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Lecture Notes on

# Nonlinear Optics

**Fredrik Jonsson**

Lectures presented at the Royal Institute of Technology
Department of Laser Physics and Quantum Optics
January 8 – March 24, 2003

**Lecture Notes on Nonlinear Optics**
Nonlinear Optics (5A5513, 5p for advanced undergraduate and doctoral students)
Course given at the Royal Institute of Technology,
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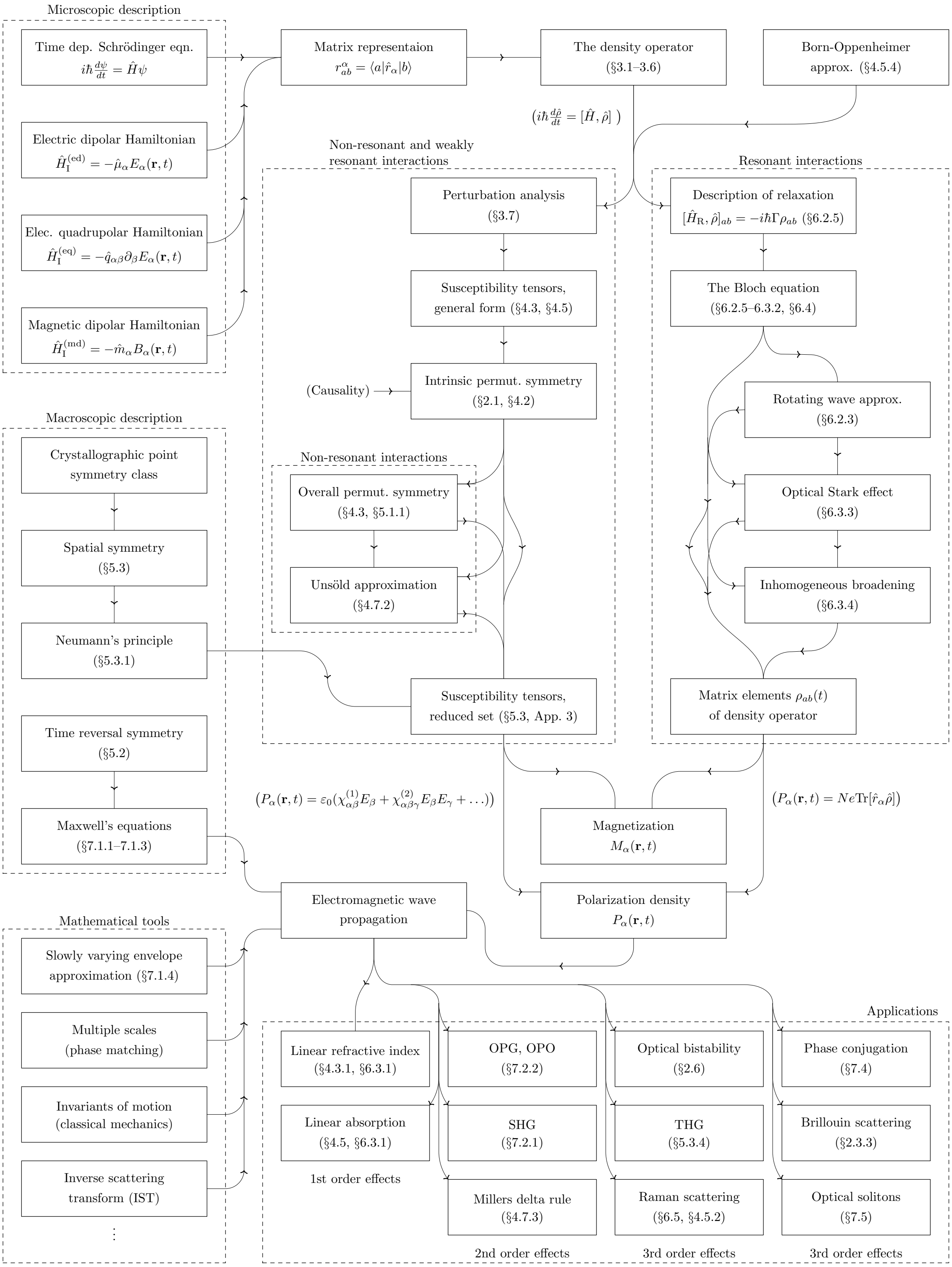

# Errors in *The Elements of Nonlinear Optics* (1991) by Paul Butcher and David Cotter

Errata written by Fredrik Jonsson

Updated as of March 16, 2003

Please send additions or corrections to `fredrik.jonsson@proximion.com`

Below follows a listing of errors found in P. N. Butcher and D. Cotters book *The Elements of Nonlinear Optics* (Cambridge University Press, Cambridge, 1991). Being a summary of the notes I have made in my personal copy of the book since June 1996, this list should by no means be considered as any kind of “official” list of errors, but rather as an attempt to collect the (rather few) misprints in the text. In the list, not only typographical misprints, but also some inconsequent notations – which do not alter the described theory – are included.

**p. 15** [*lines 14 and 16*] In order not to introduce any ambiguity of the multiple arguments of the symmetric and antisymmetric parts, the arguments $(t;\tau_1,\tau_2)$ should be explicitly written in the left-hand sides of the equations.

**p. 15** [*line 18*] “… dummy variables $\alpha\tau_1$ and $\beta\tau_2$.” should be replaced by “… dummy variables $(\alpha,\tau_1)$ and $(\beta,\tau_2)$.”, following the notation as used later in, for example, §2.3.2 and §4.3.1.

**p. 49** [*line 31*] $H_1(t)$ should be replaced by $H_{\mathrm{I}}(t)$.

**p. 54** [*lines 3, 4, and 6*] In Eq. (3.80), the upper limit of integration $t_1$ should be replaced by $\tau_1$.

**p. 54** [*lines 8 and 24*] In Eqs. (3.81) and (3.82), the upper limits of integration $t_1$ and $t_{n-1}$ should be replaced by $\tau_1$ and $\tau_{n-1}$, respectively.

**p. 60** [*line 24*] The sentence “To achieve this end we …” should be replaced by “To achieve this we …”

**p. 66** [*line 11*] In the right hand side of Eq. (4.49), one should in order not to cause confusion with the Einstein convention of summation over repeated indices explicitly state that no summation is implied, and hence the equation should be written as

$$H_0 u_i(\Theta) = \mathbb{E}_i u_i(\Theta). \quad \text{(no sum)} \tag{4.49}$$

using the common notation as used in tensor calculus.

**p. 67** [*line 6*] “… express the the unperturbed …” should be replaced by “… express the unperturbed …”

**p. 72** [*line 15*] “$(\alpha,\omega_{1)}$” should be replaced by “$(\alpha,\omega_1)$”.

**p. 72** [*last line*] “… of this type, one of which is an identity.” should be replaced by “… of this type, of which one is an identity.”

**p. 86** [*line 4*] In Eq. (4.103), “$\cdots\mathbf{f}_t(\omega_n\cdot\mathbf{e}_n\rangle$” should be replaced by “$\cdots\mathbf{f}_t(\omega_n)\cdot\mathbf{e}_n\rangle$”.

**p. 93** [*lines 12–13*] Strictly speaking, the real part of the susceptibility $\chi^{(1)}(-\omega_\sigma;\omega)$ is not proportional to the refractive index $n(\omega)$, but rather to $n^2(\omega)-1$.

**p. 97** [*lines 15 and 20*] Strictly speaking, $\Omega_{fg}$ is the transition angular frequency, and does not have the physical dimension of energy; therefore replace “$\Omega_{fg}$” in lines 15 and 20 by “$\hbar\Omega_{fg}$”.

**p. 106** [*line 9*] In the first term of Eq. (4.128), the summation should be performed over index $j$ rather than index $i$, i. e. replace $\sum_i \mathbf{p}_j$ by $\sum_j \mathbf{p}_j$.

**p. 132** [*line 8*] In Eq. (5.30), “$E_i u_i(\Theta)$” should be replaced by “$\mathbb{E}_i u_i(\Theta)$ (no sum)”.

**p. 132** [*line 25*] “$\rho_0(a)=\eta\exp(-E_a/kT)$” should be replaced by “$\rho_0(a)=\eta\exp(-\mathbb{E}_a/kT)$”.

**p. 136** [*line 19*] In Eq. (5.43), "$\chi_{ua}(1)(-\omega,\omega)$" should be replaced by "$\chi^{(1)}_{ua}(-\omega,\omega)$".

**p. 159** [*line 13*] In the left-hand side of Eq. (6.33), the Hamiltonian $H_0$ describing the system is a quantum-mechanical operator, while in the right-hand side, the matrix representation of the corresponding elements $\langle m|H_0|n\rangle = \delta_{mn}\mathbb{E}_n$ in the energy representation appears. In order to overcome this inconsistency, Eq. (6.33) should (in analogy with, for example, Eq. (6.31) for the matrix elements of the density operator) be replaced by either

$$\begin{pmatrix} \langle a|H_0|a\rangle & \langle a|H_0|b\rangle \\ \langle b|H_0|a\rangle & \langle b|H_0|b\rangle \end{pmatrix} = \begin{pmatrix} \mathbb{E}_a & 0 \\ 0 & \mathbb{E}_b \end{pmatrix},$$

or

$$\begin{pmatrix} [H_0]_{aa} & [H_0]_{ab} \\ [H_0]_{ba} & [H_0]_{bb} \end{pmatrix} = \begin{pmatrix} \mathbb{E}_a & 0 \\ 0 & \mathbb{E}_b \end{pmatrix}.$$

**p. 160** [*line 2*] In the left-hand side of Eq. (6.34), the Hamiltonian $H_{\mathrm{I}}(t)$ is a quantum-mechanical operator, while in the right-hand side, the matrix representation of its scalar elements $\langle a|H_{\mathrm{I}}(t)|b\rangle$ appears. (The same inconsistency appear in Eq. (6.33).) In order to overcome this inconsistency, Eq. (6.34) should (in analogy with, for example, Eq. (6.31) for the matrix elements of the density operator) be replaced by either

$$\begin{pmatrix} \langle a|H_{\mathrm{I}}(t)|a\rangle & \langle a|H_{\mathrm{I}}(t)|b\rangle \\ \langle b|H_{\mathrm{I}}(t)|a\rangle & \langle b|H_{\mathrm{I}}(t)|b\rangle \end{pmatrix} = \begin{pmatrix} \delta\mathbb{E}_a & -e\mathbf{r}_{ab}\cdot\mathbf{E}(t) \\ -e\mathbf{r}_{ba}\cdot\mathbf{E}(t) & \delta\mathbb{E}_b \end{pmatrix},$$

or

$$\begin{pmatrix} [H_{\mathrm{I}}(t)]_{aa} & [H_{\mathrm{I}}(t)]_{ab} \\ [H_{\mathrm{I}}(t)]_{ba} & [H_{\mathrm{I}}(t)]_{bb} \end{pmatrix} = \begin{pmatrix} \delta\mathbb{E}_a & -e\mathbf{r}_{ab}\cdot\mathbf{E}(t) \\ -e\mathbf{r}_{ba}\cdot\mathbf{E}(t) & \delta\mathbb{E}_b \end{pmatrix}.$$

**p. 164** [*line 15*] In Eq. (6.49), "$i\hbar(1-\rho_{bb})/T_b$" should be replaced by "$i\hbar(1-\rho_{aa})/T_b$".†

**p. 203** [*lines 32–33*] "Fig. 4.3" should be replaced by "Fig. 4.4(a)".

**p. 215** [*line 11*] In Eq. (7.14),

$$\ldots = \mu_0 \int_{-\infty}^{\infty} d\omega'(\omega+\omega')^2 \cdots$$

should be replaced by

$$\ldots = \frac{1}{c^2} \int_{-\infty}^{\infty} d\omega'(\omega+\omega')^2 \cdots$$

**p. 220** [*section 7.2.1*] In the example of second harmonic generation, the wave equation (7.26) is given without any explanation of which point symmetry class it applies to, and hence it is from the text virtually impossible to relate the effective nonlinear parameters to the elements of $\chi^{(2)}_{\mu\alpha\beta}(-\omega_\sigma;\omega,\omega)$.

**p. 234** [*line 31*] In Eq. (7.45), "$\ldots = iq^*\hat{E}_3^* >$" should be replaced by "$\ldots = iq^*\hat{E}_3^{**}$".

**p. 240** [*line 6*] In the first line of Eq. (7.55), there is an ambiguity of the denominator, as well as an erroneous dispersion term, and the equation

$$u = \tau\sqrt{n_2\omega/c|d^2k/d\omega^2|^2}\widehat{E}$$

should be replaced by

$$u = \tau\sqrt{n_2\omega/(c|d^2k/d\omega^2|)}\widehat{E}.$$

(The other lines of Eq. (7.55) are correct.)

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† Cf. M. D. Levenson and S. S. Kano, *Introduction to Nonlinear Laser Spectroscopy* (Academic Press, New York, 1988), p. 33, Eqs. (2.3.1)–(2.3.2).

**p. 241** [*line 30*] The fundamental bright soliton solution to the nonlinear Schrödinger equation should yield "$u(\zeta, s) = \mathrm{sech}(s)\exp(i\zeta/2)$", that is to say, *without* any minus sign in the exponential.

**p. 251** [*line 6*] In Eq. (8.5), there is parenthesis mismatch in both right- and lefthand sides;

$$f_0[(\mathbb{E}_n(\mathbf{k})] = \{\exp[\mathbb{E}_n(\mathbf{k}) - \mathbb{E}_{\mathrm{F}}]/kT + 1\}^{-1}$$

should be replaced by

$$f_0[\mathbb{E}_n(\mathbf{k})] = \{\exp[(\mathbb{E}_n(\mathbf{k}) - \mathbb{E}_{\mathrm{F}})/kT] + 1\}^{-1}.$$

**p. 252** [*line 6*] In Eq. (8.7), $n_{\mathrm{ph}}(\omega_s(\mathbf{q}))$ should be replaced by $n_{\mathrm{ph}}[\omega_s(\mathbf{q})]$, in order to follow the functional style of notation as used in, for example, Eq. (8.5).

**p. 253** [*line 20*] In Eq. (8.11), insert a "]" after $\mathbb{E}_n(\mathbf{k})$.

**p. 298** [*Table A3.2*] "... no centre of symmetry ..." should be replaced by "... no centre of inversion ...".

**p. 317** [*lines 1, 9, and 24*] In Appendix 9, there is an inconsistency in the notation for the polarisation density and the electric dipole operator, as compared to the one used in Chapters 3 and 4. While $\mathbf{P}^{\mathrm{D}}$, $\mathbf{P}^{\mathrm{Q}}$, and $\mathbf{M}^{\mathrm{D}}$ in Eq. (A9.1) (and in line 9 on the same page) denote the all-classical electric dipolar, electric quadrupolar and magnetic dipolar polarization densities of the medium, they in Eqs. (A9.6) and (A9.7) clearly denote quantum-mechanical operators. In order to overcome this inconsistency in notation, which in addition gives a wrong answer if properly inserted into the perturbation calculus etc., one should chose either of the conventions. By choosing $\mathbf{P}^{\mathrm{D}}$, $\mathbf{P}^{\mathrm{Q}}$, and $\mathbf{M}^{\mathrm{D}}$ to denote the corresponding quantum-mechanical operators, which seem to be the easiest way of correcting this inconsistency, the following corrections to the text should be made:

[*line 1*

$$\mathbf{P} = \mathbf{P}^{\mathrm{D}} + \mathbf{P}^{\mathrm{Q}}, \qquad \mathbf{M} = \mathbf{M}^{\mathrm{D}}, \tag{A9.1}$$

should be replaced by

$$\mathbf{P} = \langle\mathbf{P}^{\mathrm{D}}\rangle + \langle\mathbf{P}^{\mathrm{Q}}\rangle, \qquad \mathbf{M} = \langle\mathbf{M}^{\mathrm{D}}\rangle, \tag{A9.1}$$

[*line 9* Remove "$\mathbf{P}^{\mathrm{D}}$" or replace with "$\langle\mathbf{P}^{\mathrm{D}}\rangle$".

[*line 24* Somewhere in Appendix 9, there should be a clarifying statement that the nabla operator appearing in Eq. (A9.7) only is operating on the all-classical, macroscopic electric field of the light, and hence should be regarded as a classical vector when evaluating the quantum-mechanical trace that is involved in the expectation value of, for example, the corrected form of Eq. (A9.1).

**p. 318** [*line 12*] "$\mathbf{M} << \mathbf{H}$" should be replaced by "$|\mathbf{M}| \ll |\mathbf{H}|$".

**p. 318** [*line 23*] "$\mathbf{e}_j \cdot e\mathbf{r} + i\mathbf{k}\cdot\mathbf{q}\cdot\mathbf{e}_j + \mathbf{m}\cdot(\mathbf{k}\times\mathbf{e}_j)/\omega$" should be replaced by the same expression, though with each term divided by $e$.

**p. 333** [*line 5*] In reference Manley, J. M. and Rowe, H. E. (1956), the page numbers should yield 904 – 14.

**p. 334** [*line 44*] "Terhune, R. W. and Weinburger, D. A. ..." should be replaced by "Terhune, R. W. and Weinberger, D. A. ...".

## Notes on the "Butcher and Cotter convention" in nonlinear optics

### Convention for description of nonlinear optical polarization

As a "recipe" in theoretical nonlinear optics, Butcher and Cotter provide a very useful convention which is well worth to hold on to. For a superposition of monochromatic waves, and by invoking the general property of the intrinsic permutation symmetry, the monochromatic form of the $n$th order polarization density can be written as

$$(P^{(n)}_{\omega_\sigma})_\mu = \varepsilon_0 \sum_{\alpha_1} \cdots \sum_{\alpha_n} \sum_{\omega} K(-\omega_\sigma; \omega_1, \ldots, \omega_n) \chi^{(n)}_{\mu\alpha_1\cdots\alpha_n}(-\omega_\sigma; \omega_1, \ldots, \omega_n)(E_{\omega_1})_{\alpha_1} \cdots (E_{\omega_n})_{\alpha_n}. \tag{1}$$

The first summations in Eq. (1), over $\alpha_1, \ldots, \alpha_n$, is simply an explicit way of stating that the Einstein convention of summation over repeated indices holds. The summation sign $\sum_\omega$, however, serves as a reminder that the expression that follows is to be summed over *all distinct sets of* $\omega_1, \ldots, \omega_n$. Because of the intrinsic permutation symmetry, the frequency arguments appearing in Eq. (1) may be written in arbitrary order.

By "all distinct sets of $\omega_1, \ldots, \omega_n$", we here mean that the summation is to be performed, as for example in the case of optical Kerr-effect, over the single set of nonlinear susceptibilities that contribute to a certain angular frequency as $(-\omega; \omega, \omega, -\omega)$ *or* $(-\omega; \omega, -\omega, \omega)$ *or* $(-\omega; -\omega, \omega, \omega)$. In this example, each of the combinations are considered as *distinct*, and it is left as an arbitary choice which one of these sets that are most convenient to use (this is simply a matter of choosing notation, and does not by any means change the description of the interaction).

In Eq. (1), the degeneracy factor $K$ is formally described as

$$K(-\omega_\sigma; \omega_1, \ldots, \omega_n) = 2^{l+m-n} p$$

where

$$\begin{aligned}
p &= \text{the number of \textit{distinct} permutations of } \omega_1, \omega_2, \ldots, \omega_1, \\
n &= \text{the order of the nonlinearity}, \\
m &= \text{the number of angular frequencies } \omega_k \text{ that are zero, and} \\
l &= \begin{cases} 1, & \text{if } \omega_\sigma \neq 0, \\ 0, & \text{otherwise.} \end{cases}
\end{aligned}$$

In other words, $m$ is the number of DC electric fields present, and $l = 0$ if the nonlinearity we are analyzing gives a static, DC, polarization density, such as in the previously (in the spring model) described case of optical rectification in the presence of second harmonic fields (SHG).

A list of frequently encountered nonlinear phenomena in nonlinear optics, including the degeneracy factors as conforming to the above convention, is given in Butcher and Cotters book, Table 2.1, on page 26.

### Note on the complex representation of the optical field

Since the observable electric field of the light, in Butcher and Cotters notation taken as

$$\mathbf{E}(\mathbf{r}, t) = \frac{1}{2} \sum_{\omega_k \geq 0} [\mathbf{E}_{\omega_k} \exp(-i\omega_k t) + \mathbf{E}^*_{\omega_k} \exp(i\omega_k t)],$$

is a real-valued quantity, it follows that negative frequencies in the complex notation should be interpreted as the complex conjugate of the respective field component, or

$$\mathbf{E}_{-\omega_k} = \mathbf{E}^*_{\omega_k}.$$

**Example: Optical Kerr-effect**

Assume a monochromatic optical wave (containing forward and/or backward propagating components) polarized in the $xy$-plane,

$$\mathbf{E}(z,t) = \mathrm{Re}[\mathbf{E}_\omega(z)\exp(-i\omega t)] \in \mathbb{R}^3,$$

with all spatial variation of the field contained in

$$\mathbf{E}_\omega(z) = \mathbf{e}_x E^x_\omega(z) + \mathbf{e}_y E^y_\omega(z) \in \mathbb{C}^3.$$

Optical Kerr-effect is in isotropic media described by the third order susceptibility

$$\chi^{(3)}_{\mu\alpha\beta\gamma}(-\omega;\omega,\omega,-\omega),$$

with nonzero components of interest for the $xy$-polarized beam given in Appendix 3.3 of Butcher and Cotters book as

$$\chi^{(3)}_{xxxx} = \chi^{(3)}_{yyyy}, \quad \chi^{(3)}_{xxyy} = \chi^{(3)}_{yyxx} = \left\{ \begin{array}{c} \text{intr. perm. symm.} \\ (\alpha,\omega) \rightleftharpoons (\beta,\omega) \end{array} \right\} = \chi^{(3)}_{xyxy} = \chi^{(3)}_{yxyx}, \quad \chi^{(3)}_{xyyx} = \chi^{(3)}_{yxxy},$$

with

$$\chi^{(3)}_{xxxx} = \chi^{(3)}_{xxyy} + \chi^{(3)}_{xyxy} + \chi^{(3)}_{xyyx}.$$

The degeneracy factor $K(-\omega;\omega,\omega,-\omega)$ is calculated as

$$K(-\omega;\omega,\omega,-\omega) = 2^{l+m-n}p = 2^{1+0-3}3 = 3/4.$$

From this set of nonzero susceptibilities, and using the calculated value of the degeneracy factor in the convention of Butcher and Cotter, we hence have the third order electric polarization density at $\omega_\sigma = \omega$ given as $\mathbf{P}^{(n)}(\mathbf{r},t) = \mathrm{Re}[\mathbf{P}^{(n)}_\omega \exp(-i\omega t)]$, with

$$\begin{aligned}
\mathbf{P}^{(3)}_\omega &= \sum_\mu \mathbf{e}_\mu (P^{(3)}_\omega)_\mu \\
&= \{\text{Using the convention of Butcher and Cotter}\} \\
&= \sum_\mu \mathbf{e}_\mu \left[ \varepsilon_0 \frac{3}{4} \sum_\alpha \sum_\beta \sum_\gamma \chi^{(3)}_{\mu\alpha\beta\gamma}(-\omega;\omega,\omega,-\omega)(E_\omega)_\alpha (E_\omega)_\beta (E_{-\omega})_\gamma \right] \\
&= \{\text{Evaluate the sums over } (x,y,z) \text{ for field polarized in the } xy \text{ plane}\} \\
&= \varepsilon_0 \frac{3}{4} \{ \mathbf{e}_x [\chi^{(3)}_{xxxx} E^x_\omega E^x_\omega E^x_{-\omega} + \chi^{(3)}_{xyyx} E^y_\omega E^y_\omega E^x_{-\omega} + \chi^{(3)}_{xyxy} E^y_\omega E^x_\omega E^y_{-\omega} + \chi^{(3)}_{xxyy} E^x_\omega E^y_\omega E^y_{-\omega}] \\
&\qquad + \mathbf{e}_y [\chi^{(3)}_{yyyy} E^y_\omega E^y_\omega E^y_{-\omega} + \chi^{(3)}_{yxxy} E^x_\omega E^x_\omega E^y_{-\omega} + \chi^{(3)}_{yxyx} E^x_\omega E^y_\omega E^x_{-\omega} + \chi^{(3)}_{yyxx} E^y_\omega E^x_\omega E^x_{-\omega}]\} \\
&= \{\text{Make use of } \mathbf{E}_{-\omega} = \mathbf{E}^*_\omega \text{ and relations } \chi^{(3)}_{xxyy} = \chi^{(3)}_{yyxx}, \text{ etc.}\} \\
&= \varepsilon_0 \frac{3}{4} \{ \mathbf{e}_x [\chi^{(3)}_{xxxx} E^x_\omega |E^x_\omega|^2 + \chi^{(3)}_{xyyx} E^{y\,2}_\omega E^{x*}_\omega + \chi^{(3)}_{xyxy} |E^y_\omega|^2 E^x_\omega + \chi^{(3)}_{xxyy} E^x_\omega |E^y_\omega|^2] \\
&\qquad + \mathbf{e}_y [\chi^{(3)}_{xxxx} E^y_\omega |E^y_\omega|^2 + \chi^{(3)}_{xyyx} E^{x\,2}_\omega E^{y*}_\omega + \chi^{(3)}_{xyxy} |E^x_\omega|^2 E^y_\omega + \chi^{(3)}_{xxyy} E^y_\omega |E^x_\omega|^2]\} \\
&= \{\text{Make use of intrinsic permutation symmetry}\} \\
&= \varepsilon_0 \frac{3}{4} \{ \mathbf{e}_x [(\chi^{(3)}_{xxxx} |E^x_\omega|^2 + 2\chi^{(3)}_{xxyy} |E^y_\omega|^2) E^x_\omega + (\chi^{(3)}_{xxxx} - 2\chi^{(3)}_{xxyy}) E^{y\,2}_\omega E^{x*}_\omega \\
&\qquad \mathbf{e}_y [(\chi^{(3)}_{xxxx} |E^y_\omega|^2 + 2\chi^{(3)}_{xxyy} |E^x_\omega|^2) E^y_\omega + (\chi^{(3)}_{xxxx} - 2\chi^{(3)}_{xxyy}) E^{x\,2}_\omega E^{y*}_\omega.
\end{aligned}$$

For the optical field being linearly polarized, say in the $x$-direction, the expression for the polarization density is significantly simplified, to yield

$$\mathbf{P}^{(3)}_\omega = \varepsilon_0 (3/4) \mathbf{e}_x \chi^{(3)}_{xxxx} |E^x_\omega|^2 E^x_\omega,$$

i. e. taking a form that can be interpreted as an intensity-dependent ($\sim |E^x_\omega|^2$) contribution to the refractive index (cf. Butcher and Cotter §6.3.1).

## Recommended reference literature in nonlinear optics

[1] P. N. Butcher and D. Cotter, *The Elements of Nonlinear Optics* (Cambridge University Press, Cambridge, 1991). Uses SI units throughout the text.

[2] Y. R. Shen, *The Principles of Nonlinear Optics* (Wiley, New York, 1984). A standard text which covers most of the phenomena and applications in nonlinear optics. More directed towards the already active scientist in the field, rather than being a tutorial text. Uses Gaussian units throughout the text.

[3] D. C. Hanna, M. A. Yuratich, and D. Cotter, *Nonlinear Optics of Free Atoms and Molecules* (Springer-Verlag, Berlin, 1979). Contains a somewhat more detailed description of electric quadrupolar contributions to nonlinear susceptibilities than otherwise usually found in textbooks in nonlinear optics. Uses SI units throughout the text.

[4] R. W. Boyd, *Nonlinear Optics* (Academic Press, Boston, 1992). Starting more from an atomic physics point of view, this book contains aspects on spectroscopical applications of nonlinear optics. Uses Gaussian units throughout the text.

[5] P. Meystre and M. Sargent III, *Elements of quantum optics* (Springer-Verlag, Berlin, 1998), 3rd ed. The title of this book might be somewhat misleading, since the book contains quite a lot of interesting discussions on nonlinear optical phenomena from a classical description. This textbook provides a strict quantum-mechanical approach to linear as well as nonlinear optical interactions. The authors have chosen to include somewhat more classical tools of analysis as well, such as the Ikeda instability analysis of optical Kerr-media, and the Lorenz model for the dynamics of the homogeneously broadened singlemode unidirectional ring laser, the latter leading to solutions possessing deterministic chaos. Uses SI units throughout the text.

[6] A. C. Newell and J. V. Moloney, *Nonlinear optics* (Addison-Wesley, New York, 1992). In this book the principles of nonlinear optics are described from a more mathematical point of view. Wave propagation in nonlinear optical media is covered in more detail than in other books, and the theory of solitons is described. Contains a nice introduction to the deterministic chaos which is an intrinsic property of some numerical solutions to the wave propagation problem in certain nonlinear optical media. The book covers the Bloch equations, describing the interaction between light and matter, but does surprisingly not cover the origin of the nonlinear susceptibility tensors, even though they play a central role in the first chapters of the book. In the last chapter, a review of mathematical and computional tools that frequently are applied in nonlinear optics is presented. Uses SI units throughout the text.

[7] N. Bloembergen *Nonlinear Optics* (World Scientific, London, 1996), 4th ed. Almost to be considered as *the* historical text on nonlinear optics. The first edition of this book appeared as early as 1965, just a few years after the first observations of nonlinear optical phenomena by Franken et al. (1961). Some of the early, pioneering papers on nonlinear optics (e. g. J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, Phys. Rev. **127**, 1918 (1962); P. A. Franken, A. E. Hill, C. W. Peters, G. Weinreich, Phys. Rev. Lett. **7**, 118 (1961)) are in this book extended and presented in more detail. In 1981, Nicolaas Bloembergen and Arthur Schawlow received the Nobel prize for their contribution to the development of laser spectroscopy, in particular using nuclear magnetic resonance (NMR). Gaussian units are used throughout the text.

# Lecture I

**Lecture Notes on Nonlinear Optics**
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# LECTURE 1

Nonlinear optics is the discipline in physics in which the electric polarization density of the medium is studied as a nonlinear function of the electromagnetic field of the light. Being a wide field of research in electromagnetic wave propagation, nonlinear interaction between light and matter leads to a wide spectrum of phenomena, such as optical frequency conversion, optical solitons, phase conjugation, and Raman scattering. In addition, many of the analytical tools applied in nonlinear optics are of general character, such as the perturbative techniques and symmetry considerations, and can equally well be applied in other disciplines in nonlinear dynamics.

## The contents of this course

This course is intended as an introduction to the wide field of phenomena encountered in nonlinear optics. The course covers:

- The theoretical foundation of nonlinear interaction between light and matter.
- Perturbation analysis of nonlinear interaction between light and matter.
- The Bloch equation and its interpretation.
- Basics of soliton theory and the inverse scattering transform.

It should be emphasized that the course does not cover state-of-the-art material constants of nonlinear optical materials, etc. but rather focus on the theoretical foundations and ideas of nonlinear optical interactions between light and matter.

A central analytical technique in this course is the perurbation analysis, with its foundation in the analytical mechanics. This technique will in the course mainly be applied to the quantum-mechanical description of interaction between light and matter, but is central in a wide field of cross-disciplinary physics as well. In order to give an introduction to the analytical theory of nonlinear systems, we will therefore start with the analysis of the nonlinear equations of motion for the mechanical pendulum.

## Examples of applications of nonlinear optics

Some important applications in nonlinear optics:

- Optical parametric amplification (OPA) and oscillation (OPO), $\hbar\omega_{\rm p} \to \hbar\omega_{\rm s} + \hbar\omega_{\rm i}$.
- Second harmonic generation (SHG), $\hbar\omega + \hbar\omega \to \hbar(2\omega)$.
- Third harmonic generation (THG), $\hbar\omega + \hbar\omega + \hbar\omega \to \hbar(3\omega)$.
- Pockels effect, or the linear electro-optical effect (applications for optical switching).
- Optical bistability (optical logics).
- Optical solitons (ultra long-haul communication).

## A brief history of nonlinear optics

Some important advances in nonlinear optics:

- Townes et al. (1960), invention of the laser.[1]
- Franken et al. (1961), First observation ever of nonlinear optical effects, second harmonic generation (SHG).[2]
- Terhune et al. (1962), First observation of third harmonic generation (THG).[3]
- E. J. Woodbury and W. K. Ng (1962), first demonstration of stimulated Raman scattering.[4]
- Armstrong et al. (1962), formulation of general permutation symmetry relations in nonlinear optics.[5]
- A. Hasegawa and F. Tappert (1973), first theoretical prediction of soliton generation in optical fibers.[6]
- H. M. Gibbs et al. (1976), first demonstration and explaination of optical bistability.[7]
- L. F. Mollenauer et al. (1980), first confirmation of soliton generation in optical fibers.[8]

Recently, many advances in nonlinear optics has been made, with a lot of efforts with fields of, for example, Bose-Einstein condensation and laser cooling; these fields are, however, a bit out of focus from the subjects of this course, which can be said to be an introduction to the 1960s and 1970s advances in nonlinear optics. It should also be emphasized that many of the effects observed in nonlinear optics, such as the Raman scattering, were observed much earlier in the microwave range.

## Outline for calculations of polarization densities

### *Metals and plasmas*

From an all-classical point-of-view, the calculation of the electric polarization density of metals and plasmas, containing a free electron gas, can be performed using the model of free charges acting under the Lorenz force of an electromagnetic field,

$$m_{\rm e}\frac{d^2\mathbf{r}_{\rm e}}{dt^2} = -e\mathbf{E}(t) - e\frac{d\mathbf{r}_{\rm e}}{dt}\times\mathbf{B}(t),$$

where **E** and **B** are all-classical electric and magnetic fields of the electromagnetic field of the light. In forming the equation for the motion of the electron, the origin was chosen to coincide with the center of the nucleus.

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[1] Charles H. Townes was in 1964 awarded with the Nobel Prize for the invention of the ammonia laser.

[2] Franken et al. detected ulvtraviolet light ($\lambda = 347.1$ nm) at twice the frequency of a ruby laser beam ($\lambda = 694.2$ nm) when this beam traversed a quartz crystal; P. A. Franken, A. E. Hill, C. W. Peters, G. Weinreich, Phys. Rev. Lett. **7**, 118 (1961). Second harmonic generation is also the first nonlinear effect ever observed where a coherent input generates a coherent output.

[3] In their experiment, Terhune et al. detected only about a thousand THG photons per pulse, at $\lambda = 231.3$ nm, corresponding to a conversion of one photon out of about $10^{15}$ photons at the fundamental wavelength at $\lambda = 693.9$ nm; R. W. Terhune, P. D. Maker, and C. M. Savage, Phys. Rev. Lett. **8**, 404 (1962).

[4] E. J. Woodbury and W. K. Ng, Proc. IRE **50**, 2347 (1962).

[5] The general permutation symmetry relations of higher-order susceptibilities were published by J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, Phys. Rev. **127**, 1918 (1962).

[6] A. Hasegawa and F. Tappert, "Transmission of stationary nonliner optica pulses in dispersive optical fibers: I, Anomalous dispersion; II Normal dispersion", Appl. Phys. Lett. **23**, 142–144 and 171–172 (August 1 and 15, 1973).

[7] H. M. Gibbs, S. M. McCall, and T. N. C. Venkatesan, Phys. Rev. Lett. **36**, 1135 (1976).

[8] L. F. Mollenauer, R. H. Stolen, and J. P. Gordon, "Experimental observation of picosecond pulse narrowing and solitons in optical fibers", Phys. Rev. Lett. **45**, 1095–1098 (September 29, 1980); the first reported observation of solitons was though made in 1834 by John Scott Russell, a Scottish scientist and later famous Victorian engineer and shipbuilder, while studying water waves in the Glasgow-Edinburgh channel.

*Dielectrics*

A very useful model used by Drude and Lorentz[9] to calculate the linear electric polarization of the medium describes the electrons as harmonically bound particles.

For dielectrics in the nonlinear optical regime, as being the focus of our attention in this course, the calculation of the electric polarization density is instead performed using a nonlinear spring model of the bound charges, here quoted for one-dimensional motion as

$$m_{\mathrm{e}}\frac{d^2x_{\mathrm{e}}}{dt^2}+\Gamma_{\mathrm{e}}\frac{dx_{\mathrm{e}}}{dt}+\alpha^{(1)}x_{\mathrm{e}}+\alpha^{(2)}x_{\mathrm{e}}^2+\alpha^{(3)}x_{\mathrm{e}}^3+\ldots=-eE_x(t).$$

As in the previous case of metals and plasmas, in forming the equation for the motion of the electron, the origin was also here chosen to coincide with the center of the nucleus.

This classical mechanical model will later in this lecture be applied to the derivation of the second-order nonlinear polarzation density of the medium.

### Introduction to nonlinear dynamical systems

In this section we will, as a preamble to later analysis of quantum-mechanical systems, apply perturbation analysis to a simple mechanical system. Among the simplest nonlinear dynamical systems is the pendulum, for which the total mechanical energy of the system, considering the point of suspension as defining the level of zero potential energy, is given as the sum of the kinetic and potential energy as

$$E=T+V=\frac{1}{2m}|\mathbf{p}|^2-mgl(\cos\vartheta-1),\tag{1}$$

where $m$ is the mass, $g$ the gravitation constant, $l$ the length, and $\vartheta$ the angle of deflection of the pendulum, and where $\mathbf{p}$ is the momentum of the point mass.

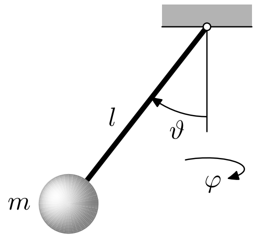

Figure 1. The mechanical pendulum.

From the total mechanical energy (1) of the system, the equations of motion for the point mass is hence given by Lagranges equations,[10]

$$\frac{d}{dt}\left(\frac{\partial L}{\partial p_j}\right)-\frac{\partial L}{\partial q_j}=0,\qquad j=x,y,z,\tag{2}$$

where $q_j$ are the generalized coordinates, $p_j=\dot{q}_j$ are the components of the generalized momentum, and $L=T-V$ is the Lagrangian of the mechanical system. In spherical coordinates $(\rho,\varphi,\vartheta)$, the momentum for the mass is given as its mass times the velocity,

$$\mathbf{p}=ml(\dot{\vartheta}\mathbf{e}_\vartheta+\dot{\varphi}\sin\vartheta\mathbf{e}_\varphi)$$

and the Lagrangian for the pendulum is hence given as

$$\begin{aligned}L&=\frac{1}{2m}(p_\varphi^2+p_\varphi^2)+mgl(\cos\vartheta-1)\\&=\frac{ml^2}{2}(\dot{\vartheta}^2+\dot{\varphi}^2\sin^2\vartheta)+mgl(\cos\vartheta-1).\end{aligned}$$

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[9] R. Becker, *Elektronen Theorie*, (Teubner, Leipzig, 1933).

[10] Herbert Goldstein, *Classical Mechanics*, 2nd ed. (Addison-Wesley, Massachusetts, 1980).

As the Lagrangian for the pendulum is inserted into Eq. (2), the resulting equations of motion are for $q_j = \varphi$ and $\vartheta$ obtained as

$$\frac{d^2\varphi}{dt^2} + \left(\frac{d\varphi}{dt}\right)^2 \sin\varphi \cos\varphi = 0, \tag{3a}$$

and

$$\frac{d^2\vartheta}{dt^2} + (g/l)\sin\vartheta = 0, \tag{3b}$$

respectively, where we may notice that the motion in $\mathbf{e}_\varphi$ and $\mathbf{e}_\vartheta$ directions are decoupled. We may also notice that the equation of motion for $\varphi$ is independent of any of the physical parameters involved in te Lagrangian, and the evolution of $\varphi(t)$ in time is entirely determined by the initial conditions at some time $t = t_0$.

In the following discussion, the focus will be on the properties of the motion of $\vartheta(t)$. The equation of motion for the $\vartheta$ coordinate is here described by the so-called Sine-Gordon equation.[11] This nonlinear differential equation is hard† to solve analytically, but if the nonlinear term is expanded as a Taylor series around $\vartheta = 0$,

$$\frac{d^2\vartheta}{dt^2} + (g/l)(\vartheta - \frac{\vartheta^3}{3!} + \frac{\vartheta^5}{5!} + \ldots) = 0,$$

Before proceeding further with the properties of the solutions to the approximative Sine-Gordon, including various orders of nonlinearities, the general properties will now be illustrated. In order to illustrate the behaviour of the Sine-Gordon equation, we may normalize it by using the normalized time $\tau = (g/l)^{1/2}t$, giving the Sine-Gordon equation in the normalized form

$$\frac{d^2\vartheta}{d\tau^2} + \sin\vartheta = 0. \tag{4}$$

The numerical solutions to the normalized Sine-Gordon equation are in Fig. 2 shown for initial conditions (a) $y(0) = 0.1$, (b) $y(0) = 2.1$, and (c) $y(0) = 3.1$, all cases with $y'(0) = 0$.

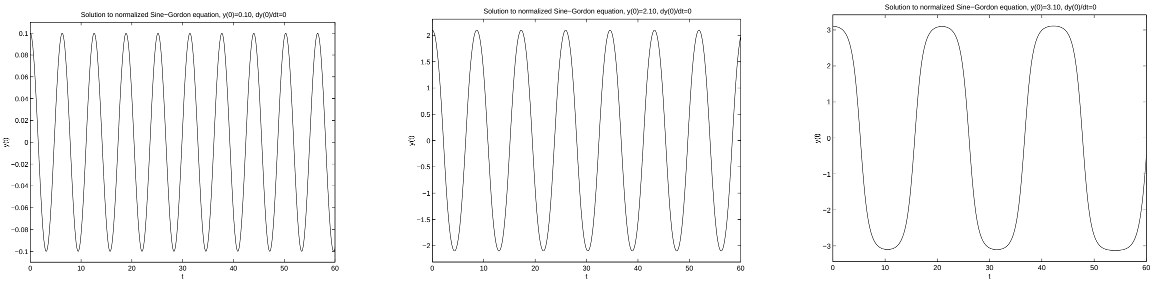

Figure 2. Numerical solutions to the normalized Sine-Gordon equation.

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[11] The term “Sine-Gordon equation” has its origin as an allegory over the similarity between the time-dependent (Sine-Gordon) equation

$$\frac{\partial^2\varphi}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2\varphi}{\partial t^2} = \mu_0^2 \sin\varphi,$$

appearing in, for example, relativistic field theories, as compared to the time-dependent Klein-Gordon equation, which takes the form

$$\frac{\partial^2\varphi}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2\varphi}{\partial t^2} = \mu_0^2 \varphi.$$

The Sine-Gordon equation is sometimes also called “pendulum equation” in the terminology of classical mechanics.

† Impossible?

First of all, we may consider the linear case, for which the approximation $\sin\vartheta \approx \vartheta$ holds. For this case, the Sine-Gordon equation (4) hence reduces to the one-dimensional linear wave-equation, with solutions $\vartheta = A\sin((g/l)^{1/2}(t-t_0))$. As seen in the frequency domain, this solution gives a delta peak at $\omega = (g/l)^{1/2}$ in the power spectrum $|\tilde{\vartheta}(\omega)|^2$, with no other frequency components present. However, if we include the nonlinearities, the previous sine-wave solution will tend to flatten at the peaks, as well as increase in period, and this changes the power spectrum to be broadened as well as flattened out. In other words, the solution to the Sine-Gordon give rise to a wide spectrum of frequencies, as compared to the delta peaks of the solutions to the linearized, approximative Sine-Gordon equation.

From the numerical solutions, we may draw the conclusion that whenever higher order nonlinear restoring forces come into play, even such a simple mechanical system as the pendulum will carry frequency components at a set of frequencies differing from the single frequency given by the linearized model of motion.

More generally, hiding the fact that for this particular case the restoring force is a simple sine function, the equation of motion for the pendulum can be written as

$$\frac{d^2\vartheta}{dt^2} + a^{(0)} + a^{(1)}\vartheta + a^{(2)}\vartheta^2 + a^{(3)}\vartheta^3 + \ldots = 0. \tag{5}$$

This equation of motion may be compared with the nonlinear wave equation for the electromagnetic field of a travelling optical wave of angular frequency $\omega$, of the form

$$\frac{\partial^2 E}{\partial z^2} + \frac{\omega^2}{c^2}E + \frac{\omega^2}{c^2}(\chi^{(1)}E + \chi^{(2)}E^2 + \chi^{(3)}E^3 + \ldots) = 0,$$

which clearly shows the similarity between the nonlinear wave propagation and the motion of the nonlinear pendulum.

Having solved the particular problem of the nonlinear pendulum, we may ask ourselves if the equations of motion may be altered in some way in order to give insight in other areas of nonlinear physics as well. For example, the series (5) that define the feedback that tend to restore the mechanical pendulum to its rest position clearly defines equations of motion that conserve the total energy of the mechanical system. This, however, in generally not true for an arbitrary series of terms of various power for the restoring force. As we will later on see, in nonlinear optics we generally have a complex, though in many cases most predictable, transfer of energy between modes of different frequencies and directions of propagation.

### The anharmonic oscillator

Among the simplest models of interaction between light and matter is the all-classical one-electron oscillator, consisting of a negatively charged particle (electron) with mass $m_{\rm e}$, mutually interacting with a positively charged particle (proton) with mass $m_{\rm p}$, through attractive Coulomb forces.

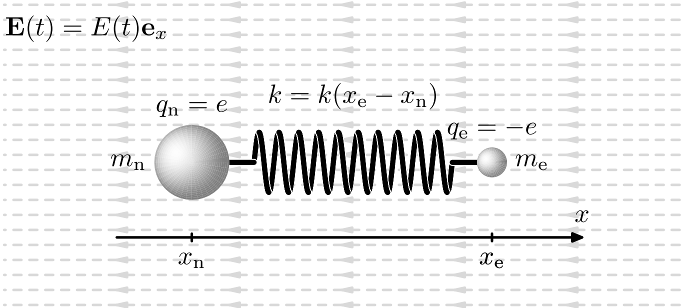

Figure 5. Setup of the one-dimensional undamped spring model.

In the one-electron oscillator model, several levels of approximations may be applied to the problem, with increasing algebraic complexity. At the first level of approximation, the proton is assumed to be fixed in space, with the electron free to oscillate around the proton. Quite generally, at least within the scope of linear optics, the restoring spring force which confines the electron can be assumed to be linear with the displacement distance of the electron from the central position. Providing the very basic models of the concept of refractive index and optical dispersion, this model has been applied by numerous authors, such as Feynman [R. P. Feynman, *Lectures on Physics* (Addison-Wesley, Massachusetts, 1963)], and Born and Wolf [M. Born and E. Wolf, *Principles of Optics* (Cambridge University Press, Cambridge, 1980)].

Moving on to the next level of approximation, the bound proton-electron pair may be considered as constituting a two-body central force problem of classical mechanics, in which one may assume a fixed center of mass of the system, around which the proton as well as the electron are free to oscillate. In this level of approximation, by introducing the concept of reduced mass for the two moving particles, the equations of motion for the two particles can be reduced to one equation of motion, for the evolution of the electric dipole moment of the system.

The third level of approximation which may be identified is when the center of mass is allowed to oscillate as well, in which case an equation of motion for the center of mass appears in addition to the one for the evolution of the electric dipole moment.

In each of the models, nonlinearities of the restoring central force field may be introduced as to include nonlinear interactions as well. It should be emphasized that the spring model, as now will be introduced, gives an identical form of the set of nonzero elements of the susceptibility tensors, as compared with those obtained using a quantum mechanical analysis.

Throughout this analysis, the wavelength of the electromagnetic field will be assumed to be sufficiently large in order to neglect any spatial variations of the fields over the spatial extent of the oscillator system. In this model, the central force field is modelled by a mechanical spring force with spring constant $k_{\rm e}$, as shown schematically in Fig. 5, and the all-classical Newton's equations of motion for the electron and nucleus are

$$\begin{aligned} m_{\rm e}\frac{\partial^2 x_{\rm e}}{\partial t^2} &= \underbrace{-eE(t)}_{\text{optical}} - \underbrace{k_0(x_{\rm e}-x_{\rm n}) + k_1(x_{\rm e}-x_{\rm n})^2}_{\text{spring}},\\ m_{\rm n}\frac{\partial^2 x_{\rm n}}{\partial t^2} &= \underbrace{+eE(t)}_{\text{optical}} + \underbrace{k_0(x_{\rm e}-x_{\rm n}) - k_1(x_{\rm e}-x_{\rm n})^2}_{\text{spring}}, \end{aligned}$$

corresponding to a system of two particles connected by a spring with spring "constant"

$$k = -\frac{\partial F_{\rm e}^{(\rm spring)}}{\partial(x_{\rm e}-x_{\rm n})} = \frac{\partial F_{\rm n}^{(\rm spring)}}{\partial(x_{\rm e}-x_{\rm n})} = k_0 - 2k_1(x_{\rm e}-x_{\rm n}).$$

By introducing the reduced mass[12] $m_{\rm r} = m_{\rm e}m_{\rm n}/(m_{\rm e}+m_{\rm n})$ of the system, the equation of motion for the electric dipole moment $p = -e(x_{\rm e}-x_{\rm n})$ is then obtained as

$$\frac{\partial^2 p}{\partial t^2} + \frac{k_0}{m_{\rm r}}p + \frac{k_1}{em_{\rm r}}p^2 = \frac{e^2}{m_{\rm r}}E(t). \tag{6}$$

This inhomogeneous nonlinear ordinary differential equation for the electric dipole moment is the primary interest in the discussion that now is to follow.

The electric dipole moment of the anharmonic oscillator is now expressed in terms of a perturbation series as

$$p(t) = p^{(0)}(t) + \underbrace{p^{(1)}(t)}_{\propto E(t)} + \underbrace{p^{(2)}(t)}_{\propto E^2(t)} + \underbrace{p^{(3)}(t)}_{\propto E^3(t)} + \ldots,$$

where each term in the series is proportional to the applied electrical field strength to the power as indicated in the superscript of repective term, and formulate the system of $n+1$ equations for $p^{(k)}$, $k = 0, 1, 2, \ldots, n$, that define the time evolution of the electric dipole. By inserting the

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[12] Herbert Goldstein, *Classical Mechanics*, 2nd ed. (Addison-Wesley, Massachusetts, 1980).

perturbation series into Eq. (6), we hence have the equation

$$\begin{aligned}\frac{\partial^2 p^{(0)}}{\partial t^2} &+ \frac{\partial^2 p^{(1)}}{\partial t^2} + \frac{\partial^2 p^{(2)}}{\partial t^2} + \frac{\partial^2 p^{(3)}}{\partial t^2} + \ldots\\ &+ \frac{k_0}{m_\mathrm{r}} p^{(0)} + \frac{k_0}{m_\mathrm{r}} p^{(1)} + \frac{k_0}{m_\mathrm{r}} p^{(2)} + \frac{k_0}{m_\mathrm{r}} p^{(3)} + \ldots\\ &+ \frac{k_1}{e m_\mathrm{r}} (p^{(0)} + p^{(1)} + p^{(2)} + \ldots)(p^{(0)} + p^{(1)} + p^{(2)} + \ldots) = \frac{e^2}{m_\mathrm{r}} E(t).\end{aligned}$$

Since this equation is to hold for an arbitrary electric field $E(t)$, that is to say, at least within the limits of the validity of the perturbation analysis, each set of terms with equal power dependence of the electric field must individually satisfy the relation. By sorting out the various powers and identifying terms in the left and right hand sides of the equation, we arrive at the system of equations

$$\begin{aligned}&\frac{\partial^2 p^{(0)}}{\partial t^2} + \frac{k_0}{m_\mathrm{r}} p^{(0)} + \frac{k_1}{e m_\mathrm{r}} p^{(0)2} = 0,\\ &\frac{\partial^2 p^{(1)}}{\partial t^2} + \frac{k_0}{m_\mathrm{r}} p^{(1)} + \frac{k_1}{e m_\mathrm{r}} 2p^{(0)} p^{(1)} = \frac{e^2}{m_\mathrm{r}} E(t),\\ &\frac{\partial^2 p^{(2)}}{\partial t^2} + \frac{k_0}{m_\mathrm{r}} p^{(2)} + \frac{k_1}{e m_\mathrm{r}} (2p^{(0)} p^{(2)} + p^{(1)2}) = 0,\\ &\frac{\partial^2 p^{(3)}}{\partial t^2} + \frac{k_0}{m_\mathrm{r}} p^{(3)} + \frac{k_1}{e m_\mathrm{r}} (2p^{(0)} p^{(3)} + 2p^{(1)} p^{(2)}) = 0,\end{aligned}$$

where we kept terms with powers of the electric field up to and including order three. At a first glance, this system seem to suggest that only the first order of the perturbation series depends on the applied electric field of the light; however, taking a closer look at the system, one can easily verify that all orders of the dipole moment is coupled directly to the lower order terms. The system of equations for $p^{(k)}$ can now be solved for $k = 0, 1, 2, \ldots$, in that order, to successively provide the basis of solutions for higher and higher order terms, until reaching some $k = n$ after which we may safely neglect the reamaining terms, hence providing an approximate solution.[13]

The zeroth order term in the perturbation series is decribed by a nonlinear ordinary differential equation of order two, a so-calles *Riccati equation*, which analytically can be solved exactly, either by directly applying the theory of Jacobian elliptic integrals of by applying the Riccati transormation.[14] However, by considering a system starting from rest, at a state of equilibrium, we can immediately draw the conclusion that $p^{(0)}(t)$ must be identically zero for all times $t$. This, of course, only holds for this particular model; in many molecular systems, such in water, a permanent static dipole moment is present, something that is left out in this particular spring model of ours. (Not to be confused with the static polarization induced by the electric field, which by definition of the terms in the perturbation series is included in higher order terms, depending on the power of the electric field.)

The first order term in the perturbation series is the first and only one with an explicit dependence of the electric field of the light. Since the zeroth order perturbation term is zero, the differential equation for the first order term is linear, which simplifies the calculus. However, since it is an inhomogeneous differential equation, we must generally look for a total solution to the equation as a sum of a homogeneous solution (with zero right hand side) and a particular solution (with the electric field in the right hand side present). The homogeneous solution, which will contain two constants of integration (since we are considering second-order ordinary differential equation) will though only give the part of the solution which depend on initial conditions, that is to say,

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[13] It should though be emphasized that in the limit $n \to \infty$, the described theory still is an exact description of the motion of the electric dipole moment within this model of interaction between light and matter.

[14] For examples of the application of the Riccati transformation, see Zwillinger, *Handbook of Differential Equations*, 2nd ed. (Academic Press, Boston, 1992).

in this case a harmonic natural oscillation of the spring system which in the presence of damping terms rapidly would decrease to zero. This implies that in order to find steady-state solutions, in which the oscillation of the dipole moment directly follows the oscillation of the electric field of the light, we may directly start looking for the particular solution. For a time harmonic electric field, here taken as

$$E(t) = E_\omega \sin(\omega t),$$

the particular solution for the first order term is after some straightforward algebra given as[15]

$$p^{(1)} = \frac{(e^2/m_{\rm r})}{(k_0/m_{\rm r} - \omega^2)} E_\omega \sin(\omega t), \qquad \omega^2 \neq (k_0/m_{\rm r}).$$

For a material consisting of $N$ dipoles per unit volume, and by following the conventions for the linear electric susceptibility in SI units, this corresponds to a first order electric polarization density of the form

$$\begin{aligned} P^{(1)}(t) &= P^{(1)}_\omega \sin(\omega t) \\ &= \varepsilon_0 \chi^{(1)}(\omega) E_\omega \sin(\omega t), \end{aligned}$$

with the *first order (linear) electric susceptibility* given as

$$\chi^{(1)}(\omega) = \chi^{(1)}(-\omega;\omega) = \frac{N}{\varepsilon_0} \frac{(e^2/m_{\rm r})}{(\Omega^2 - \omega^2)},$$

where the resonance frequency $\Omega^2 = k_0/m_{\rm r}$ was introduced. The Lorenzian shape of the frequency dependence is shown in Fig. 6.

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[15] For the sake of self consistency, the general solution for the first order term is given as

$$p^{(1)} = A\cos((k_0/m_{\rm r})^{1/2} t) + B\sin((k_0/m_{\rm r})^{1/2} t) + \frac{e^2/m_{\rm r}}{k_0/m_{\rm r} - \omega^2} E_\omega \sin(\omega t),$$

where $A$ and $B$ are constants of integration, determined by initial conditions.

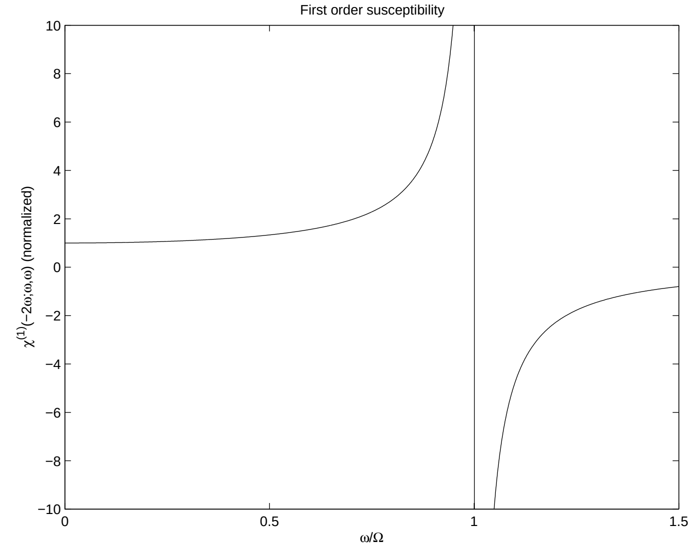

Figure 6. Lorenzian shape of the linear susceptibility $\chi^{(1)}(-\omega;\omega)$.

Continuing with the second order perturbation term, some straightforward algebra gives that the particular solution for the second order term of the electric dipole moment becomes

$$\begin{aligned} p^{(2)}(t) = &-\frac{k_1 e^3}{2k_0 m_{\mathrm{r}}^2}\frac{1}{(\Omega^2-\omega^2)}E_\omega^2 + \frac{k_1 e^3}{2m_{\mathrm{r}}^3}\frac{1}{(\Omega^2-\omega^2)(\Omega^2-4\omega^2)}E_\omega^2 \\ &+ \frac{k_1 e^3}{m_{\mathrm{r}}^3}\frac{1}{(\Omega^2-\omega^2)(\Omega^2-4\omega^2)}E_\omega^2 \sin^2(\omega t). \end{aligned}$$

In terms of the polarization density of the medium, still with $N$ dipoles per unit volume and following the conventions in regular SI units, this can be written as

$$\begin{aligned} P^{(2)}(t) &= P_0^{(0)} + P_{2\omega}^{(0)}\sin(2\omega t) \\ &= \underbrace{\varepsilon_0\chi^{(2)}(0;\omega,-\omega)E_\omega E_\omega}_{\text{DC polarization}} + \underbrace{\varepsilon_0\chi^{(2)}(-2\omega;\omega,\omega)E_\omega E_\omega\sin(2\omega)}_{\text{second harmonic polarization}} \end{aligned}$$

with the *second order (quadratic) electric susceptibility* given as

$$\begin{aligned} \chi^{(2)}(0;\omega,-\omega) &= \frac{N}{\varepsilon_0}\frac{k_1 e^3}{2m_{\mathrm{r}}^3}\left[\frac{1}{(\Omega^2-\omega^2)(\Omega^2-4\omega^2)} - \frac{1}{\Omega^2(\Omega^2-\omega^2)}\right], \\ \chi^{(2)}(2\omega;\omega,\omega) &= \frac{N}{\varepsilon_0}\frac{k_1 e^3}{m_{\mathrm{r}}^3}\frac{1}{(\Omega^2-\omega^2)(\Omega^2-4\omega^2)}. \end{aligned}$$

From this we may notice that for one-photon resonances, the nonlinearities are enhanced whenever $\omega \approx \Omega$ or $2\omega \approx \Omega$, for the induced DC as well as the second harmonic polarization density.

The explicit frequency dependencies of the susceptibilities $\chi^{(2)}(-2\omega;\omega,\omega)$ and $\chi^{(2)}(0;\omega,-\omega)$ are shown in Figs. 7 and 8.

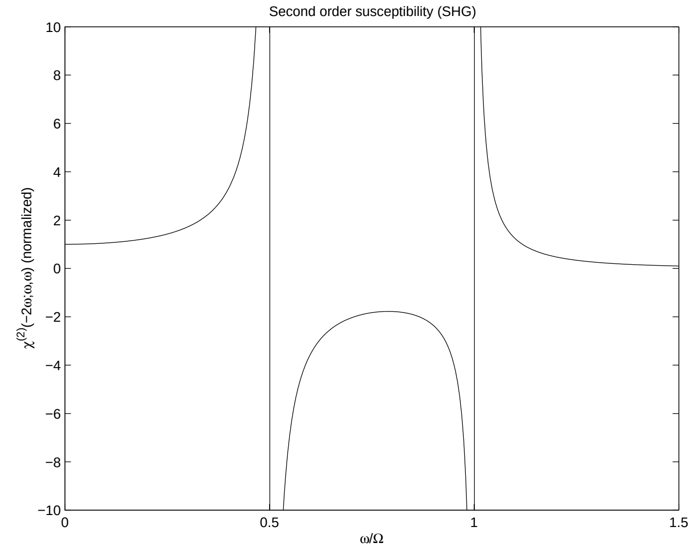

Figure 7. Lorenzian shape of the linear susceptibility $\chi^{(2)}(-2\omega;\omega,\omega)$ (SHG).

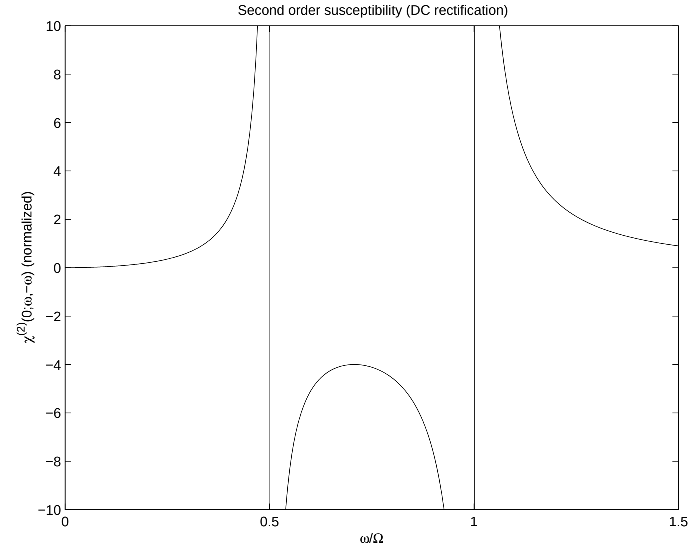

Figure 8. Lorenzian shape of the linear susceptibility $\chi^{(2)}(0;\omega,-\omega)$ (DC).

A well known fact in electromagnetic theory is that an electric dipole that oscillates at a certain angular frequency, say at $2\omega$, also emits electromagnetic radiation at this frequency. In particular, this implies that the term described by the susceptibility $\chi^{(2)}(-2\omega;\omega,\omega)$ will generate light at twice the angular frequency of the light, hence generating a second harmonic light wave.

# Lecture II

**Lecture Notes on Nonlinear Optics**
Nonlinear Optics (5A5513, 5p for advanced undergraduate and doctoral students)
Course given at the Royal Institute of Technology,
Department of Laser Physics and Quantum Optics
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## LECTURE 2

### Nonlinear polarization density

From the introductory perturbation analysis of the all-classical anharmonic oscillator in the previous lecture, we now *a priori* know that it is possible to express the electric polarization density as a power series in the electric field of the optical wave.

Loosely formulated, the electric polarization density in complex notation can be taken as the series

$$\begin{aligned} P_\mu(\omega_\sigma) = \varepsilon_0[ &\underbrace{\chi^{(1)}_{\mu\alpha}(-\omega_\sigma;\omega_\sigma)E_\alpha(\omega_\sigma)}_{\sim P^{(1)}_\mu(\omega_\sigma)} + \underbrace{\chi^{(2)}_{\mu\alpha\beta}(-\omega_\sigma;\omega_1,\omega_2)E_\alpha(\omega_1)E_\beta(\omega_2)}_{\sim P^{(2)}_\mu(\omega_\sigma),\ \omega_\sigma=\omega_1+\omega_2} \\ &+ \underbrace{\chi^{(3)}_{\mu\alpha\beta\gamma}(-\omega_\sigma;\omega_1,\omega_2,\omega_3)E_\alpha(\omega_1)E_\beta(\omega_2)E_\gamma(\omega_3)}_{\sim P^{(3)}_\mu(\omega_\sigma),\ \omega_\sigma=\omega_1+\omega_2+\omega_3} + \ldots] \end{aligned}$$

where we adopted Einstein's convention of summation for terms with repeated subscripts. (A more formal formulation of the polarization density will be described later.)

### Symmetries in nonlinear optics

There are essentially four classes of symmetries that we will encounter in this course:

[1] Intrinsic permutation symmetry:

$$\begin{aligned} \chi^{(3)}_{\mu\alpha\beta\gamma}(-\omega_\sigma;\omega_1,\omega_2,\omega_3) &= \chi^{(3)}_{\mu\beta\alpha\gamma}(-\omega_\sigma;\omega_2,\omega_1,\omega_3) \\ &= \chi^{(3)}_{\mu\beta\gamma\alpha}(-\omega_\sigma;\omega_2,\omega_3,\omega_1) \\ &= \chi^{(3)}_{\mu\gamma\beta\alpha}(-\omega_\sigma;\omega_3,\omega_2,\omega_1), \end{aligned}$$

that is to say, invariance under the $n!$ possible permutations of $(\alpha_k,\omega_k)$, $k=1,2,\ldots,n$. This principle applies generally to resonant as well as nonresonant media. (*Described in this lecture.*)

[2] Overall permutation symmetry:

$$\begin{aligned} \chi^{(3)}_{\mu\alpha\beta\gamma}(-\omega_\sigma;\omega_1,\omega_2,\omega_3) &= \chi^{(3)}_{\alpha\mu\beta\gamma}(\omega_1;-\omega_\sigma,\omega_2,\omega_3) \\ &= \chi^{(3)}_{\alpha\beta\mu\gamma}(\omega_1;\omega_2,-\omega_\sigma,\omega_3) \\ &= \chi^{(3)}_{\alpha\beta\gamma\mu}(\omega_1;\omega_2,\omega_3,-\omega_\sigma), \end{aligned}$$

that is to say, invariance under the $(n+1)!$ possible permutations of $(\mu,-\omega_\sigma)$, $(\alpha_k,\omega_k)$, $k=1,2,\ldots,n$. This principle applies to nonresonant media, where all optical frequencies appearing in the formula for the susceptibility are removed far from the transition frequencies of the medium. (*Described in lecture 4.*)

[3] Kleinman symmetry:

$$\begin{aligned} \chi^{(3)}_{\mu\alpha\beta\gamma}(-\omega_\sigma;\omega_1,\omega_2,\omega_3) &= \chi^{(3)}_{\alpha\mu\beta\gamma}(-\omega_\sigma;\omega_1,\omega_2,\omega_3) \\ &= \chi^{(3)}_{\alpha\beta\mu\gamma}(-\omega_\sigma;\omega_1,\omega_2,\omega_3) \\ &= \chi^{(3)}_{\alpha\beta\gamma\mu}(-\omega_\sigma;\omega_1,\omega_2,\omega_3), \end{aligned}$$

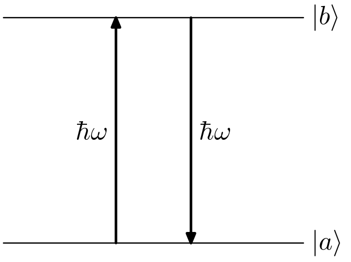
that is to say, invariance under the $(n+1)!$ possible permutations of the subscripts $\mu, \alpha_1, \ldots, \alpha_n$. This principle is a consequence of the overall permutation symmetry, and applies in the low-frequency limit of nonresonant media.

[4] Spatial symmetries, given by the point symmetry class of the medium. (*Described in lecture 6.*)

**Conditions for observing nonlinear optical interactions**

Loosely formulated, a nonlinear response between light and matter depends on one of two key indgredients: either there is a resonance between the light wave and some natural oscillation mode of the medium, or the light is sufficiently intense. *Direct resonance* can occur in isolated intervals of the electromagnetic spectrum at

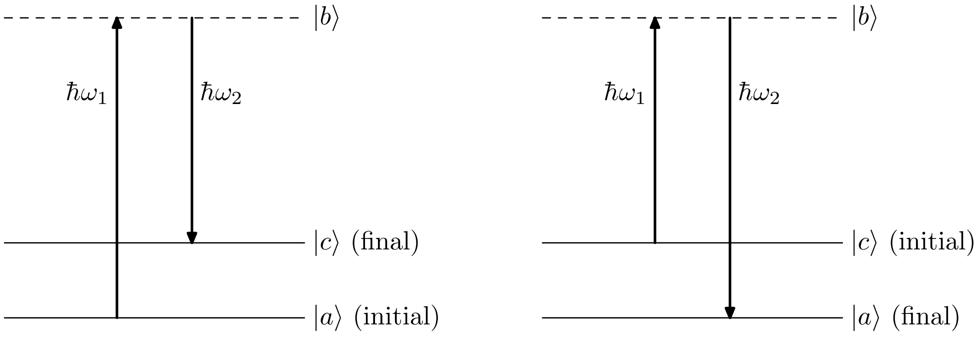
- ultraviolet and visible frequencies ($10^{15}$ s$^{-1}$) where the oscillator corresponds to an electronic transition of the medium,
- infrared ($10^{13}$ s$^{-1}$), where the medium has vibrational modes, and
- the far infrared-microwave range ($10^{11}$ s$^{-1}$), where there are rotational modes. These interactions are also called *one-photon processes*, and are schematically illustrated in Fig. 3.



The one-photon transition.

Figure 3. Transition scheme of the one-photon process.

The lower frequency modes can be excited at optical frequencies ($10^{15}$ s$^{-1}$) through indirect resonant processes in which the difference in frequencies and wave vectors of two light waves, called the pump and Stokes wave, respectively, matches the frequency and wave vector of one of these lower frequency modes. These three-frequency interactions are sometime called *two-photon processes*. In the case where there "lower frequency" mode is an electronic transition or in the vibrational range (in which case the Stokes frequency can be of the same order of magnitude as that of the light wave), this process is called *Raman scattering*.



The Stokes Raman transition. The Anti-Stokes Raman transition.

Figure 4. Transition schemes of stimulated Raman Scattering.

The stimulated Raman scattering is essentially a two-photon process in which one photon at $\omega_1$ is absorbed and one photon at $\omega_2$ is emitted, while the material makes a transition from the initial state $|a\rangle$ to the final state $|c\rangle$, as shown in Fig. 4. Energy conservation requires the Raman resonance frequency (electronic or vibrational) to satisfy $\hbar\Omega_{ca} = \hbar(\omega_1 - \omega_2)$, and hence we may classify the Stokes and Anti-Stokes transitions as

$$\begin{aligned} \hbar\Omega_{ca} > 0 \quad &\Leftrightarrow \quad \text{Stokes Raman} \\ \hbar\Omega_{ca} < 0 \quad &\Leftrightarrow \quad \text{Anti} - \text{Stokes Raman} \end{aligned}$$

When the lower frequency mode instead is an *acoustic* mode of the material, the process is instead called *Brillouin scattering*.

As will be shown explicitly later on in the course, optical resonance with transitions of the material is an important tool for "boosting up" the nonlinearities, with enhanced possibilities of applications. However, at single-photon resonance we have a strong absorption at the frequency of the optical field, and in most cases this is a non-desirable effect, since it decreases the optical intensity, even though it meanwhile also enhances the nonlinearity of the material.

However, by instead exploiting the two-photon resonance, the desired nonlinearity can be significantly enhanced whilst at the same time the competing absorption process can be minimized by avoiding coincidences between optical frequencies and *single-photon* resonances.

One general drawback with the resonant enhancement is that the response time of the polarization density of the material is slowed down, affecting applications such as optical switching or modulation, where speed is of importance.

### Phenomenological description of the susceptibility tensors

Before entering the full quantum-mechanical formalism, we will assume the medium to possess a temporal response described by *time response functions* $R(t)$.

The very first step in the analysis is to express the electric polarization density, which in classical electrodynamical terms is expressed as the sum over all $M$ electric charges $q_k$ in a small volume $V$ centered at $\mathbf{r}$,

$$\mathbf{P}(\mathbf{r},t) = \frac{1}{V}\sum_{k=1}^{M} q_k \mathbf{r}_k(t),$$

as a series expansion

$$\mathbf{P}(\mathbf{r},t) = \mathbf{P}^{(0)}(\mathbf{r},t) + \mathbf{P}^{(1)}(\mathbf{r},t) + \mathbf{P}^{(2)}(\mathbf{r},t) + \ldots + \mathbf{P}^{(n)}(\mathbf{r},t) + \ldots,$$

where $\mathbf{P}^{(1)}(\mathbf{r},t)$ is linear in the electric field, $\mathbf{P}^{(2)}(\mathbf{r},t)$ is quadratic in the electric field, etc. The field-independent $\mathbf{P}^{(0)}(\mathbf{r},t)$ corresponds to eventually appearing static polarization of the medium. It should be emphasized that any of these terms may be linear as well as nonlinear functions of the electric field of the optical wave; i. e. an externally applied static electric field may together with the electric field of the light interact through, for example, $\mathbf{P}^{(3)}(\mathbf{r},t)$ to induce an electric polarization that is linear in the optical field.

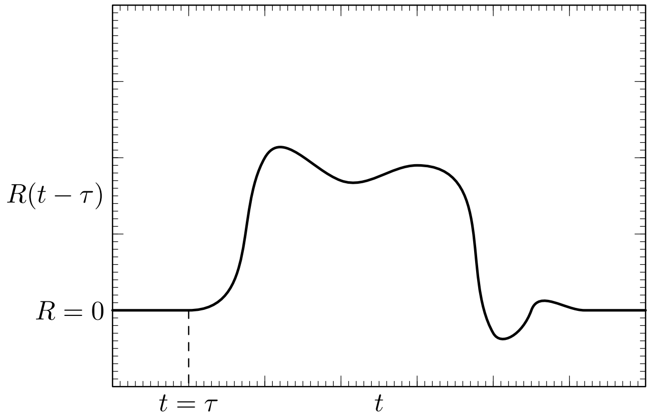


Figure 9. Schematic form of a possible response function in time domain.

In the analysis that now is to follow, we will a priori assume that it is possible to express the polarization density as a perturbation series. Later on, we will show how each of the terms can be derived from a more stringent basis, where it also will be stated which conditions that must hold in order to apply perturbation theory.

**Linear polarization response function**

Since $\mathbf{P}^{(1)}(\mathbf{r},t)$ is taken as linear in the electric field, we may express the linear polarization density of the medium as being related to the optical field as

$$P_{\mu}^{(1)}(\mathbf{r},t)=\varepsilon_0\int_{-\infty}^{\infty}T_{\mu\alpha}^{(1)}(t;\tau)E_{\alpha}(\mathbf{r},\tau)\,d\tau, \tag{7}$$

where $T_{\mu\alpha}^{(1)}(t;\tau)$ is a rank-two tensor that weights all contributions in time from the electric field of the light. A few required properties of $T_{\mu\alpha}^{(1)}(t;\tau)$ can immediately be stated:

- *Causality.* We require that no optically induced contribution can occur *before* the field is applied, i. e. $T_{\mu\alpha}^{(1)}(t;\tau)=0$ for $t\le\tau$.
- *Time invariance.* Under most circumstances, we may in addition assume that the material parameters are constant in time, such that $P_{\mu}^{(1)}(\mathbf{r},t')$ is identical to the polarisation as induced by the time-displaced electric field $E_{\alpha}(\mathbf{r},t')$.

The second of these properties is essentially a manifestation of an adiabatically following change of the carrier wave of the optical field.

By using Eq. (7), the time invariance of the constitutive relation gives that

$$\begin{aligned}P_{\mu}^{(1)}(\mathbf{r},t+t_0)&=\varepsilon_0\int_{-\infty}^{\infty}T_{\mu\alpha}^{(1)}(t+t_0;\tau)E_{\alpha}(\mathbf{r},\tau)\,d\tau\\&=\big\{\text{Should equal to polarization induced by } E_{\alpha}(\mathbf{r},\tau+t_0)\big\}\\&=\varepsilon_0\int_{-\infty}^{\infty}T_{\mu\alpha}^{(1)}(t;\tau)E_{\alpha}(\mathbf{r},\tau+t_0)\,d\tau\\&=\big\{\tau'=\tau+t_0\big\}\\&=\varepsilon_0\int_{-\infty}^{\infty}T_{\mu\alpha}^{(1)}(t;\tau'-t_0)E_{\alpha}(\mathbf{r},\tau')\,d\tau',\end{aligned}$$

from which we, by changing the "dummy" variable of integration back to $\tau$, obtain the relation

$$T_{\mu\alpha}^{(1)}(t+t_0;\tau)=T_{\mu\alpha}^{(1)}(t;\tau-t_0).$$

In particular, by setting $t=0$ and replacing the arbitrary time displacement $t_0$ by $t$, one finds that the response of the medium depends only of the time difference $\tau-t$,

$$\begin{aligned}T_{\mu\alpha}^{(1)}(t;\tau)&=T_{\mu\alpha}^{(1)}(0;\tau-t)\\&=R_{\mu\alpha}^{(1)}(\tau-t)\end{aligned}$$

where we defined the linear polarization response function $R_{\mu\alpha}^{(1)}(\tau-t)$, being a rank-two tensor depending only on the time difference $\tau-t$.

To summarize, the linear contribution to the electric polarization density is given in terms of the linear polarization response function as

$$\begin{aligned}P_{\mu}^{(1)}(\mathbf{r},t)&=\varepsilon_0\int_{-\infty}^{\infty}R_{\mu\alpha}^{(1)}(t-\tau)E_{\alpha}(\mathbf{r},\tau)\,d\tau,\\&=\varepsilon_0\int_{-\infty}^{\infty}R_{\mu\alpha}^{(1)}(\tau')E_{\alpha}(\mathbf{r},t-\tau')\,d\tau',\end{aligned} \tag{8}$$

and the causality condition for the linear polarization response function requires that

$$R_{\mu\alpha}^{(1)}(\tau-t)=0,\qquad t\le\tau,$$

and in addition, since the relation (8) is to hold for arbitrary time evolution of the electrical field, and since the polarization density $P_{\mu}^{(1)}(\mathbf{r},t)$ and the electric field $E_{\alpha}^{(1)}(\mathbf{r},t)$ both are real-valued quantities, we also require the polarization response function $R_{\mu\alpha}^{(1)}(\tau-t)$ to be a real-valued function.

**Quadratic polarization response function**

The second-order, quadratic polarization density can in similar to the linear one be phenomenologically be written as the sum of all infinitesimal previous contributions in time. In this case, we must though include the possibility that not all contributions origin in the same time scale, and hence the proper formulation of the second order polarization density is as a two-dimensional integral,

$$P_\mu^{(2)}(\mathbf{r},t) = \varepsilon_0 \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} T^{(2)}_{\mu\alpha\beta}(t;\tau_1,\tau_2)E_\alpha(\mathbf{r},\tau_1)E_\beta(\mathbf{r},\tau_2)\,d\tau_1\,d\tau_2. \tag{9}$$

The tensor $T^{(2)}_{\mu\alpha\beta}(t;\tau_1,\tau_2)$ uniquely determines the quadratic, second-order polarization of the medium. However, the tensor $T^{(2)}_{\mu\alpha\beta}(t;\tau_1,\tau_2)$ is not itself unique. To see this, we may express the tensor as a sum of a symmetric part and an antisymmetric part,

$$\begin{aligned}
T^{(2)}_{\mu\alpha\beta}(t;\tau_1,\tau_2) &= \underbrace{\frac{1}{2}\Big[T^{(2)}_{\mu\alpha\beta}(t;\tau_1,\tau_2)+T^{(2)}_{\mu\beta\alpha}(t;\tau_2,\tau_1)\Big]}_{\text{symmetric}} + \underbrace{\frac{1}{2}\Big[T^{(2)}_{\mu\alpha\beta}(t;\tau_1,\tau_2)-T^{(2)}_{\mu\beta\alpha}(t;\tau_2,\tau_1)\Big]}_{\text{antisymmetric}}\\
&= \underbrace{S^{(2)}_{\mu\alpha\beta}(t;\tau_1,\tau_2)}_{=S^{(2)}_{\mu\beta\alpha}(t;\tau_2,\tau_1)} + \underbrace{A^{(2)}_{\mu\alpha\beta}(t;\tau_1,\tau_2)}_{=-A^{(2)}_{\mu\beta\alpha}(t;\tau_2,\tau_1)}
\end{aligned}$$

As this form of the response function is inserted into the original expression (9) for the second order polarization density, we immediately find that it is left invariant under the interchange of the dummy variables $(\alpha,\tau_1)$ and $(\beta,\tau_2)$ (since the integration is performed from minus to plus infinity in time). In particular, it from this follows that

$$\begin{aligned}
P_\mu^{(2)}(\mathbf{r},t) &= \varepsilon_0 \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} [S^{(2)}_{\mu\alpha\beta}(t;\tau_1,\tau_2)+A^{(2)}_{\mu\alpha\beta}(t;\tau_1,\tau_2)]E_\alpha(\mathbf{r},\tau_1)E_\beta(\mathbf{r},\tau_2)\,d\tau_1\,d\tau_2\\
&= \underbrace{\varepsilon_0 \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} S^{(2)}_{\mu\alpha\beta}(t;\tau_1,\tau_2)E_\alpha(\mathbf{r},\tau_1)E_\beta(\mathbf{r},\tau_2)\,d\tau_1\,d\tau_2}_{\equiv I_1}\\
&\qquad + \underbrace{\varepsilon_0 \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} A^{(2)}_{\mu\alpha\beta}(t;\tau_1,\tau_2)E_\alpha(\mathbf{r},\tau_1)E_\beta(\mathbf{r},\tau_2)\,d\tau_1\,d\tau_2}_{\equiv I_2}\\
&= \varepsilon_0 \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} S^{(2)}_{\mu\beta\alpha}(t;\tau_2,\tau_1)E_\alpha(\mathbf{r},\tau_1)E_\beta(\mathbf{r},\tau_2)\,d\tau_1\,d\tau_2\\
&\qquad - \varepsilon_0 \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} A^{(2)}_{\mu\beta\alpha}(t;\tau_2,\tau_1)E_\alpha(\mathbf{r},\tau_1)E_\beta(\mathbf{r},\tau_2)\,d\tau_1\,d\tau_2\\
&= \underbrace{\varepsilon_0 \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} S^{(2)}_{\mu\beta\alpha}(t;\tau_2,\tau_1)E_\beta(\mathbf{r},\tau_2)E_\alpha(\mathbf{r},\tau_1)\,d\tau_1\,d\tau_2}_{=I_1}\\
&\qquad - \underbrace{\varepsilon_0 \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} A^{(2)}_{\mu\beta\alpha}(t;\tau_2,\tau_1)E_\beta(\mathbf{r},\tau_2)E_\alpha(\mathbf{r},\tau_1)\,d\tau_1\,d\tau_2}_{=I_2}
\end{aligned}$$

i. e. the symmetric part satisfy the trivial identity $I_1 = I_1$, while the antisymmetric part satisfy $I_2 = -I_2$, i. e. the antisymmetric part does not contribute to the polarization density, and that we may set the antiymmetric part to zero, without imposing any constraint on the validity of the theory.

The time response $T^{(2)}_{\mu\alpha\beta}(t;\tau_1,\tau_2)$ is then unique and now chosen to be symmetric,

$$T^{(2)}_{\mu\alpha\beta}(t;\tau_1,\tau_2) = T^{(2)}_{\mu\beta\alpha}(t;\tau_2,\tau_1).$$

This procedure of symmetrization may seem like an all theoretical contruction, somewhat out of focus of what lies ahead, but in fact it turns out to be an extremely useful property that we later on will exploit extensively. The previously described symmetric property will later on, for susceptibility tensors in the frequency domain, be denoted as the *intrinsic permutation symmetry*, a general property that will hold irregardless of whether the nonlinear interaction under analysis is highly resonant or far from resonance.

By again applying the arguments of time invariance, as previously for the linear response function, we find that

$$T^{(2)}_{\mu\alpha\beta}(t+t_0;\tau_1,\tau_2) = T^{(2)}_{\mu\alpha\beta}(t;\tau_1-t_0,\tau_2-t_0)$$

for all $t$, $\tau_1$, and $\tau_2$. Hence, by setting $t=0$ and then replacing th arbitrary time $t_0$ by $t$, again as previously done for the linear case, one finds that $T^{(2)}(t;\tau_1,\tau_2)$ depends only on the two time differences $t-\tau_1$ and $t-\tau_2$. To make this fact explicit, we may hence write the response function as

$$T^{(2)}_{\mu\alpha\beta}(t;\tau_1,\tau_2) = R^{(2)}_{\mu\alpha\beta}(t-\tau_1,t-\tau_2),$$

giving the canonical form of the quadratic polarization density as

$$\begin{aligned} P^{(2)}_{\mu}(\mathbf{r},t) &= \varepsilon_0 \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} R^{(2)}_{\mu\alpha\beta}(t-\tau_1,t-\tau_2)E_\alpha(\mathbf{r},\tau_1)E_\beta(\mathbf{r},\tau_2)\,d\tau_1\,d\tau_2 \\ &= \varepsilon_0 \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} R^{(2)}_{\mu\alpha\beta}(\tau_1',\tau_2')E_\alpha(\mathbf{r},t-\tau_1')E_\beta(\mathbf{r},t-\tau_2')\,d\tau_1'\,d\tau_2'. \end{aligned} \tag{10}$$

The tensor $R^{(2)}_{\mu\alpha\beta}(\tau_1,\tau_2)$ is called the quadratic electric polarization response of the medium, and in similar with the linear response function, arguments of causality require the response function to be zero whenever $\tau_1$ and/or $\tau_2$ is negative. Similarly, the reality condition on $E_\alpha(\mathbf{r},t)$ and $P_\alpha(\mathbf{r},t)$ requires that $R^{(2)}_{\mu\alpha\beta}(\tau_1,\tau_2)$ is a real-valued function.

### Higher order polarization response functions

The $n$th order polarization density can in similar to the linear ($n=1$ and quadratic ($n=2$) ones be written as

$$\begin{aligned} P^{(n)}_{\mu}(\mathbf{r},t) &= \varepsilon_0 \int_{-\infty}^{\infty}\cdots\int_{-\infty}^{\infty} R^{(n)}_{\mu\alpha_1\cdots\alpha_n}(t-\tau_1,\ldots,t-\tau_n)E_{\alpha_1}(\mathbf{r},\tau_1)\cdots E_{\alpha_n}(\mathbf{r},\tau_n)\,d\tau_1\,\cdots\,d\tau_n \\ &= \varepsilon_0 \int_{-\infty}^{\infty}\cdots\int_{-\infty}^{\infty} R^{(n)}_{\mu\alpha_1\cdots\alpha_n}(\tau_1',\ldots,\tau_n')E_{\alpha_1}(\mathbf{r},t-\tau_1')\cdots E_{\alpha_n}(\mathbf{r},t-\tau_n')\,d\tau_1'\,\cdots\,d\tau_n', \end{aligned} \tag{11}$$

where the $n$th order response function is a tensor of rank $n+1$, and a real-valued function of the $n$ parameters $\tau_1,\ldots,\tau_n$, vanishing whenever any $\tau_k<0$, $k=1,2,\ldots,n$, and with the *intrinsic permutation symmetry* that it is left invariant under any of the $n!$ pairwise permutations of $(\alpha_1,\tau_1),\ldots,(\alpha_n,\tau_n)$.

# Lecture III

**Lecture Notes on Nonlinear Optics**
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# LECTURE 3

## Susceptibility tensors in the frequency domain

The susceptibility tensors in the frequency domain arise when the electric field $E_\alpha(t)$ of the light is expressed in terms of its Fourier transform $E_\alpha(\omega)$, by means of the Fourier integral identity

$$E_\alpha(t) = \int_{-\infty}^{\infty} E_\alpha(\omega)\exp(-i\omega t)\,d\omega = \mathfrak{F}^{-1}[E_\alpha](t), \tag{1'}$$

with inverse relation

$$E_\alpha(\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} E_\alpha(\tau)\exp(i\omega\tau)\,d\tau = \mathfrak{F}[E_\alpha](\omega). \tag{1''}$$

This convention of inclusion of the factor of $2\pi$, as well as the sign convention, is commonly used in quantum mechanics; however, it should be emphasized that this convention is not a commonly adopted standard in optics, neither in linear nor in nonlinear optical regimes.

The sign convention here used leads to wave solutions of the form $f(kz-\omega t)$ for monochromatic waves propagating in the positive $z$-direction, which might be somewhat more intuitive than the alternative form $f(\omega t - kz)$, which is obtained if one instead apply the alternative sign convention.

The convention for the inclusion of $2\pi$ in the Fourier transform in Eq. $(1'')$ is here convenient for description of electromagnetic wave propagation in the frequency domain (going from the time domain description, in terms of the polarization response functions, to the frequency domain, in terms of the linear and nonlinear susceptibilities), since it enables us to omit any multiple of $2\pi$ of the Fourier transformed fields.

## First order susceptibility tensor

By inserting Eq. $(1')$ is inserted into the previously obtained[1] relation for the first order, linear polarization density, one obtains

$$\begin{aligned}
P^{(1)}_\mu(\mathbf{r},t) &= \varepsilon_0 \int_{-\infty}^{\infty} R^{(1)}_{\mu\alpha}(\tau)E_\alpha(\mathbf{r},t-\tau)\,d\tau,\\
&= \{\text{express } E_\alpha(\mathbf{r},t-\tau) \text{ in frequency domain}\}\\
&= \varepsilon_0 \int_{-\infty}^{\infty} R^{(1)}_{\mu\alpha}(\tau)\int_{-\infty}^{\infty} E_\alpha(\mathbf{r},\omega)\exp[-i\omega(t-\tau)]\,d\omega\,d\tau,\\
&= \{\text{change order of integration}\}\\
&= \varepsilon_0 \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} R^{(1)}_{\mu\alpha}(\tau)E_\alpha(\mathbf{r},\omega)\exp(i\omega\tau)\,d\tau\ \exp(-i\omega t)\,d\omega,\\
&= \varepsilon_0 \int_{-\infty}^{\infty} \chi^{(1)}_{\mu\alpha}(-\omega;\omega)E_\alpha(\mathbf{r},\omega)\exp(-i\omega t)\,d\omega,
\end{aligned} \tag{2}$$

where the *linear electric dipolar susceptibility*,

$$\chi^{(1)}_{\mu\alpha}(-\omega;\omega) = \int_{-\infty}^{\infty} R^{(1)}_{\mu\alpha}(\tau)\exp(i\omega\tau)\,d\tau = \mathfrak{F}[R^{(1)}_{\mu\alpha}](\omega), \tag{3}$$

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[1] Expressions for the first order, second order, and $n$th order polarization densities were obtained in lecture two.

was introduced. In this expression for the susceptibility, $\omega_\sigma = \omega$, and the reasons for the somewhat peculiar notation of arguments of the susceptibility will be explained later on in the context of nonlinear susceptibilities.

**Second order susceptibility tensor**

In similar to the linear susceptibility tensor, by inserting Eq. ($1'$) into the previously obtained relation for the second order, quadratic polarization density, one obtains

$$\begin{aligned} P^{(2)}_\mu(\mathbf{r},t) &= \varepsilon_0 \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} R^{(2)}_{\mu\alpha\beta}(\tau_1,\tau_2) E_\alpha(\mathbf{r},\omega_1) E_\beta(\mathbf{r},\omega_2) \\ &\qquad\qquad \times \exp[-i(\omega_1(t-\tau_1)+\omega_2(t-\tau_2))]\, d\tau_1\, d\tau_2\, d\omega_1\, d\omega_2 \\ &= \varepsilon_0 \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \chi^{(2)}_{\mu\alpha\beta}(-\omega_\sigma;\omega_1,\omega_2) E_\alpha(\mathbf{r},\omega_1) E_\beta(\mathbf{r},\omega_2) \exp[-i\underbrace{(\omega_1+\omega_2)}_{\equiv\omega_\sigma} t]\, d\omega_1\, d\omega_2 \end{aligned} \tag{4}$$

where the *quadratic electric dipolar susceptibility*,

$$\chi^{(2)}_{\mu\alpha\beta}(-\omega_\sigma;\omega_1,\omega_2) = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} R^{(2)}_{\mu\alpha\beta}(\tau_1,\tau_2) \exp[i(\omega_1\tau_1+\omega_2\tau_2)]\, d\tau_1\, d\tau_2, \tag{5}$$

was introduced. In this expression for the susceptibility, $\omega_\sigma = \omega_1 + \omega_2$, and the reason for the notation of arguments should now be somewhat more clear: the first angular frequency argument of the susceptibility tensor is simply the sum of all driving angular frequencies of the optical field.

The intrinsic permutation symmetry of $R^{(2)}_{\mu\alpha\beta}(\tau_1,\tau_2)$, the second order polarization response function, carries over to the second order susceptibility tensor as well, in the sense that

$$\chi^{(2)}_{\mu\alpha\beta}(-\omega_\sigma;\omega_1,\omega_2) = \chi^{(2)}_{\mu\beta\alpha}(-\omega_\sigma;\omega_2,\omega_1),$$

i. e. the second order susceptibility is invariant under any of the $2! = 2$ pairwise permutations of $(\alpha,\omega_1)$ and $(\beta,\omega_2)$.

**Higher order susceptibility tensors**

In similar to the linear and quadratic susceptibility tensors, by inserting Eq. ($1'$) into the previously obtained relation for the $n$th order polarization density, one obtains

$$\begin{aligned} P^{(n)}_\mu(\mathbf{r},t) = \varepsilon_0 \int_{-\infty}^{\infty}\cdots\int_{-\infty}^{\infty} \chi^{(n)}_{\mu\alpha_1\cdots\alpha_n}(-\omega_\sigma;\omega_1,\ldots,\omega_n) E_{\alpha_1}(\mathbf{r},\omega_1)\cdots E_{\alpha_n}(\mathbf{r},\omega_n) \\ \times \exp[-i\underbrace{(\omega_1+\ldots+\omega_n)}_{\equiv\omega_\sigma} t]\, d\omega_1 \cdots d\omega_n, \end{aligned} \tag{6}$$

where the *nth order electric dipolar susceptibility*,

$$\chi^{(n)}_{\mu\alpha_1\cdots\alpha_n}(-\omega_\sigma;\omega_1,\ldots,\omega_n) = \int_{-\infty}^{\infty}\cdots\int_{-\infty}^{\infty} R^{(n)}_{\mu\alpha_1\cdots\alpha_n}(\tau_1,\ldots,\tau_n) \exp[i(\omega_1\tau_1+\ldots+\omega_n\tau_n)]\, d\tau_1 \cdots d\tau_n, \tag{7}$$

was introduced, and where, as previously,

$$\omega_\sigma = \omega_1 + \omega_2 + \ldots + \omega_n.$$

The intrinsic permutation symmetry of $R^{(n)}_{\mu\alpha_1\cdots\alpha_n}(\tau_1,\ldots,\tau_n)$, the $n$th order polarization response function, also in this general case carries over to the $n$th order susceptibility tensor as well, in the sense that

$$\chi^{(n)}_{\mu\alpha_1\alpha_2\cdots\alpha_n}(-\omega_\sigma;\omega_1,\omega_2,\ldots,\omega_n)$$

is invariant under any of the $n!$ pairwise permutations of $(\alpha_1,\omega_1)$, $(\alpha_2,\omega_2)$, …, $(\alpha_n,\omega_n)$.

**Monochromatic fields**

It should at this stage be emphasized that even though the electric field via the Fourier integral identity can be seen as a superposition of infinitely many infinitesimally narrow band monochromatic components, the superposition principle of linear optics, which states that the wave equation may be independently solved for each frequency component of the light, generally does *not* hold in nonlinear optics.

For monochromatic light, the electric field can be written as a superposition of a set of distinct terms in time domain as

$$\mathbf{E}(\mathbf{r},t)=\sum_k \mathrm{Re}[\mathbf{E}_{\omega_k}\exp(-i\omega_k t)],$$

with the convention that the involved angular frequencies all are taken as positive, $\omega_k \geq 0$, and the electric field in the frequency domain simply becomes a superposition of delta peaks in the spectrum,

$$\begin{aligned}
\mathbf{E}(\mathbf{r},\omega) &= \frac{1}{2\pi}\int_{-\infty}^{\infty}\mathbf{E}(\mathbf{r},\tau)\exp(i\omega\tau)\,d\tau\\
&= \{\text{express as monochromatic field}\}\\
&= \frac{1}{2\pi}\sum_k\int_{-\infty}^{\infty}\mathrm{Re}[\mathbf{E}_{\omega_k}\exp(-i\omega_k\tau)]\exp(i\omega\tau)\,d\tau\\
&= \{\text{by definition}\}\\
&= \frac{1}{4\pi}\sum_k\int_{-\infty}^{\infty}[\mathbf{E}_{\omega_k}\exp(i(\omega-\omega_k)\tau)+\mathbf{E}^*_{\omega_k}\exp(i(\omega+\omega_k)\tau)]\,d\tau\\
&= \frac{1}{2}\sum_k\Big[\mathbf{E}_{\omega_k}\underbrace{\frac{1}{2\pi}\int_{-\infty}^{\infty}\exp(i(\omega-\omega_k)\tau)\,d\tau}_{\equiv\delta(\omega-\omega_k)}+\mathbf{E}^*_{\omega_k}\underbrace{\frac{1}{2\pi}\int_{-\infty}^{\infty}\exp(i(\omega+\omega_k)\tau)\,d\tau}_{\equiv\delta(\omega+\omega_k)}\Big]\\
&= \{\text{definition of the delta function}\}\\
&= \frac{1}{2}\sum_k[\mathbf{E}_{\omega_k}\delta(\omega-\omega_k)+\mathbf{E}^*_{\omega_k}\delta(\omega+\omega_k)].
\end{aligned}$$

By inserting this form of the electric field (taken in the frequency domain) into the polarization density, one obtains the polarization density in the monochromatic form

$$\mathbf{P}^{(n)}(\mathbf{r},t)=\sum_{\omega_\sigma\geq 0}\mathrm{Re}[\mathbf{P}^{(n)}_{\omega_\sigma}\exp(-i\omega_\sigma t)],$$

with complex-valued Cartesian components at angular frequency $\omega_\sigma$ given as

$$\begin{aligned}
(\mathbf{P}^{(n)}_{\omega_\sigma})_\mu = 2\varepsilon_0\sum_{\alpha_1}\cdots\sum_{\alpha_n} &\chi^{(n)}_{\mu\alpha_1\alpha_2\cdots\alpha_n}(-\omega_\sigma;\omega_1,\omega_2,\dots,\omega_n)(E_{\omega_1})_{\alpha_1}(E_{\omega_2})_{\alpha_2}\cdots(E_{\omega_n})_{\alpha_n}\\
&+\chi^{(n)}_{\mu\alpha_1\alpha_2\cdots\alpha_n}(-\omega_\sigma;\omega_2,\omega_1,\dots,\omega_n)(E_{\omega_2})_{\alpha_1}(E_{\omega_1})_{\alpha_2}\cdots(E_{\omega_n})_{\alpha_n}\\
&+\text{all other distinguishable terms}
\end{aligned}\tag{8}$$

where, as previously, $\omega_\sigma=\omega_1+\omega_2+\ldots+\omega_n$. In the right hand side of Eq. (8), the summation is performed over all distinguishble terms, that is to say over all the possible combinations of $\omega_1,\omega_2,\dots,\omega_n$ that give rise to the particular $\omega_\sigma$. Within this respect, a certain frequency and its negative counterpart are to be considered as distinct frequencies when appearing in the set. In general, there are several possible combinations that give rise to a certain $\omega_\sigma$; for example $(\omega,\omega,-\omega)$, $(\omega,-\omega,\omega)$, and $(-\omega,\omega,\omega)$ form the set of distinct combinations of optical frequencies that give rise to optical Kerr-effect (a field dependent contribution to the polarization density at $\omega_\sigma=\omega+\omega-\omega=\omega$).

A general conclusion of the form of Eq. (8), keeping the intrinsic permutation symmetry in mind, is that only one term needs to be written, and the number of times this term appears in the expression for the polarization density should consequently be equal to the number of distinguishable combinations of $\omega_1,\omega_2,\dots,\omega_n$.

### Convention for description of nonlinear optical polarization

As a "recipe" in theoretical nonlinear optics, Butcher and Cotter provide a very useful convention which is well worth to hold on to. For a superposition of monochromatic waves, and by invoking the general property of the intrinsic permutation symmetry, the monochromatic form of the $n$th order polarization density can be written as

$$(P^{(n)}_{\omega_\sigma})_\mu = \varepsilon_0 \sum_{\alpha_1} \cdots \sum_{\alpha_n} \sum_{\omega} K(-\omega_\sigma;\omega_1,\dots,\omega_n)\chi^{(n)}_{\mu\alpha_1\cdots\alpha_n}(-\omega_\sigma;\omega_1,\dots,\omega_n)(E_{\omega_1})_{\alpha_1}\cdots(E_{\omega_n})_{\alpha_n}. \tag{9}$$

The first summations in Eq. (9), over $\alpha_1,\dots,\alpha_n$, is simply an explicit way of stating that the Einstein convention of summation over repeated indices holds. The summation sign $\sum_\omega$, however, serves as a reminder that the expression that follows is to be summed over *all distinct sets of* $\omega_1,\dots,\omega_n$. Because of the intrinsic permutation symmetry, the frequency arguments appearing in Eq. (9) may be written in arbitrary order.

By "all distinct sets of $\omega_1,\dots,\omega_n$", we here mean that the summation is to be performed, as for example in the case of optical Kerr-effect, over the single set of nonlinear susceptibilities that contribute to a certain angular frequency as $(-\omega;\omega,\omega,-\omega)$ *or* $(-\omega;\omega,-\omega,\omega)$ *or* $(-\omega;-\omega,\omega,\omega)$. In this example, each of the combinations are considered as *distinct*, and it is left as an arbitary choice which one of these sets that are most convenient to use (this is simply a matter of choosing notation, and does not by any means change the description of the interaction).

In Eq. (9), the degeneracy factor $K$ is formally described as

$$K(-\omega_\sigma;\omega_1,\dots,\omega_n) = 2^{l+m-n}p$$

where

$$\begin{aligned} p &= \text{the number of } \textit{distinct} \text{ permutations of } \omega_1,\omega_2,\dots,\omega_1,\\ n &= \text{the order of the nonlinearity},\\ m &= \text{the number of angular frequencies } \omega_k \text{ that are zero, and}\\ l &= \begin{cases} 1, & \text{if } \omega_\sigma \neq 0,\\ 0, & \text{otherwise.}\end{cases} \end{aligned}$$

In other words, $m$ is the number of DC electric fields present, and $l = 0$ if the nonlinearity we are analyzing gives a static, DC, polarization density, such as in the previously (in the spring model) described case of optical rectification in the presence of second harmonic fields (SHG).

A list of frequently encountered nonlinear phenomena in nonlinear optics, including the degeneracy factors as conforming to the above convention, is given in Butcher and Cotters book, Table 2.1, on page 26.

### Note on the complex representation of the optical field

Since the observable electric field of the light, in Butcher and Cotters notation taken as

$$\mathbf{E}(\mathbf{r},t) = \frac{1}{2}\sum_{\omega_k \geq 0}[\mathbf{E}_{\omega_k}\exp(-i\omega_k t) + \mathbf{E}^*_{\omega_k}\exp(i\omega_k t)],$$

is a real-valued quantity, it follows that negative frequencies in the complex notation should be interpreted as the complex conjugate of the respective field component, or

$$\mathbf{E}_{-\omega_k} = \mathbf{E}^*_{\omega_k}.$$

### Example: Optical Kerr-effect

Assume a monochromatic optical wave (containing forward and/or backward propagating components) polarized in the $xy$-plane,

$$\mathbf{E}(z,t) = \mathrm{Re}[\mathbf{E}_\omega(z)\exp(-i\omega_t)] \in \mathbb{R}^3,$$

with all spatial variation of the field contained in

$$\mathbf{E}_\omega(z) = \mathbf{e}_x E^x_\omega(z) + \mathbf{e}_y E^y_\omega(z) \in \mathbb{C}^3.$$

Optical Kerr-effect is in isotropic media described by the third order susceptibility

$$\chi^{(3)}_{\mu\alpha\beta\gamma}(-\omega;\omega,\omega,-\omega),$$

with nonzero components of interest for the $xy$-polarized beam given in Appendix 3.3 of Butcher and Cotters book as

$$\chi^{(3)}_{xxxx} = \chi^{(3)}_{yyyy}, \quad \chi^{(3)}_{xxyy} = \chi^{(3)}_{yyxx} = \left\{\begin{matrix}\text{intr. perm. symm.}\\ (\alpha,\omega) \rightleftharpoons (\beta,\omega)\end{matrix}\right\} = \chi^{(3)}_{xyxy} = \chi^{(3)}_{yxyx}, \quad \chi^{(3)}_{xyyx} = \chi^{(3)}_{yxxy},$$

with

$$\chi^{(3)}_{xxxx} = \chi^{(3)}_{xxyy} + \chi^{(3)}_{xyxy} + \chi^{(3)}_{xyyx}.$$

The degeneracy factor $K(-\omega;\omega,\omega,-\omega)$ is calculated as

$$K(-\omega;\omega,\omega,-\omega) = 2^{l+m-n}p = 2^{1+0-3}3 = 3/4.$$

From this set of nonzero susceptibilities, and using the calculated value of the degeneracy factor in the convention of Butcher and Cotter, we hence have the third order electric polarization density at $\omega_\sigma = \omega$ given as $\mathbf{P}^{(n)}(\mathbf{r},t) = \mathrm{Re}[\mathbf{P}^{(n)}_\omega \exp(-i\omega t)]$, with

$$\begin{aligned}
\mathbf{P}^{(3)}_\omega &= \sum_\mu \mathbf{e}_\mu (P^{(3)}_\omega)_\mu \\
&= \{\text{Using the convention of Butcher and Cotter}\} \\
&= \sum_\mu \mathbf{e}_\mu \Big[\varepsilon_0 \frac{3}{4} \sum_\alpha \sum_\beta \sum_\gamma \chi^{(3)}_{\mu\alpha\beta\gamma}(-\omega;\omega,\omega,-\omega)(E_\omega)_\alpha (E_\omega)_\beta (E_{-\omega})_\gamma\Big] \\
&= \{\text{Evaluate the sums over } (x,y,z) \text{ for field polarized in the } xy \text{ plane}\} \\
&= \varepsilon_0 \frac{3}{4}\{\mathbf{e}_x[\chi^{(3)}_{xxxx} E^x_\omega E^x_\omega E^x_{-\omega} + \chi^{(3)}_{xyyx} E^y_\omega E^y_\omega E^x_{-\omega} + \chi^{(3)}_{xyxy} E^y_\omega E^x_\omega E^y_{-\omega} + \chi^{(3)}_{xxyy} E^x_\omega E^y_\omega E^y_{-\omega}] \\
&\qquad + \mathbf{e}_y[\chi^{(3)}_{yyyy} E^y_\omega E^y_\omega E^y_{-\omega} + \chi^{(3)}_{yxxy} E^x_\omega E^x_\omega E^y_{-\omega} + \chi^{(3)}_{yxyx} E^x_\omega E^y_\omega E^x_{-\omega} + \chi^{(3)}_{yyxx} E^y_\omega E^x_\omega E^x_{-\omega}]\} \\
&= \{\text{Make use of } \mathbf{E}_{-\omega} = \mathbf{E}^*_\omega \text{ and relations } \chi^{(3)}_{xxyy} = \chi^{(3)}_{yyxx}, \text{ etc.}\} \\
&= \varepsilon_0 \frac{3}{4}\{\mathbf{e}_x[\chi^{(3)}_{xxxx} E^x_\omega |E^x_\omega|^2 + \chi^{(3)}_{xyyx} E^{y\,2}_\omega E^{x*}_\omega + \chi^{(3)}_{xyxy} |E^y_\omega|^2 E^x_\omega + \chi^{(3)}_{xxyy} E^x_\omega |E^y_\omega|^2] \\
&\qquad + \mathbf{e}_y[\chi^{(3)}_{xxxx} E^y_\omega |E^y_\omega|^2 + \chi^{(3)}_{xyyx} E^{x\,2}_\omega E^{y*}_\omega + \chi^{(3)}_{xyxy} |E^x_\omega|^2 E^y_\omega + \chi^{(3)}_{xxyy} E^y_\omega |E^x_\omega|^2]\} \\
&= \{\text{Make use of intrinsic permutation symmetry}\} \\
&= \varepsilon_0 \frac{3}{4}\{\mathbf{e}_x[(\chi^{(3)}_{xxxx}|E^x_\omega|^2 + 2\chi^{(3)}_{xxyy}|E^y_\omega|^2)E^x_\omega + (\chi^{(3)}_{xxxx} - 2\chi^{(3)}_{xxyy})E^{y\,2}_\omega E^{x*}_\omega \\
&\qquad \mathbf{e}_y[(\chi^{(3)}_{xxxx}|E^y_\omega|^2 + 2\chi^{(3)}_{xxyy}|E^x_\omega|^2)E^y_\omega + (\chi^{(3)}_{xxxx} - 2\chi^{(3)}_{xxyy})E^{x\,2}_\omega E^{y*}_\omega.
\end{aligned}$$

For the optical field being linearly polarized, say in the $x$-direction, the expression for the polarization density is significantly simplified, to yield

$$\mathbf{P}^{(3)}_\omega = \varepsilon_0 (3/4) \mathbf{e}_x \chi^{(3)}_{xxxx} |E^x_\omega|^2 E^x_\omega,$$

i. e. taking a form that can be interpreted as an intensity-dependent ($\sim |E^x_\omega|^2$) contribution to the refractive index (cf. Butcher and Cotter §6.3.1).

# Lecture IV

**Lecture Notes on Nonlinear Optics**
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## Lecture 4

### The Truth of polarization densitites

So far, we have performed the analysis in a theoretical framework that has been exclusively formulated in terms of phenomenological models, such as the anharmonic oscillator and the phenomenologically introduced polarization response function of the medium. In the real world application of nonlinear optics, however, we should not restrict the theory just to phenomenological models, but rather take advantage over the full quantum-mechanical framework of analysis of interaction between light and matter.

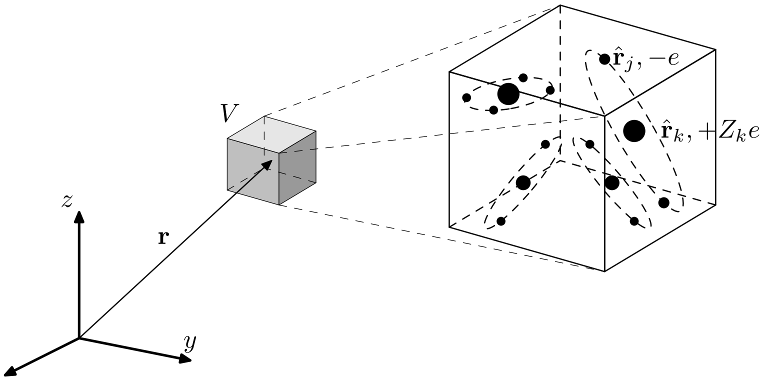

Figure 1. Schematic figure of the ensemble in the "small volume".

In a small volume $V$ (smaller than the wavelength of the light, to ensure that the natural spatial variation of the light is not taken into account, but large enough in order to contain a sufficcient number of molecules in order to ignore the quantum-mechanical fluctuations of the dipole moment density), we consider the applied electric field to be homogeneous, and the electric polarization density of the medium is then given as the expectation value of the *electric dipole operator of the ensemble of molecules* divided by the volume, as

$$P_\mu(\mathbf{r}, t) = \langle \hat{Q}_\mu \rangle / V,$$

where the electric dipole operator of the ensemble contained in $V$ can be written as a sum over all electrons and nuclei as

$$\hat{\mathbf{Q}} = \underbrace{-e \sum_j \hat{\mathbf{r}}_j}_{\text{electrons}} + \underbrace{e \sum_k Z_k \hat{\mathbf{r}}_k}_{\text{nuclei}} .$$

The expectation value $\langle \hat{Q}_\mu \rangle$ can in principle be calculated directly from the compound, time-dependent wave function of the ensemble of molecules in the small volume, considering any kind of interaction between the molecules, which may be of an arbitrary composition. However, we will here describe the interactions that take place in terms of the *quantum mechanical density operator* of the ensemble, in which case the expectation value is calculated from the *quantum mechanical trace* as

$$P_\mu(\mathbf{r}, t) = \mathrm{Tr}[\hat{\rho}(t) \hat{Q}_\mu] / V.$$

## Outline

Previously, in lecture one, we applied the mathematical tool of perturbation analysis to a classical mechanical model of the dipole moment. This analysis will now essentially be repeated, but now we will instead consider a perturbation series for the quantum mechanical density operator, with the series being of the form

$$\hat{\rho}(t) = \underbrace{\hat{\rho}_0}_{\sim[E(t)]^0} + \underbrace{\hat{\rho}_1(t)}_{\sim[E(t)]^1} + \underbrace{\hat{\rho}_2(t)}_{\sim[E(t)]^2} + \ldots + \underbrace{\hat{\rho}_n(t)}_{\sim[E(t)]^n} + \ldots$$

As this perturbation series is inserted into the expression for the electric polarization density, we will obtain a resulting series for the polarization density as

$$P_\mu(\mathbf{r},t) = \sum_{m=0}^{\infty} \underbrace{\mathrm{Tr}[\hat{\rho}_m(t)\hat{Q}_\mu]/V}_{=P_\mu^{(m)}(\mathbf{r},t)} \approx \sum_{m=0}^{n} P_\mu^{(m)}(\mathbf{r},t).$$

## Quantum mechanics

We consider an ensemble of molecules, where each molecule may be different from the other molecules of the ensemble, as well as being affected by some mutual interaction between the other members of the ensemble. The Hamiltonian for this ensemble is generally taken as

$$\hat{H} = \hat{H}_0 + \hat{H}_\mathrm{I}(t),$$

where $\hat{H}_0$ is the Hamiltonian at thermal equilibrium, with no external forces present, and $\hat{H}_\mathrm{I}(t)$ is the interaction Hamiltonial (in the Schrödinger picture), which for electric dipolar interactions take the form:

$$\hat{H}_\mathrm{I}(t) = -\hat{\mathbf{Q}}\cdot\mathbf{E}(\mathbf{r},t) = -\hat{Q}_\alpha E_\alpha(\mathbf{r},t),$$

where $\hat{\mathbf{Q}}$ is the electric dipole operator of the *ensemble* of molecules contained in the small volume $V$ (see Fig. 1). This expression may be compared with the all-classical electrostatic energy of an electric dipole moment in a electric field, $V = -\mathbf{p}\cdot\mathbf{E}(\mathbf{r},t)$.

In order to provide a proper description of the interaction between light and matter at molecular level, we must be means of some quantum mechanical description evaluate all properties of the molecule, such as electric dipole moment, magnetic dipole moment, etc., by means of *quantum mechanical expectation values*.

The description that we here will apply is by means of the *density operator formalism*, with the density operator defined in terms of orthonormal set of wave functions $|a\rangle$ of the system as

$$\hat{\rho} = \sum_a p_a |a\rangle\langle a| = \hat{\rho}(t),$$

where $p_a$ are the normalized probabilities of the system to be in state $|a\rangle$, with

$$\sum_a p_a = 1.$$

From the density operator, the expectation value of any arbitrary quantum mechanical operator $\hat{O}$ of the ensemble is obtained from the *quantum mechanical trace* as

$$\langle\hat{O}\rangle = \mathrm{Tr}(\hat{\rho}\hat{O}) = \sum_k \langle k|\hat{\rho}\hat{O}|k\rangle.$$

The equation of motion for the density operator is given in terms of the Hamiltonian as

$$\begin{aligned} i\hbar\frac{d\hat{\rho}}{dt} &= [\hat{H},\hat{\rho}] = \hat{H}\hat{\rho} - \hat{\rho}\hat{H} \\ &= [\hat{H}_0,\hat{\rho}] + [\hat{H}_\mathrm{I}(t),\hat{\rho}] \end{aligned} \qquad (1)$$

In this context, the terminology of "equation of motion" can be pictured as

$$\left\{\begin{array}{c}\text{A change of the density}\\ \text{operator } \hat{\rho}(t) \text{ in time}\end{array}\right\} \quad\Leftrightarrow\quad \left\{\begin{array}{c}\text{A change of density}\\ \text{of states in time}\end{array}\right\} \quad\Leftrightarrow\quad \left\{\begin{array}{c}\text{Change of a general}\\ \text{property } \langle\hat{O}\rangle \text{ in time}\end{array}\right\}$$

Whenever external forces are absent, that is to say, whenever the applied electromagnetic field is zero, the equation of motion for the density operator takes the form

$$i\hbar\frac{d\hat{\rho}}{dt} = [\hat{H}_0, \hat{\rho}],$$

with the solution[1]

$$\begin{aligned}\hat{\rho}(t) &= \hat{\rho}_0 = \eta\exp(-\hat{H}_0/k_{\mathrm{B}}T)\\ &\left\{= \eta\sum_{j=1}^{\infty}\frac{1}{j!}(-\hat{H}_0/k_{\mathrm{B}}T)^j\right\}\end{aligned}$$

being the time-independent density operator at thermal equilibrium, with the normalization constant $\eta$ chosen so that $\mathrm{Tr}(\hat{\rho}) = 1$, i. e.,

$$\eta = 1/\mathrm{Tr}[\exp(-\hat{H}_0/k_{\mathrm{B}}T)].$$

**Perturbation analysis of the density operator**

The task is now o obtain a solution of the equation of motion (1) by means of a perturbation series, in similar to the analysis performed for the anharmonic oscillator in the first lecture of this course. The perturbation series is, in analogy to the mechanical spring oscillator under influence of an electromagnetic field, taken as

$$\hat{\rho}(t) = \underbrace{\hat{\rho}_0}_{\sim[E(t)]^0} + \underbrace{\hat{\rho}_1(t)}_{\sim[E(t)]^1} + \underbrace{\hat{\rho}_2(t)}_{\sim[E(t)]^2} + \ldots + \underbrace{\hat{\rho}_n(t)}_{\sim[E(t)]^n} + \ldots$$

The boundary condition of the perturbation series is taken as the initial condition that sometime in the past, the external forces has been absent, i. e.

$$\hat{\rho}(-\infty) = \hat{\rho}_0,$$

which, since the perturbation series is to be valid for *all possible evolutions in time of the externally applied electric field*, leads to the boundary conditions for each individual term of the perturbation series as

$$\hat{\rho}_j(-\infty) = 0, \qquad j = 1, 2, \ldots$$

By inserting the perturbation series for the density operator into the equation of motion (1), one hence obtains

$$\begin{aligned}i\hbar\frac{d}{dt}(\hat{\rho}_0 + \hat{\rho}_1(t) + \hat{\rho}_2(t) + \ldots + \hat{\rho}_n(t) + \ldots) &= [\hat{H}_0, \hat{\rho}_0 + \hat{\rho}_1(t) + \hat{\rho}_2(t) + \ldots + \hat{\rho}_n(t) + \ldots]\\ &\quad + [\hat{H}_{\mathrm{I}}(t), \hat{\rho}_0 + \hat{\rho}_1(t) + \hat{\rho}_2(t) + \ldots + \hat{\rho}_n(t) + \ldots],\end{aligned}$$

---

[1] For any macroscopic system, the probability that the system is in a particular energy eigenstate $\psi_n$, with associated energy $\mathbb{E}_n$, is given by the familiar Boltzmann distribution

$$p_n = \eta\exp(-\mathbb{E}_n/k_{\mathrm{B}}T),$$

where $\eta$ is a normalization constant chosen so that $\sum_n p_n = 1$, $k_{\mathrm{B}}$ is the Boltzmann constant, and $T$ the absolute temperature. This probability distribution is in this course to be considered as being an axiomatic fact, and the origin of this probability distribution can readily be obtained from textbooks on thermodynamics or statistical mechanics.

and by equating terms with equal power dependence of the applied electric field in the right and left hand sides, one obtains the system of equations

$$\begin{aligned}
i\hbar\frac{d\hat{\rho}_0}{dt} &= [\hat{H}_0, \hat{\rho}_0], \\
i\hbar\frac{d\hat{\rho}_1(t)}{dt} &= [\hat{H}_0, \hat{\rho}_1(t)] + [\hat{H}_{\mathrm{I}}(t), \hat{\rho}_0], \\
i\hbar\frac{d\hat{\rho}_2(t)}{dt} &= [\hat{H}_0, \hat{\rho}_2(t)] + [\hat{H}_{\mathrm{I}}(t), \hat{\rho}_1(t)], \\
&\vdots \\
i\hbar\frac{d\hat{\rho}_n(t)}{dt} &= [\hat{H}_0, \hat{\rho}_n(t)] + [\hat{H}_{\mathrm{I}}(t), \hat{\rho}_{n-1}(t)], \\
&\vdots
\end{aligned} \tag{2}$$

for the variuos order terms of the perturbation series. In Eq. (2), we may immediately notice that the first equation simply is the identity stating the thermal equilibrium condition for the zeroth order term $\hat{\rho}_0$, while all other terms may be obtained by consecutively solve the equations of order $j = 1, 2, \ldots, n$, in that order.

**The interaction picture**

We will now turn our attention to the problem of actually solving the obtained system of equations for the terms of the perturbation series for the density operator. In a classical picture, the obtained equations are all of the form similar to

$$\frac{d\rho}{dt} = f(t)\rho + g(t), \tag{3}$$

for known functions $f(t)$ and $g(t)$. To solve these equations, we generally look for an integrating factor $I(t)$ satisfying

$$I(t)\frac{d\rho}{dt} - I(t)f(t)\rho = \frac{d}{dt}[I(t)\rho]. \tag{4}$$

By carrying out the differentiation in the right hand side of the equation, we find that the integrating factor should satisfy

$$\frac{dI(t)}{dt} = -I(t)f(t),$$

which is solved by[2]

$$I(t) = I(0)\exp\big[-\int_0^t f(\tau)\,d\tau\big].$$

The original ordinary differential equation (3) is hence solved by multiplying with the intagrating factor $I(t)$ and using the property (4) of the integrating factor, giving the equation

$$\frac{d}{dt}[I(t)\rho] = I(t)g(t),$$

from which we hence obtain the solution for $\rho(t)$ as

$$\rho(t) = \frac{1}{I(t)}\int_0^t I(\tau)g(\tau)\,d\tau.$$

From this preliminary discussion we may anticipate that equations of motion for the various order perturbation terms of the density operator can be solved in a similar manner, using integrating

---

[2] Butcher and Cotter have in their classical description of integrating factors chosen to put $I(0) = 1$.

factors. However, it should be kept in mind that we here are dealing with *operators* and not classical quantities, and since we do not know if the integrating factor is to be multiplied from left or right.

In order not to loose any generality, we may look for a set of two integrating factors $\hat{V}_0(t)$ and $\hat{U}_0(t)$, in operator sense, that we left and right multiply the unknown terms of the $n$th order equation by, and we require these operators to have the effective impact

$$\hat{V}_0(t)\left\{i\hbar\frac{d\hat{\rho}_n(t)}{dt}-[\hat{H}_0,\hat{\rho}_n(t)]\right\}\hat{U}_0(t)=i\hbar\frac{d}{dt}[\hat{V}_0(t)\hat{\rho}_n(t)\hat{U}_0(t)]. \tag{5}$$

By carrying out the differentiation in the right-hand side, expanding the commutator in the left hand side, and rearranging terms, one then obtains the equation

$$\left\{i\hbar\frac{d}{dt}\hat{V}_0(t)+\hat{V}_0(t)\hat{H}_0\right\}\hat{\rho}_n(t)\hat{U}_0(t)+\hat{V}_0(t)\hat{\rho}_n(t)\left\{i\hbar\frac{d}{dt}\hat{U}_0(t)-\hat{H}_0\hat{U}_0(t)\right\}=0$$

for the operators $\hat{V}_0(t)$ and $\hat{U}_0(t)$. This equation clearly is satisfied if both of the braced expressions simultaneously are zero for all times, in other words, if the so-called *time-development operators* $\hat{V}_0(t)$ and $\hat{U}_0(t)$ are chosen to satisfy

$$\begin{aligned}i\hbar\frac{d\hat{V}_0(t)}{dt}+\hat{V}_0(t)\hat{H}_0&=0,\\ i\hbar\frac{d\hat{U}_0(t)}{dt}-\hat{H}_0\hat{U}_0(t)&=0,\end{aligned}$$

with solutions

$$\begin{aligned}\hat{U}_0(t)&=\exp(-i\hat{H}_0t/\hbar),\\ \hat{V}_0(t)&=\exp(i\hat{H}_0t/\hbar)=\hat{U}_0(-t).\end{aligned}$$

In these expressions, the exponentials are to be regarded as being defined by their series expansion. In particular, each term of the series expansion contains an operator part being a power of the thermal equilibrium Hamiltonian $\hat{H}_0$, which commute with any of the other powers. We may easily verify that the obtained solutions, in a strict operator sense, satisfy the relations

$$\hat{U}_0(t)\hat{U}_0(t')=\hat{U}_0(t+t'),$$

with, in particular, the corollary

$$\hat{U}_0(t)\hat{U}_0(-t)=\hat{U}_0(0)=1.$$

Let us now again turn our attention to the original equation of motion that was the starting point for this discussion. By multiplying the $n$th order subequation of Eq. (2) with $\hat{U}_0(-t)$ from the left, and multiplying with $\hat{U}_0(t)$ from the right, we by using the relation (5) obtain

$$i\hbar\frac{d}{dt}\left\{\hat{U}_0(-t)\hat{\rho}_n(t)\hat{U}_0(t)\right\}=\hat{U}_0(-t)[\hat{H}_{\text{I}}(t),\hat{\rho}_{n-1}(t)]\hat{U}_0(t),$$

which is integrated to yield the solution

$$\hat{U}_0(-t)\hat{\rho}_n(t)\hat{U}_0(t)=\frac{1}{i\hbar}\int_{-\infty}^{t}\hat{U}_0(-\tau)[\hat{H}_{\text{I}}(\tau),\hat{\rho}_{n-1}(\tau)]\hat{U}_0(\tau)\,d\tau,$$

where the lower limit of integration was fixed in accordance with the initial condition $\hat{\rho}_n(-\infty)=0$, $n=1,2,\ldots$. In some sense, we may consider the obtained solution as being the end point of this discussion; however, we may simplify the expression somewhat by making a few notes on the properties of the time development operators. By expanding the right hand side of the solution, and inserting $\hat{U}_0(\tau)\hat{U}_0(-\tau) = 1$ between $\hat{H}_{\rm I}(\tau)$ and $\hat{\rho}_{n-1}(\tau)$ in the two terms, we obtain

$$\begin{aligned}
\hat{U}_0(-t)\hat{\rho}_n(t)\hat{U}_0(t) &= \frac{1}{i\hbar}\int_{-\infty}^{t} \hat{U}_0(-\tau)[\hat{H}_{\rm I}(\tau)\hat{\rho}_{n-1}(\tau) - \hat{\rho}_{n-1}(\tau)\hat{H}_{\rm I}(\tau)]\hat{U}_0(\tau)\,d\tau\\
&= \frac{1}{i\hbar}\int_{-\infty}^{t} \hat{U}_0(-\tau)\hat{H}_{\rm I}(\tau)\underbrace{\hat{U}_0(\tau)\hat{U}_0(-\tau)}_{=1}\hat{\rho}_{n-1}(\tau)\hat{U}_0(\tau)\,d\tau\\
&\qquad - \frac{1}{i\hbar}\int_{-\infty}^{t} \hat{U}_0(-\tau)\hat{\rho}_{n-1}(\tau)\underbrace{\hat{U}_0(\tau)\hat{U}_0(-\tau)}_{=1}\hat{H}_{\rm I}(\tau)\hat{U}_0(\tau)\,d\tau\\
&= \frac{1}{i\hbar}\int_{-\infty}^{t} [\underbrace{\hat{U}_0(-\tau)\hat{H}_{\rm I}(\tau)\hat{U}_0(\tau)}_{\equiv\hat{H}'_{\rm I}(t)}, \underbrace{\hat{U}_0(-\tau)\hat{\rho}_{n-1}(\tau)\hat{U}_0(\tau)}_{\equiv\hat{\rho}'_{n-1}(t)}]\,d\tau,
\end{aligned}$$

and hence, by introducing the primed notation in the *interaction picture* for the quantum mechanical operators,

$$\begin{aligned}
\hat{\rho}'_n(t) &= \hat{U}_0(-t)\hat{\rho}_n(t)\hat{U}_0(t),\\
\hat{H}'_{\rm I}(t) &= \hat{U}_0(-t)\hat{H}_{\rm I}(t)\hat{U}_0(t),
\end{aligned}$$

the solutions of the system of equations for the terms of the perturbation series for the density operator *in the interaction picture* take the simplified form

$$\hat{\rho}'_n(t) = \frac{1}{i\hbar}\int_{-\infty}^{t} [\hat{H}'_{\rm I}(\tau), \hat{\rho}'_{n-1}(\tau)]\,d\tau, \qquad n = 1, 2, \ldots,$$

with the variuos order solutions expressed in the original Schrödinger picture by means of the inverse transformation

$$\hat{\rho}_n(t) = \hat{U}_0(t)\hat{\rho}'_n(t)\hat{U}_0(-t).$$

### The first order polarization density

With the quantum mechanical perturbative description of the interaction between light and matter in fresh mind, we are now in the position of formulating the polarization density of the medium from a quantum mechanical description. A minor note should though be made regarding the Hamiltonian, which now is expressed in the interaction picture, and hence the electric dipolar operator (since the electric field here is considered to be a macroscopic, classical quantity) is given in the interaction picture as well,

$$\begin{aligned}
\hat{H}'_{\rm I}(\tau) &= \hat{U}_0(-\tau)\underbrace{[-\hat{Q}_\alpha E_\alpha(\tau)]}_{=\hat{H}_{\rm I}(\tau)}\hat{U}_0(\tau)\\
&= -\hat{U}_0(-\tau)\hat{Q}_\alpha\hat{U}_0(\tau)E_\alpha(\tau)\\
&= -\hat{Q}_\alpha(\tau)E_\alpha(\tau)
\end{aligned}$$

where $\hat{Q}_\alpha(\tau)$ denotes the electric dipolar operator of the ensemble, taken *in the interaction picture.*

By inserting the expression for the first order term of the perturbation series for the density operator into the quantum mechanical trace of the first order electric polarization density of the medium, one obtains

$$\begin{aligned}
P^{(1)}_\mu(\mathbf{r},t) &= \frac{1}{V}\,\mathrm{Tr}[\hat\rho_1(t)\hat Q_\mu]\\
&= \frac{1}{V}\,\mathrm{Tr}\Big[\underbrace{\Big(\hat U_0(t)\underbrace{\frac{1}{i\hbar}\int_{-\infty}^{t}[\hat H'_{\mathrm{I}}(\tau),\hat\rho_0]\,d\tau}_{=\hat\rho'_1(t)}\,\hat U_0(-t)\Big)}_{=\hat\rho_1(t)}\hat Q_\mu\Big]\\
&= \left\{\begin{array}{c} E_\mu(\tau)\text{ is a classical field (omit space dependence }\mathbf{r}),\\ {[\hat H'_{\mathrm{I}}(\tau),\hat\rho_0] = [-\hat Q_\alpha(\tau)E_\alpha(\tau),\hat\rho_0] = -E_\alpha(\tau)[\hat Q_\alpha(\tau),\hat\rho_0]}\end{array}\right\}\\
&= -\frac{1}{Vi\hbar}\,\mathrm{Tr}\Big\{\hat U_0(t)\int_{-\infty}^{t}E_\alpha(\tau)[\hat Q_\alpha(\tau),\hat\rho_0]\,d\tau\,\hat U_0(-t)\,\hat Q_\mu\Big\}\\
&= \{\text{Pull out } E_\alpha(\tau)\text{ and the integral outside the trace}\}\\
&= -\frac{1}{Vi\hbar}\int_{-\infty}^{t}E_\alpha(\tau)\,\mathrm{Tr}\{\hat U_0(t)[\hat Q_\alpha(\tau),\hat\rho_0]\hat U_0(-t)\,\hat Q_\mu\}\,d\tau\\
&= \Big\{\text{Express } E_\alpha(\tau)\text{ in frequency domain, } E_\alpha(\tau)=\int_{-\infty}^{\infty}E_\alpha(\omega)\exp(-i\omega\tau)\,d\omega\Big\}\\
&= -\frac{1}{Vi\hbar}\int_{-\infty}^{\infty}\int_{-\infty}^{t}E_\alpha(\omega)\,\mathrm{Tr}\{\hat U_0(t)[\hat Q_\alpha(\tau),\hat\rho_0]\hat U_0(-t)\,\hat Q_\mu\}\exp(-i\omega\tau)\,d\tau\,d\omega\\
&= \{\text{Use } \exp(-i\omega\tau)=\exp(-i\omega t)\exp[-i\omega(\tau-t)]\}\\
&= -\frac{1}{Vi\hbar}\int_{-\infty}^{\infty}\int_{-\infty}^{t}E_\alpha(\omega)\,\mathrm{Tr}\{\hat U_0(t)[\hat Q_\alpha(\tau),\hat\rho_0]\hat U_0(-t)\,\hat Q_\mu\}\\
&\qquad\qquad\times\exp[-i\omega(\tau-t)]\,d\tau\,\exp(-i\omega t)\,d\omega\\
&= \varepsilon_0\int_{-\infty}^{\infty}\chi^{(1)}_{\mu\alpha}(-\omega;\omega)E_\alpha(\omega)\exp(-i\omega t)\,d\omega,
\end{aligned}$$

where the first order (linear) electric susceptibility is defined as

$$\begin{aligned}
\chi^{(1)}_{\mu\alpha}(-\omega;\omega) &= -\frac{1}{\varepsilon_0 Vi\hbar}\int_{-\infty}^{t}\mathrm{Tr}\{\underbrace{\hat U_0(t)[\hat Q_\alpha(\tau),\hat\rho_0]\hat U_0(-t)}_{=[\hat Q_\alpha(\tau-t),\hat\rho_0]}\,\hat Q_\mu\}\exp[-i\omega(\tau-t)]\,d\tau\\
&= \left\{\begin{array}{c}\text{Expand the commutator and insert } \hat U_0(-t)\hat U_0(t)\equiv 1\\ \text{in middle of each of the terms, using } [\hat U_0(t),\hat\rho_0]=0\\ \Rightarrow \hat U_0(t)[\hat Q_\alpha(\tau),\hat\rho_0]\hat U_0(-t) = [\hat Q_\alpha(\tau-t),\hat\rho_0]\end{array}\right\}\\
&= -\frac{1}{\varepsilon_0 Vi\hbar}\int_{-\infty}^{t}\mathrm{Tr}\{[\hat Q_\alpha(\tau-t),\hat\rho_0]\hat Q_\mu\}\exp[-i\omega(\tau-t)]\,d\tau\\
&= \Big\{\text{Change variable of integration } \tau'=\tau-t;\ \int_{-\infty}^{t}\cdots d\tau\to\int_{-\infty}^{0}\cdots d\tau'\Big\}\\
&= -\frac{1}{\varepsilon_0 Vi\hbar}\int_{-\infty}^{0}\underbrace{\mathrm{Tr}\{[\hat Q_\alpha(\tau'),\hat\rho_0]\hat Q_\mu\}}_{=\mathrm{Tr}\{\hat\rho_0[\hat Q_\mu,\hat Q_\alpha(\tau')]\}}\exp(-i\omega\tau')\,d\tau'\\
&= \{\text{Cyclic permutation of the arguments in the trace}\}\\
&= -\frac{1}{\varepsilon_0 Vi\hbar}\int_{-\infty}^{0}\mathrm{Tr}\{\hat\rho_0[\hat Q_\mu,\hat Q_\alpha(\tau)]\}\exp(-i\omega\tau)\,d\tau.
\end{aligned}$$

# Lecture V

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## LECTURE 5

In the previous lecture, the quantum mechanical origin of the linear and nonlinear susceptibilities was discussed. In particular, a perturbation analysis of the density operator was performed, and the resulting system of equations was solved recursively for the $n$th order density operator $\hat{\rho}_n(t)$ in terms of $\hat{\rho}_{n-1}(t)$, where the zeroth order term (independent of the applied electric field of the light) is given by the Boltzmann distribution at thermal equilibrium.

So far we have obtained the linear optical properties of the medium, in terms of the first order susceptibility tensor (of rank-two), and we will now proceed with the next order of interaction, giving the second order electric susceptibility tensor (of rank three).

### The second order polarization density

For the second order interaction, the corresponding term in the perturbation series of the density operator *in the interaction picture* becomes[1]

$$\begin{aligned}\hat{\rho}'_2(t) &= \frac{1}{i\hbar}\int_{-\infty}^{t}[\hat{H}'_{\mathrm{I}}(\tau_1),\hat{\rho}'_1(\tau_1)]\,d\tau_1\\ &= \frac{1}{i\hbar}\int_{-\infty}^{t}[\hat{H}'_{\mathrm{I}}(\tau_1),\frac{1}{i\hbar}\int_{-\infty}^{\tau_1}[\hat{H}'_{\mathrm{I}}(\tau_2),\hat{\rho}_0]\,d\tau_2\,]\,d\tau_1 \qquad (1)\\ &= \frac{1}{(i\hbar)^2}\int_{-\infty}^{t}\int_{-\infty}^{\tau_1}[\hat{H}'_{\mathrm{I}}(\tau_1),[\hat{H}'_{\mathrm{I}}(\tau_2),\hat{\rho}_0]]\,d\tau_2\,d\tau_1\end{aligned}$$

In order to simplify the expression for the second order susceptibility, we will in the following analysis make use of a generalization of the cyclic perturbation of the terms in the commutator inside the trace, as

$$\mathrm{Tr}\{[\hat{Q}_\alpha(\tau_1),[\hat{Q}_\beta(\tau_2),\hat{\rho}_0]]\hat{Q}_\mu\} = \mathrm{Tr}\{\hat{\rho}_0[[\hat{Q}_\mu,\hat{Q}_\alpha(\tau_1)],\hat{Q}_\beta(\tau_2)]\}. \qquad (2)$$

By inserting the expression for the second order term of the perturbation series for the density operator into the quantum mechanical trace of the second order electric polarization density of the medium, one obtains

$$\begin{aligned}P^{(2)}_\mu(\mathbf{r},t) &= \frac{1}{V}\,\mathrm{Tr}[\hat{\rho}_2(t)\hat{Q}_\mu]\\ &= \frac{1}{V}\,\mathrm{Tr}\Big[\underbrace{\Big(\hat{U}_0(t)\underbrace{\frac{1}{(i\hbar)^2}\int_{-\infty}^{t}\int_{-\infty}^{\tau_1}[\hat{H}'_{\mathrm{I}}(\tau_1),[\hat{H}'_{\mathrm{I}}(\tau_2),\hat{\rho}_0]]\,d\tau_2\,d\tau_1}_{=\hat{\rho}'_2(t)\quad\text{(interaction picture)}}\hat{U}_0(-t)\Big)}_{=\hat{\rho}_2(t)\quad\text{(Schrödinger picture)}}\hat{Q}_\mu\Big]\\ &= \{E_\alpha(\tau_1)\text{ and }E_\alpha(\tau_1)\text{ are classical fields (omit space dependence }\mathbf{r})\}\\ &= \frac{1}{V(i\hbar)^2}\,\mathrm{Tr}\Big\{\hat{U}_0(t)\int_{-\infty}^{t}\int_{-\infty}^{\tau_1}[\hat{Q}_\alpha(\tau_1),[\hat{Q}_\beta(\tau_2),\hat{\rho}_0]]E_\alpha(\tau_1)E_\beta(\tau_2)\,d\tau_2\,d\tau_1\hat{U}_0(-t)\,\hat{Q}_\mu\Big\}\\ &= \{\text{Pull out }E_{\alpha_1}(\tau_1)E_{\alpha_2}(\tau_2)\text{ and the integrals outside the trace}\}\end{aligned}$$

---

[1] It should be noticed that the form given in Eq. (1) not only applies to an ensemble of molecules, of arbitrary composition, but also to *any* kind of level of approximation for the interaction, such as the inclusion of magnetic dipolar interactions or electric quadrupolar interactions as well. These interactions should (of course) be incorporated in the expression for the interaction Hamiltonian $\hat{H}'_{\mathrm{I}}(\tau)$, here described in the interaction picture.

$$= \frac{1}{V(i\hbar)^2}\int_{-\infty}^{t}\int_{-\infty}^{\tau_1}\mathrm{Tr}\left\{\hat{U}_0(t)[\hat{Q}_\alpha(\tau_1),[\hat{Q}_\beta(\tau_2),\hat{\rho}_0]]\hat{U}_0(-t)\,\hat{Q}_\mu\right\}E_\alpha(\tau_1)E_\beta(\tau_2)\,d\tau_2\,d\tau_1.$$

In analogy with the results as obtained for the first order (linear) optical properties, now express the term $E_{\alpha_1}(\tau_1)E_{\alpha_2}(\tau_2)$ in the frequency domain, by using the Fourier identity

$$E_{\alpha_k}(\tau_k) = \int_{-\infty}^{\infty} E_{\alpha_k}(\omega_k)\exp(-i\omega\tau_k)\,d\omega,$$

which hence gives the second order polarization density expressed in terms of the electric field in the frequency domain as

$$\begin{aligned}
P_\mu^{(2)}(\mathbf{r},t) &= \frac{1}{V(i\hbar)^2}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{t}\int_{-\infty}^{\tau_1}\mathrm{Tr}\left\{\hat{U}_0(t)[\hat{Q}_\alpha(\tau_1),[\hat{Q}_\beta(\tau_2),\hat{\rho}_0]]\hat{U}_0(-t)\,\hat{Q}_\mu\right\}E_\alpha(\omega_1)E_\beta(\omega_2)\\
&\qquad\times\exp(-i\omega_1\tau_1)\exp(-i\omega_2\tau_2)\,d\tau_2\,d\tau_1\,d\omega_2\,d\omega_1\\
&= \{\text{Use }\exp(-i\omega\tau)=\exp(-i\omega t)\exp[-i\omega(\tau-t)]\}\\
&= \frac{1}{V(i\hbar)^2}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{t}\int_{-\infty}^{\tau_1}\mathrm{Tr}\left\{\hat{U}_0(t)[\hat{Q}_\alpha(\tau_1),[\hat{Q}_\beta(\tau_2),\hat{\rho}_0]]\hat{U}_0(-t)\,\hat{Q}_\mu\right\}E_\alpha(\omega_1)E_\beta(\omega_2)\\
&\qquad\times\exp[-i\omega_1(\tau_1-t)-i\omega_2(\tau_2-t)]\,d\tau_2\,d\tau_1\,\exp[-i\underbrace{(\omega_1+\omega_2)}_{=\omega_\sigma}t]\,d\omega_2\,d\omega_1\\
&= \varepsilon_0\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\chi^{(2)}_{\mu\alpha\beta}(-\omega_\sigma;\omega_1,\omega_2)E_\alpha(\omega_1)E_\beta(\omega_2)\exp(-i\omega_\sigma t)\,d\omega_2\,d\omega_1,
\end{aligned}$$

where the second order (quadratic) electric susceptibility is defined as

$$\begin{aligned}
&\chi^{(2)}_{\mu\alpha\beta}(-\omega_\sigma;\omega_1,\omega_2)\\
&\quad= \frac{1}{\varepsilon_0 V(i\hbar)^2}\int_{-\infty}^{t}\int_{-\infty}^{\tau_1}\mathrm{Tr}\left\{\hat{U}_0(t)[\hat{Q}_\alpha(\tau_1),[\hat{Q}_\beta(\tau_2),\hat{\rho}_0]]\hat{U}_0(-t)\,\hat{Q}_\mu\right\}\\
&\qquad\times\exp[-i\omega_1(\tau_1-t)-i\omega_2(\tau_2-t)]\,d\tau_2\,d\tau_1\\
&\quad= \{\text{Make use of Eq. (2) and take }\tau_1'=\tau_1-t\}\\
&\quad= \ldots\\
&\quad= \frac{1}{\varepsilon_0 V(i\hbar)^2}\int_{-\infty}^{0}\int_{-\infty}^{\tau_1'}\mathrm{Tr}\{\hat{\rho}_0[[\hat{Q}_\mu,\hat{Q}_\alpha(\tau_1')],\hat{Q}_\beta(\tau_2')]\}\exp[-i(\omega_1\tau_1'+\omega_2\tau_2')]\,d\tau_2'\,d\tau_1'.
\end{aligned}$$

This obtained expression for the second order electric susceptibility does not possess the property of intrinsic permutation symmetry. However, by using the same arguments as discussed in the analysis of the polarization response functions in lecture two, we can easily verify that this tensor can be cast into a symmetric and antisymmetric part as

$$\begin{aligned}
\chi^{(2)}_{\mu\alpha\beta}(-\omega_\sigma;\omega_1,\omega_2) &= \frac{1}{2}\underbrace{[\chi^{(2)}_{\mu\alpha\beta}(-\omega_\sigma;\omega_1,\omega_2)+\chi^{(2)}_{\mu\beta\alpha}(-\omega_\sigma;\omega_2,\omega_1)]}_{\text{symmetric part}}\\
&\qquad+\frac{1}{2}\underbrace{[\chi^{(2)}_{\mu\alpha\beta}(-\omega_\sigma;\omega_1,\omega_2)-\chi^{(2)}_{\mu\beta\alpha}(-\omega_\sigma;\omega_2,\omega_1)]}_{\text{antisymmetric part}},
\end{aligned}$$

and since the antisymmetric part, again following the arguments for the second order polarization response function, does not contribute to the polarization density, it is customary (in the Butcher and Cotter convention as well as all other conventions in nonlinear optics) to cast the second order susceptibility into the form

$$\begin{aligned}
&\chi^{(2)}_{\mu\alpha\beta}(-\omega_\sigma;\omega_1,\omega_2)\\
&\quad= \frac{1}{\varepsilon_0 V(i\hbar)^2}\frac{1}{2!}\mathbf{S}\int_{-\infty}^{0}\int_{-\infty}^{\tau_1'}\mathrm{Tr}\{\hat{\rho}_0[[\hat{Q}_\mu,\hat{Q}_\alpha(\tau_1')],\hat{Q}_\beta(\tau_2')]\}\exp[-i(\omega_1\tau_1'+\omega_2\tau_2')]\,d\tau_2'\,d\tau_1',
\end{aligned}$$

where **S**, commonly called the *symmetrizing operator*, denotes that the expression that follows is to be summed over the $2! = 2$ possible pairwise permutations of $(\alpha, \omega_1)$ and $(\beta, \omega_2)$, hence ensuring that the second order susceptibility possesses the intrinsic permutation symmetry,

$$\chi^{(2)}_{\mu\alpha\beta}(-\omega_\sigma; \omega_1, \omega_2) = \chi^{(2)}_{\mu\beta\alpha}(-\omega_\sigma; \omega_2, \omega_1).$$

**Higher order polarization densities**

The previously described principle of deriving the susceptibilities of first and second order are straightforward to extend to the $n$th order interaction. In this case, we will make use of the following generalization of Eq. (2),

$$\begin{aligned}&\mathrm{Tr}\{[\hat{Q}_{\alpha_1}(\tau_1), [\hat{Q}_{\alpha_2}(\tau_2), \ldots, [\hat{Q}_{\alpha_n}(\tau_n), \hat{\rho}_0]] \ldots]\hat{Q}_\mu\}\\ &\qquad = \mathrm{Tr}\{\hat{\rho}_0[\ldots[[\hat{Q}_\mu, \hat{Q}_{\alpha_1}(\tau_1)], \hat{Q}_{\alpha_2}(\tau_2)], \ldots \hat{Q}_{\alpha_n}(\tau_n)]\},\end{aligned}$$

which, when applied in the evaluation of the expectation value of the electric dipole operator of the ensemble, gives the $n$th order electric susceptibility as

$$\begin{aligned}&\chi^{(n)}_{\mu\alpha_1\cdots\alpha_n}(-\omega_\sigma; \omega_1, \ldots, \omega_n)\\ &\quad = \frac{1}{\varepsilon_0 V(-i\hbar)^n}\frac{1}{n!}\mathbf{S}\int_{-\infty}^{0}\int_{-\infty}^{\tau_1}\cdots\int_{-\infty}^{\tau_{n-1}} \mathrm{Tr}\{\hat{\rho}_0[\ldots[[\hat{Q}_\mu, \hat{Q}_{\alpha_1}(\tau_1)], \hat{Q}_{\alpha_2}(\tau_2)], \ldots \hat{Q}_{\alpha_n}(\tau_n)]\}\\ &\qquad\qquad \times \exp[-i(\omega_1\tau_1 + \omega_2\tau_2 + \ldots + \omega_n\tau_n)]\, d\tau_n \cdots d\tau_2\, d\tau_1,\end{aligned}$$

where now the symmetrizing operator **S** indicates that the expression following it should be summed over all the $n!$ pairwise permutations of $(\alpha_1, \omega_1), \ldots, (\alpha_n, \omega_n)$.

It should be emphasized the symmetrizing operator **S** always implies summation over *all* the $n!$ pairwise permutations of $(\alpha_1, \omega_1), \ldots, (\alpha_n, \omega_n)$, *irregardless of whether the permutations are distinct or not*. This is due to that eventually occuring degenerate permutations are taken care of in the degeneracy coefficient $K(-\omega_\sigma; \omega_1, \ldots, \omega_n)$ in Butcher and Cotters convention, as described in lecture three and in additional notes that has been handed out in lecture four.

# Lecture VI

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## LECTURE 6

### Assembly of independent molecules

So far the description of the interaction between light and matter has been in a very general form, where it was assumed merely that the interaction is local, and in the electric dipolar approximation, where magnetic dipolar and electric quadrupolar interactions (as well as higher order terms) were neglected. The ensemble of molecules has so far no constraints in terms of composition or mutual interaction, and the electric dipolar operator is so far taken for the *whole ensemble of molecules*, rather than as the dipole operator for the individual molecules.

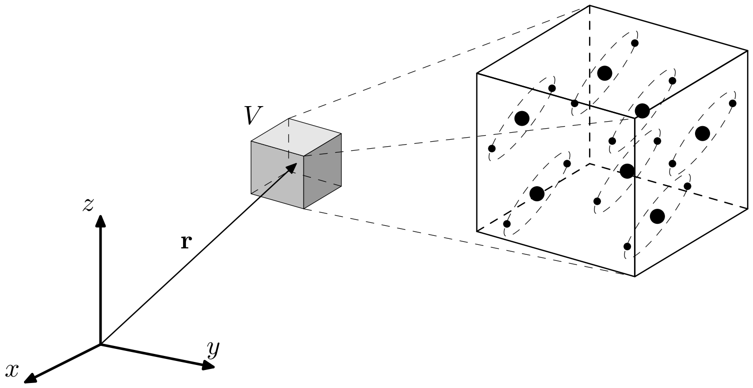

Figure 1. The ensemble of identical, similarly oriented, and mutually independent molecules.

For many practical applications, however, the obtained general form is somewhat inconvenient when it comes to the numerical evaluation of the susceptibilities, since tables of wave functions, transition frequencies and their corresponding electric dipole moments etc. often are tabulated exclusively for the individual molecules themselves. Thus, we will now apply the obtained general theory to an ensemble of identical molecules, making a transition from the wave function and electric dipole operator of the whole ensemble to the wave function and electric dipole operator of the individual molecule, and find an explicit form of the electric susceptibilities in terms of the *quantum mechanical matrix elements of the molecular dipole operator*.

We will now apply three assumptions of the ensemble of molecules, which introduce a significant simplification to the task of expressing the susceptibilities and macroscopic polarization density in terms of the individual molecular properties:

1. The molecules of the ensemble are all identical.
2. The molecules of the ensemble are mutually non-interacting.
3. The molecules of the ensemble are identically oriented.

From a wave functional approach, it is straightforward to make the transition from the description of the ensemble down to the molecular level in a strict quantum mechanical perspective (as shown in Butcher and Cotters book). However, from a practical engineering point of view, the results are quite intuitively derived if we make the following observations, which immediately follow from the assumptions listed above:

- The positions of the individual molecules does not affect the electric dipole moment of the whole ensemble in $V$, if we neglect the mutual interaction between the molecules. (This holds only if the molecules are neutrally charged, since they otherwise could build up a total electric dipole moment of the ensemble.)

- Since the mutual interaction between the molecules is neglected, we may just as well consider the individual molecules as constituting a set of "sub-ensembles" of the general ensemble picture. In this picture, all quantum mechanical expectation values, involved commutators, matrix elements, etc., should be considered for each subensemble instead, and the macroscopic polarization density will in this case become

$$\mathbf{P}(\mathbf{r},t)=\frac{\left\{\begin{matrix}\text{the number of molecules}\\ \text{within the volume } V\end{matrix}\right\}\times\left\{\begin{matrix}\text{the expectation value of the}\\ \text{molecular dipole operator}\end{matrix}\right\}}{\{\text{the volume } V\}}.$$

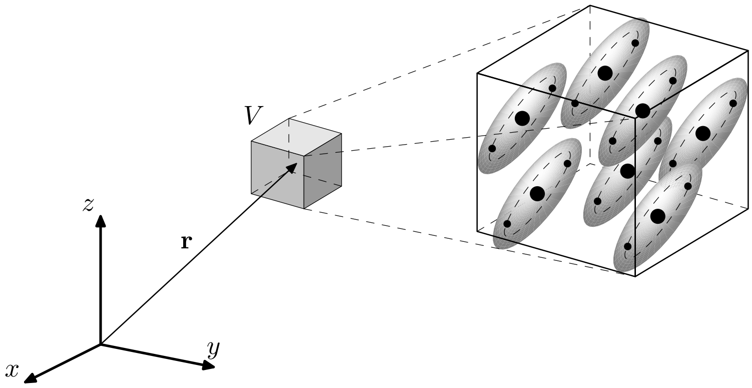

Figure 2. The general ensemble seen as an ensemble of identical mono-molecular sub-ensembles.

Assuming there are $M$ mutually non-interacting and similarly oriented molecules in the volume $V$, the macroscopic polarization density of the medium can be written as

$$\begin{aligned}
\mathbf{P}(\mathbf{r},t)&=\frac{1}{V}\langle\hat{\mathbf{Q}}\rangle=\frac{1}{V}\langle\underbrace{-e\sum_j\hat{\mathbf{r}}_j}_{\text{electrons}}+\underbrace{e\sum_k Z_k\hat{\mathbf{r}}_k}_{\text{nuclei}}\rangle\\
&=\{\text{arrange terms as sum over the molecules of the ensemble}\}\\
&=\frac{1}{V}\langle\underbrace{\sum_{m=1}^{M}\underbrace{\Big(\underbrace{-e\sum_j\hat{\mathbf{r}}_j^{(m)}}_{\text{electrons}}+\underbrace{e\sum_k Z_k^{(m)}\hat{\mathbf{r}}_k^{(m)}}_{\text{nuclei}}\Big)}_{\text{molecular elec. dipole operator, } e\hat{\mathbf{r}}^{(m)}}}_{\text{electric dipole moment of ensemble}}\rangle\\
&=\frac{1}{V}\langle\sum_{m=1}^{M}e\hat{\mathbf{r}}^{(m)}\rangle\\
&=\{\text{the molecules are mutually noninteracting}\}\\
&=\frac{1}{V}\sum_{m=1}^{M}\langle e\hat{\mathbf{r}}^{(m)}\rangle\\
&=\{\text{the molecules are identical and similarly oriented}\}\\
&=\frac{1}{V}\sum_{m=1}^{M}\langle e\hat{\mathbf{r}}\rangle\\
&=N\langle e\hat{\mathbf{r}}\rangle
\end{aligned}$$

where $N = M/V$ is the number of molecules per unit volume, and

$$\langle e\hat{\mathbf{r}}\rangle = \mathrm{Tr}[\hat{\varrho}(t)e\hat{\mathbf{r}}]$$

is the expectation value of the *mono-molecular* electric dipole operator, with $\hat{\varrho}(t)$ (that is to say, $\hat{\rho}(t)$ with a "kink"[1] to indicate the difference to the density operator of the general ensemble) is the molecular density operator. Notice that the form $\mathbf{P}(\mathbf{r},t) = N\langle e\hat{\mathbf{r}}\rangle$ is identical to the previous form for the general ensemble, though with the factor $1/V$ replaced by $N = M/V$, and with the electric dipole operator $\hat{Q}_\alpha$ of the ensemble replaced by the molecular dipole moment operator $e\hat{r}_\alpha$.

When making this transition, the condition that the molecules are mutually independent is simply a statement that we *locally* assume the superposition principle of the properties of the molecules (wave functions, electric dipole moments, etc.) to hold.

Going in the limit of non-interacting molecules, we may picture the situation as in Fig. 2, with each molecule defining a sub-ensemble, which we are free to choose as our "small volume" of charged particles. As long as we do not make any claim to determine the exact individual positions of the charged particles together with their respective momentum (or any other pair of canonical variables which would violate the Heisenberg uncertainty relation), this is a perfectly valid picture, which provides the statistical expectation values of any observable property of the medium as $M/V$ times the average statistical molecular observations.

The previously described operators of a general ensemble of charged particles should in this case be replaced by their corresponding mono-molecular equivalents, as listed in the following table.

| General ensemble | | Molecular representation | |
|---|---|---|---|
| $\hat{Q}_\mu$ | → | $e\hat{r}_\mu$ | (Electric dipole operator) |
| $\hat{\rho}_0$ | → | $\hat{\varrho}_0$ | (Density operator of thermal equilibrium) |
| $\hat{\rho}_n(t)$ | → | $\hat{\varrho}_n(t)$ | ($n$th order term of density operator) |
| $\hat{H}_0$ | → | $\hat{\mathcal{H}}_0$ | (Hamiltonian of thermal equilibrium) |
| $1/V$ | → | $N = M/V$ | (Number density of molecules) |
| $\rho_0(a)$ | → | $\varrho_0(a)$ | (Molecular population density at state $\|a\rangle$) |

Whenever an operator is expressed in the interaction picture, we should keep in mind that the corresponding time development operators $\hat{U}_0(t)$, which originally were expressed in terms of the thermal equilibrium Hamiltonian $\hat{H}_0$ of the ensemble, now should be expressed in terms of the mono-molecular thermal equilibrium Hamiltonian $\hat{\mathcal{H}}_0$.

The matrix elements of the mono-molecular electric dipole operator, taken in the interaction picture, become

$$\begin{aligned}
\langle a|e\hat{r}_\alpha(t)|b\rangle &= \langle a|\hat{U}_0(-t)e\hat{r}_\alpha\hat{U}_0(t)|b\rangle \\
&= \{\text{the closure principle, B.\&C. Eq. (3.11)}, [\hat{O}\hat{O}']_{ab} = \sum_k O_{ak}O_{kb}\} \\
&= \sum_k\sum_l \langle a|\hat{U}_0(-t)|k\rangle\langle k|e\hat{r}_\alpha|l\rangle\langle l|\hat{U}_0(t)|b\rangle \\
&= \{\text{energy representation, B.\&C. Eq. (4.54)}, [\hat{U}_0(t)]_{ab} = \exp(-i\mathbb{E}_a t/\hbar)\delta_{ab}\} \\
&= \sum_k\sum_l \exp(-i\mathbb{E}_a(-t)/\hbar)\delta_{ak}\langle k|e\hat{r}_\alpha|l\rangle\exp(-i\mathbb{E}_l t/\hbar)\delta_{lb} \\
&= \langle a|e\hat{r}_\alpha|b\rangle\exp[i\,\underbrace{(\mathbb{E}_a-\mathbb{E}_b)t/\hbar}_{\equiv\Omega_{ab}t}] \\
&= \langle a|e\hat{r}_\alpha|b\rangle\exp(i\Omega_{ab}t),
\end{aligned} \tag{3}$$

---

[1] **kink** *n.* **1.** a sharp twist or bend in a wire, rope, hair, etc. [*Collins Concise Dictionary*, Harper-Collins (1995)].

where $\Omega_{ab} = (\mathbb{E}_a - \mathbb{E}_b)/\hbar$ is the molecular transition frequency between the molecular states $|a\rangle$ and $|b\rangle$. We may thus, at least in some sense, interpret the interaction picture for the matrix elements of the molecular operators as an out-separation of the naturally occurring transition frequencies associated with a change from state $|a\rangle$ to $|b\rangle$ of the molecule.

*First order electric susceptibility*

From the general form of the first order (linear) electric susceptibility, as derived in the previous lecture, one obtains

$$\begin{aligned}
\chi^{(1)}_{\mu\alpha}(-\omega;\omega) &= -\frac{N}{\varepsilon_0 i\hbar}\int_{-\infty}^{0} \mathrm{Tr}\{\hat{\varrho}_0[e\hat{r}_\mu, e\hat{r}_\alpha(\tau)]\}\exp(-i\omega\tau)\,d\tau \\
&= -\frac{Ne^2}{\varepsilon_0 i\hbar}\int_{-\infty}^{0} \sum_a \langle a|\hat{\varrho}_0(\hat{r}_\mu\hat{r}_\alpha(\tau) - \hat{r}_\alpha(\tau)\hat{r}_\mu)|a\rangle \exp(-i\omega\tau)\,d\tau \\
&= \{\text{the closure principle}\} \\
&= -\frac{Ne^2}{\varepsilon_0 i\hbar}\int_{-\infty}^{0} \sum_a\sum_k \langle a|\hat{\varrho}_0|k\rangle\langle k|\hat{r}_\mu\hat{r}_\alpha(\tau) - \hat{r}_\alpha(\tau)\hat{r}_\mu|a\rangle \exp(-i\omega\tau)\,d\tau \\
&= \{\text{use that } \langle a|\hat{\varrho}_0|k\rangle = \varrho_0(a)\delta_{ak}\} \\
&= -\frac{Ne^2}{\varepsilon_0 i\hbar}\int_{-\infty}^{0} \sum_a \varrho_0(a)\big[\langle a|\hat{r}_\mu\hat{r}_\alpha(\tau)|a\rangle - \langle a|\hat{r}_\alpha(\tau)\hat{r}_\mu|a\rangle\big] \exp(-i\omega\tau)\,d\tau \\
&= \{\text{the closure principle again}\} \\
&= -\frac{Ne^2}{\varepsilon_0 i\hbar}\int_{-\infty}^{0} \sum_a \varrho_0(a)\sum_b \big[\langle a|\hat{r}_\mu|b\rangle\langle b|\hat{r}_\alpha(\tau)|a\rangle - \langle a|\hat{r}_\alpha(\tau)|b\rangle\langle b|\hat{r}_\mu|a\rangle\big] \exp(-i\omega\tau)\,d\tau \\
&= \{\text{Use results of Eq. (3) and shorthand notation } \langle a|\hat{r}_\alpha|b\rangle = r^\alpha_{ab}\} \\
&= -\frac{Ne^2}{\varepsilon_0 i\hbar}\sum_a \varrho_0(a)\sum_b \int_{-\infty}^{0} \big[r^\mu_{ab}r^\alpha_{ba}\exp(i\Omega_{ba}\tau) - r^\alpha_{ab}r^\mu_{ba}\exp(-i\Omega_{ba}\tau)\big]\exp(-i\omega\tau)\,d\tau \\
&= \{\text{evaluate integral } \int_{-\infty}^{0}\exp(iC\tau)\,d\tau \to 1/(iC)\} \\
&= -\frac{Ne^2}{\varepsilon_0 i\hbar}\sum_a \varrho_0(a)\sum_b \Big[\frac{r^\mu_{ab}r^\alpha_{ba}}{i(\Omega_{ba}-\omega)} - \frac{r^\alpha_{ab}r^\mu_{ba}}{-i(\Omega_{ba}+\omega)}\Big] \\
&= \frac{Ne^2}{\varepsilon_0\hbar}\sum_a \varrho_0(a)\sum_b \Big(\frac{r^\mu_{ab}r^\alpha_{ba}}{\Omega_{ba}-\omega} + \frac{r^\alpha_{ab}r^\mu_{ba}}{\Omega_{ba}+\omega}\Big).
\end{aligned}$$

This result, which was derived entirely under the assumption that all resonances of the medium are located far away from the angular frequency of the light, possesses a new type of symmetry, which can be seen if we make the interchange

$$(-\omega,\mu) \leftrightharpoons (\omega,\alpha),$$

which by using the above result for the linear susceptibility gives

$$\begin{aligned}
\chi^{(1)}_{\alpha\mu}(\omega;-\omega) &= \frac{Ne^2}{\varepsilon_0\hbar}\sum_a \varrho_0(a)\sum_b \Big(\frac{r^\alpha_{ab}r^\mu_{ba}}{\Omega_{ba}+\omega} + \frac{r^\mu_{ab}r^\alpha_{ba}}{\Omega_{ba}-\omega}\Big) \\
&= \{\text{change order of appearance of the terms}\} \\
&= \frac{Ne^2}{\varepsilon_0\hbar}\sum_a \varrho_0(a)\sum_b \Big(\frac{r^\mu_{ab}r^\alpha_{ba}}{\Omega_{ba}-\omega} + \frac{r^\alpha_{ab}r^\mu_{ba}}{\Omega_{ba}+\omega}\Big) \\
&= \chi^{(1)}_{\mu\alpha}(-\omega;\omega),
\end{aligned}$$

which is a signature of the *overall permutation symmetry* that applies to nonresonant interactions. With this result, the reason for the peculiar notation with "$(-\omega;\omega)$" should be all clear, namely that it serves as an explicit way of notation of the overall permutation symmetry, whenever it applies.

It should be noticed that while intrinsic permutation symmetry is a general property that solely applies to nonlinear optical interactions, the overall permutation symmetry applies to linear interactions as well.

*Second order electric susceptibility*

In similar to the linear electric susceptibility, starting from the general form of the second order (quadratic) electric susceptibility, one obtains

$$\chi^{(2)}_{\mu\alpha\beta}(-\omega_\sigma;\omega_1,\omega_2)$$

$$= \frac{Ne^3}{\varepsilon_0(i\hbar)^2}\frac{1}{2!}\mathbf{S}\int_{-\infty}^{0}\int_{-\infty}^{\tau_1}\mathrm{Tr}\{\hat\varrho_0[[\hat r_\mu,\hat r_\alpha(\tau_1)],\hat r_\beta(\tau_2)]\}\exp[-i(\omega_1\tau_1+\omega_2\tau_2)]\,d\tau_2\,d\tau_1$$

$$= \frac{Ne^3}{\varepsilon_0(i\hbar)^2}\frac{1}{2!}\mathbf{S}\int_{-\infty}^{0}\int_{-\infty}^{\tau_1}\sum_a\langle a|\hat\varrho_0[\underbrace{[\hat r_\mu,\hat r_\alpha(\tau_1)]}_{\hat r_\mu\hat r_\alpha(\tau_1)-\hat r_\alpha(\tau_1)\hat r_\mu},\hat r_\beta(\tau_2)]|a\rangle\exp[-i(\omega_1\tau_1+\omega_2\tau_2)]\,d\tau_2\,d\tau_1$$

$$= \{\text{expand the commutator}\}$$

$$= \frac{Ne^3}{\varepsilon_0(i\hbar)^2}\frac{1}{2!}\mathbf{S}\int_{-\infty}^{0}\int_{-\infty}^{\tau_1}\sum_a\Big(\langle a|\hat\varrho_0\hat r_\mu\hat r_\alpha(\tau_1)\hat r_\beta(\tau_2)|a\rangle-\langle a|\hat\varrho_0\hat r_\alpha(\tau_1)\hat r_\mu\hat r_\beta(\tau_2)|a\rangle$$

$$-\langle a|\hat\varrho_0\hat r_\beta(\tau_2)\hat r_\mu\hat r_\alpha(\tau_1)|a\rangle+\langle a|\hat\varrho_0\hat r_\beta(\tau_2)\hat r_\alpha(\tau_1)\hat r_\mu|a\rangle\Big)\exp[-i(\omega_1\tau_1+\omega_2\tau_2)]\,d\tau_2\,d\tau_1$$

$$= \{\text{apply the closure principle and Eq. (3)}\}$$

$$= \frac{Ne^3}{\varepsilon_0(i\hbar)^2}\frac{1}{2!}\mathbf{S}\int_{-\infty}^{0}\int_{-\infty}^{\tau_1}\sum_a\sum_k\sum_b\sum_c\Big(\langle a|\hat\varrho_0|k\rangle\langle k|\hat r_\mu|b\rangle\langle b|\hat r_\alpha(\tau_1)|b\rangle\langle c|\hat r_\beta(\tau_2)|a\rangle$$

$$-\langle a|\hat\varrho_0|k\rangle\langle k|\hat r_\alpha(\tau_1)|b\rangle\langle b|\hat r_\mu|c\rangle\langle c|\hat r_\beta(\tau_2)|a\rangle$$

$$-\langle a|\hat\varrho_0|k\rangle\langle k|\hat r_\beta(\tau_2)|b\rangle\langle b|\hat r_\mu|c\rangle\langle c|\hat r_\alpha(\tau_1)|a\rangle$$

$$+\langle a|\hat\varrho_0|k\rangle\langle k|\hat r_\beta(\tau_2)|b\rangle\langle b|\hat r_\alpha(\tau_1)|c\rangle\langle c|\hat r_\mu|a\rangle\Big)\exp[-i(\omega_1\tau_1+\omega_2\tau_2)]\,d\tau_2\,d\tau_1$$

$$= \{\text{use that }\langle a|\hat\varrho_0|k\rangle=\varrho_0(a)\delta_{ak}\text{ and apply Eq. (3)}\}$$

$$= \frac{Ne^3}{\varepsilon_0(i\hbar)^2}\frac{1}{2!}\mathbf{S}\sum_a\varrho_0(a)\sum_b\sum_c\int_{-\infty}^{0}\int_{-\infty}^{\tau_1}\Big\{r^\mu_{ab}r^\alpha_{bc}r^\beta_{ca}\exp[i(\Omega_{bc}\tau_1+\Omega_{ca}\tau_2)]$$

$$-r^\alpha_{ab}r^\mu_{bc}r^\beta_{ca}\exp[i(\Omega_{ab}\tau_1+\Omega_{ca}\tau_2)]-r^\beta_{ab}r^\mu_{bc}r^\alpha_{ca}\exp[i(\Omega_{ca}\tau_1+\Omega_{ab}\tau_2)]$$

$$+r^\beta_{ab}r^\alpha_{bc}r^\mu_{ca}\exp[i(\Omega_{bc}\tau_1+\Omega_{ab}\tau_2)]\Big\}\exp[-i(\omega_1\tau_1+\omega_2\tau_2)]\,d\tau_2\,d\tau_1$$

$$= \frac{Ne^3}{\varepsilon_0(i\hbar)^2}\frac{1}{2!}\mathbf{S}\sum_a\varrho_0(a)\sum_b\sum_c\int_{-\infty}^{0}\Big\{\frac{r^\mu_{ab}r^\alpha_{bc}r^\beta_{ca}}{i(\Omega_{ca}-\omega_2)}\exp[i\underbrace{(\Omega_{bc}+\Omega_{ca})}_{=\Omega_{ba}}\tau_1]$$

$$-\frac{r^\alpha_{ab}r^\mu_{bc}r^\beta_{ca}}{i(\Omega_{ca}-\omega_2)}\exp[i\underbrace{(\Omega_{ab}+\Omega_{ca})}_{=\Omega_{cb}}\tau_1]-\frac{r^\beta_{ab}r^\mu_{bc}r^\alpha_{ca}}{i(\Omega_{ab}-\omega_2)}\exp[i\underbrace{(\Omega_{ca}+\Omega_{ab})}_{=\Omega_{cb}}\tau_1)]$$

$$+\frac{r^\beta_{ab}r^\alpha_{bc}r^\mu_{ca}}{i(\Omega_{ab}-\omega_2)}\exp[i\underbrace{(\Omega_{bc}+\Omega_{ab})}_{=-\Omega_{ca}}\tau_1]\Big\}\exp[-i\underbrace{(\omega_1+\omega_2)}_{=\omega_\sigma}\tau_1]\,d\tau_1$$

$$= \frac{Ne^3}{\varepsilon_0(i\hbar)^2}\frac{1}{2!}\mathbf{S}\sum_a\varrho_0(a)\sum_b\sum_c\Big\{-\frac{r^\mu_{ab}r^\alpha_{bc}r^\beta_{ca}}{(\Omega_{ca}-\omega_2)(\Omega_{ba}-\omega_\sigma)}+\frac{r^\alpha_{ab}r^\mu_{bc}r^\beta_{ca}}{(\Omega_{ca}-\omega_2)(\Omega_{cb}-\omega_\sigma)}$$

$$+\frac{r^\beta_{ab}r^\mu_{bc}r^\alpha_{ca}}{(\Omega_{ab}-\omega_2)(\Omega_{cb}-\omega_\sigma)}-\frac{r^\beta_{ab}r^\alpha_{bc}r^\mu_{ca}}{(\Omega_{ab}-\omega_2)(-\Omega_{ca}-\omega_\sigma)}\Big\}$$

*... continued on next page ...*

*. . . continuing from previous page . . .*

$$= \frac{Ne^3}{\varepsilon_0\hbar^2}\frac{1}{2!}\mathbf{S}\sum_a \varrho_0(a)\sum_b\sum_c \Big\{ \frac{r^\mu_{ab} r^\alpha_{bc} r^\beta_{ca}}{(\Omega_{ac}+\omega_2)(\Omega_{ab}+\omega_\sigma)} - \frac{r^\alpha_{ab} r^\mu_{bc} r^\beta_{ca}}{(\Omega_{ac}+\omega_2)(\Omega_{bc}+\omega_\sigma)} - \frac{r^\beta_{ab} r^\mu_{bc} r^\alpha_{ca}}{(\Omega_{ba}+\omega_2)(\Omega_{bc}+\omega_\sigma)} + \frac{r^\beta_{ab} r^\alpha_{bc} r^\mu_{ca}}{(\Omega_{ba}+\omega_2)(\Omega_{ca}+\omega_\sigma)}\Big\}.$$

Now it is easily seen that if we, for example, interchange

$$(-\omega_\sigma, \mu) \rightleftharpoons (\omega_2, \beta),$$

one obtains

$$\begin{aligned}
&\chi^{(2)}_{\beta\alpha\mu}(\omega_2; \omega_1, -\omega_\sigma)\\
&\quad= \frac{Ne^3}{\varepsilon_0\hbar^2}\frac{1}{2!}\mathbf{S}\sum_a \varrho_0(a)\sum_b\sum_c \Big\{ \frac{r^\beta_{ab} r^\alpha_{bc} r^\mu_{ca}}{(\Omega_{ac}-\omega_\sigma)(\Omega_{ab}-\omega_2)} - \frac{r^\alpha_{ab} r^\beta_{bc} r^\mu_{ca}}{(\Omega_{ac}-\omega_\sigma)(\Omega_{bc}-\omega_2)}\\
&\qquad\qquad - \frac{r^\mu_{ab} r^\beta_{bc} r^\alpha_{ca}}{(\Omega_{ba}-\omega_\sigma)(\Omega_{bc}-\omega_2)} + \frac{r^\mu_{ab} r^\alpha_{bc} r^\beta_{ca}}{(\Omega_{ba}-\omega_\sigma)(\Omega_{ca}-\omega_2)}\Big\}\\
&\quad= \{\text{use } \Omega_{ab} = -\Omega_{ba},\ \text{etc.}\}\\
&\quad= \frac{Ne^3}{\varepsilon_0\hbar^2}\frac{1}{2!}\mathbf{S}\sum_a \varrho_0(a)\sum_b\sum_c \Big\{ \frac{r^\beta_{ab} r^\alpha_{bc} r^\mu_{ca}}{\underbrace{(\Omega_{ca}+\omega_\sigma)(\Omega_{ba}+\omega_2)}_{\text{identify 4th term}}} - \frac{r^\alpha_{ab} r^\beta_{bc} r^\mu_{ca}}{(\Omega_{ca}+\omega_\sigma)(\Omega_{cb}+\omega_2)}\\
&\qquad\qquad - \frac{r^\mu_{ab} r^\beta_{bc} r^\alpha_{ca}}{(\Omega_{ab}+\omega_\sigma)(\Omega_{cb}+\omega_2)} + \frac{r^\mu_{ab} r^\alpha_{bc} r^\beta_{ca}}{\underbrace{(\Omega_{ab}+\omega_\sigma)(\Omega_{ac}+\omega_2)}_{\text{identify 1st term}}}\Big\}\\
&\quad= \{\text{interchange dummy indices } a \to c \to b \to a \text{ in 2nd term}\}\\
&\quad= \{\text{interchange dummy indices } a \to b \to c \to a \text{ in 3rd term}\}\\
&\quad= \frac{Ne^3}{\varepsilon_0\hbar^2}\frac{1}{2!}\mathbf{S}\sum_a \varrho_0(a)\sum_b\sum_c \Big\{ \frac{r^\beta_{ab} r^\alpha_{bc} r^\mu_{ca}}{\underbrace{(\Omega_{ca}+\omega_\sigma)(\Omega_{ba}+\omega_2)}_{\text{identify 4th term}}} - \frac{r^\alpha_{ca} r^\beta_{ab} r^\mu_{bc}}{\underbrace{(\Omega_{bc}+\omega_\sigma)(\Omega_{ba}+\omega_2)}_{\text{identify as 3rd term}}}\\
&\qquad\qquad - \frac{r^\mu_{bc} r^\beta_{ca} r^\alpha_{ab}}{\underbrace{(\Omega_{bc}+\omega_\sigma)(\Omega_{ac}+\omega_2)}_{\text{identify as 2nd term}}} + \frac{r^\mu_{ab} r^\alpha_{bc} r^\beta_{ca}}{\underbrace{(\Omega_{ab}+\omega_\sigma)(\Omega_{ac}+\omega_2)}_{\text{identify 1st term}}}\Big\}\\
&\quad= \chi^{(2)}_{\mu\beta\alpha}(-\omega_\sigma; \omega_1, \omega_2),
\end{aligned}$$

that is to say, the second order susceptibility is left invariant under any of the $(2+1)! = 6$ possible pairwise permutations of $(-\omega_\sigma, \mu)$, $(\omega_1, \alpha)$, and $(\omega_2, \beta)$; this is the *overall permutation symmetry for the second order susceptibility*, and applies whenever the interaction is moved far away from any resonance.

We recapitulate that when deriving the form of the nonlinear susceptibilities that lead to intrinsic permutation symmetry, either in terms of polarization response functions in time domain or in terms of a mechanical spring model, nothing actually had to be stated regarding the nature of interaction. This is rather different from what we just obtained for the overall permutation symmetry, as being a signature of a nonresonant interaction between the light and matter, and this symmetry cannot (in contrary to the intrinsic permutation symmetry) be expressed unless the origin of interaction is considered.[2]

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[2] It should though be noticed that the derivation of the susceptibilities still may be performed within a mechanical spring model, as long as the resonance frequencies of the oscillator are removed far from the angular frequencies of the present light.

# Lecture VII

**Lecture Notes on Nonlinear Optics**
Nonlinear Optics (5A5513, 5p for advanced undergraduate and doctoral students)
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*Nonlinear Optics 5A5513 (2003)*
*Lecture notes*

# Lecture 7

So far, this course has mainly dealt with the dependence of the angular frequency of the light and molecular interaction strength in the description of nonlinear optics. In this lecture, we will now end this development of the description of interaction between light and matter, in favour of more engineering practical techniques for describing the theory of an experimental setup in a certain geometry, and for reducing the number of necessary tensor elements needed for describing a mediumof a certain crystallographic point-symmetry group.

## Motivation for analysis of susceptibilities in rotated coordinate systems

For a given experimental setup, it is often convenient to introduce some kind of reference coordinate frame, in which one for example express the wave propagation as a linear motion along some Cartesian coordinate axis. This laboratory reference frame might be chosen, for example, with the $z$-axis coinciding with the direction of propagation of the optical wave at the laser output, in the phase-matched direction of an optical parametric oscillator (OPO), after some beam aligning mirror, etc.

In some cases, it might be so that this laboratory frame coincide with the natural coordinate frame[1] of the nonlinear crystal, in which case the coordinate indices of the linear as well as nonlinear susceptibility tensors take the same values as the coordinates of the laboratory frame. However, we cannot generally assume the coordinate frame of the crystal to coincide with a conveniently chosen laboratory reference frame, and this implies that we generally should be prepared to spatially transform the susceptibility tensors to arbitrarily rotated coordinate frames.

Having formulated these spatial transformation rules, we will also directly benefit in another aspect of the description of nonlinear optical interactions, namely the reduction of the susceptibility tensors to the minimal set of nonzero elements. This is typically performed by using the knowledge of the so called *crystallographic point ymmetry group* of the medium, which essentially is a description of the spatial operations (rotations, inversions etc.) that define the symmetry operations of the medium.

As a particular example of the applicability of the spatial transformation rules (which we soon will formulate) is illustrated in Figs. 1 and 2. In Fig. 1, the procedure for analysis of sum or frequency difference generation is outlined. Starting from the description of the linear and nonlinear susceptibility tensors of the medium, as we previously have derived the relations from a first principle approach in Lectures 1–6, we obtain the expressions for the electric polarization densities of the medium as functions of the applied electric fields of the optical wave inside the nonlinear crystal. These polarization densities are then inserted into the wave equation, which basically is derived from Maxwell's equations of motion for the electromagnetic field. In the wave equation, the polarization densities act as source terms in an otherwise homogeneous equation for the motion of the electromagnetic field in vacuum.

As the wave equation is solved for the electric field, here taken in complex notation, we have solved for the general output from the crystal, and we can then design the experiment in such a way that an optimal efficiency is obtained.

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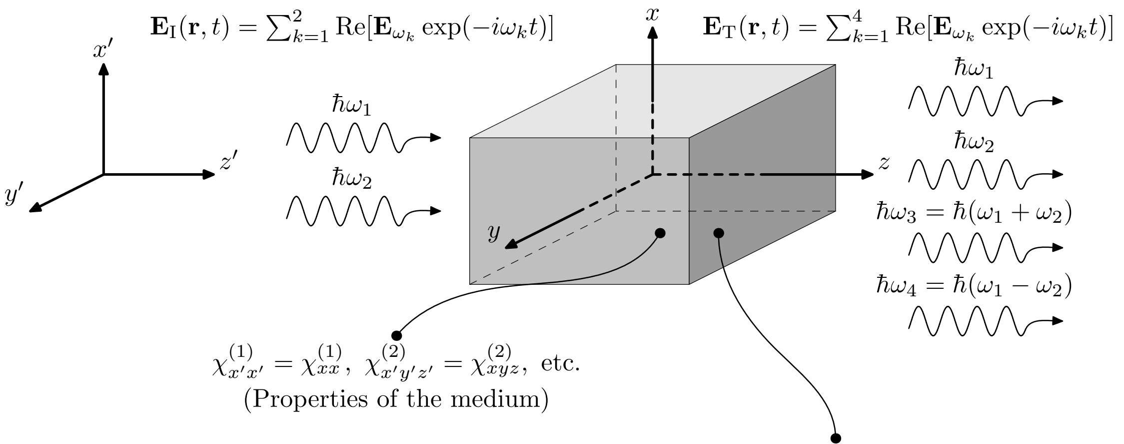
[1] The natural coordinate frame of the crystal is often chosen such that some particular symmetry axis is chosen as one of the Cartesian axes.

Laboratory frame $(x', y', z')$ Crystal frame $(x, y, z)$



$\chi^{(1)}_{x'x'}=\chi^{(1)}_{xx},\ \chi^{(2)}_{x'y'z'}=\chi^{(2)}_{xyz}$, etc.
(Properties of the medium)

$$
\begin{aligned}
\Rightarrow\quad \mathbf{P}^{(1)}_{\omega_k} &= \varepsilon_0\mathbf{e}_x\chi^{(1)}_{xx}(-\omega_k;\omega_k)(\mathbf{e}_x\cdot\mathbf{E}_{\omega_k}),\ \text{etc.}\\
\mathbf{P}^{(2)}_{\omega_3} &= \varepsilon_0\mathbf{e}_x\chi^{(2)}_{xyz}(-\omega_3;\omega_1,\omega_2)(\mathbf{e}_y\cdot\mathbf{E}_{\omega_1})(\mathbf{e}_z\cdot\mathbf{E}_{\omega_2})\\
&\quad+\varepsilon_0\mathbf{e}_y\chi^{(2)}_{yxx}(-\omega_3;\omega_1,\omega_2)(\mathbf{e}_x\cdot\mathbf{E}_{\omega_1})(\mathbf{e}_x\cdot\mathbf{E}_{\omega_2}),\ \text{etc.}\\
\mathbf{P}^{(2)}_{\omega_4} &= \varepsilon_0\mathbf{e}_x\chi^{(2)}_{xyz}(-\omega_4;\omega_1,-\omega_2)(\mathbf{e}_y\cdot\mathbf{E}_{\omega_1})(\mathbf{e}_z\cdot\mathbf{E}^*_{\omega_2})\\
&\quad+\varepsilon_0\mathbf{e}_y\chi^{(2)}_{yxx}(-\omega_4;\omega_1,-\omega_2)(\mathbf{e}_x\cdot\mathbf{E}_{\omega_1})(\mathbf{e}_x\cdot\mathbf{E}^*_{\omega_2}),\ \text{etc.}
\end{aligned}
$$

$$\Rightarrow\quad \frac{d^2\mathbf{E}_{\omega_k}}{dz'^2}+\frac{\omega_k^2}{c^2}\mathbf{E}_{\omega_k}=-\mu_0\omega^2(\mathbf{P}^{(1)}_{\omega_k}+\mathbf{P}^{(2)}_{\omega_k}),\quad k=1,2,3,4$$

(Nonlinear wave equation inside crystal)

$$\Rightarrow\quad \mathbf{E}_{\omega_k}=\mathbf{E}_{\omega_k}(z),\quad k=1,2,3,4$$

(Solution for optical fields)

Figure 1. The setup in which the orientation of the laboratory and crystal frames coincide.

In Fig. 1, this outline is illustrated for the case where the natural coordinate frame of the crystal happens to coincide with the coordinate system of the laboratory frame. In this case, all elements of the susceptibilities taken in the coordinate frame of the crystal (which naturally is the coordinate frame in which we can obtain tabulated sets of tensor elements) will coincide with the elements as taken in the laboratory frame, and the design and interpretation of the experiment is straightforward.

However, this setup clearly constitutes a rare case, since we have infinitely many other possibilities of orienting the crystal relative the laboratory coordinate frame. Sometimes the experiment is *designed* with the crystal and laboratory frames coinciding, in order to simplify the interpretation of an experiment, and sometimes it is instead *necessary* to rotate the crytal, in order to achieve phase-matching of nonlinear process, as is the case in for example most schemes for second-order optical parametric amplification.

If now the crystal frame is rotated with respect to the laboratory frame, as shown in Fig. 2, we should make up our mind in which system we would like the wave propagation to be analyzed. In some cases, it might be so that the output of the experimental setup is most easily interpreted in the coordinate frame of the crystal, but in most cases, we have a fixed laboratory frame (fixed by the orientation of the laser, positions of mirrors, etc.) in which we would like to express the wave propagation and interaction between light and matter.

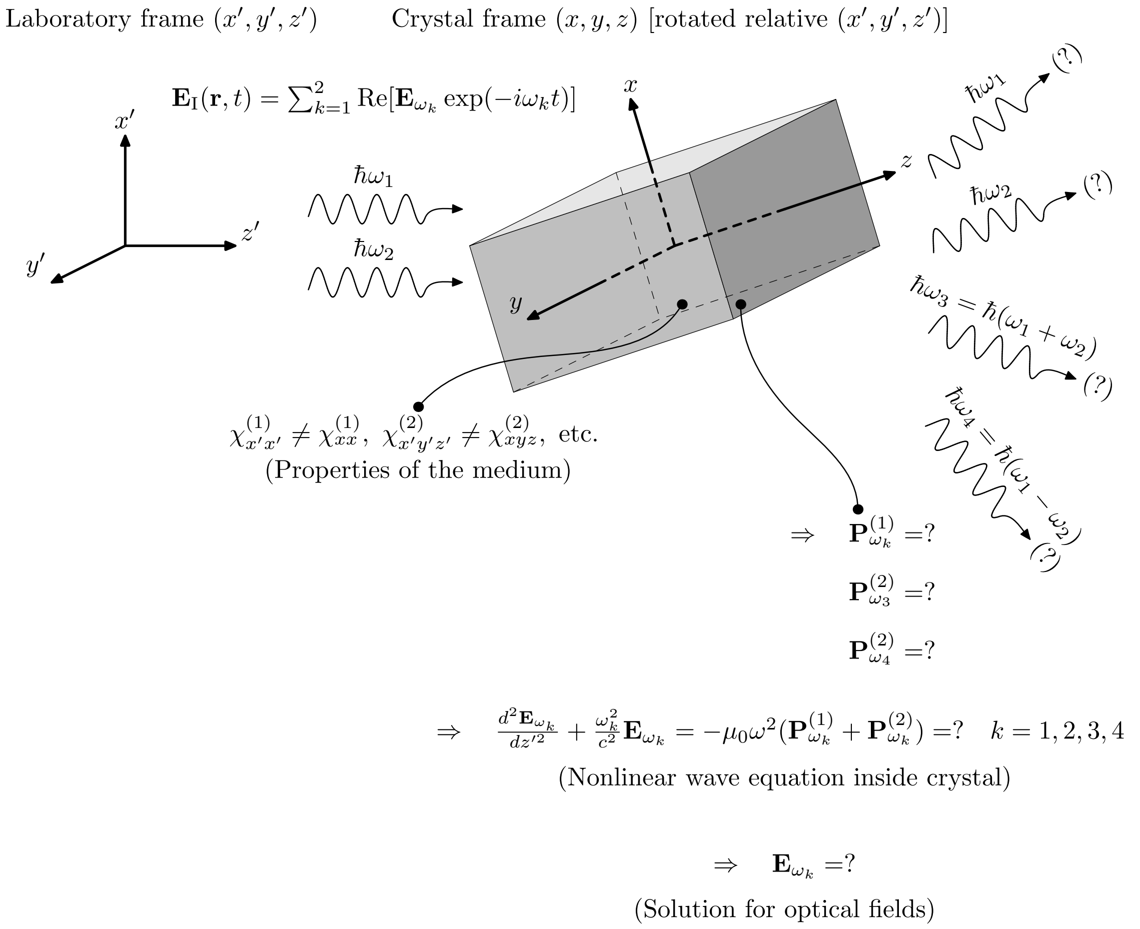


Figure 2. The setup in which the crystal frame is rotated relative the laboratory frame.

In Fig. 2, we would, in order to express the nonlinear process in the laboratory frame, like to obtain the naturally appearing susceptibilities $\chi^{(2)}_{xyz}$, $\chi^{(2)}_{xxx}$, etc., in the laboratory frame instead, as $\chi^{(2)}_{x'y'z'}$, $\chi^{(2)}_{x'x'x'}$, etc.

Just to summarize, why are then the transformation rules and spatial symmetries of the meduim so important?

- Hard to make physical conclusions about generated optical fields unless orientation of the laboratory and crystal frames coincide.
- Spatial symmetries often significantly simplifies the wave propagation problem (by choosing a suitable polarization state and direction of propagation of the light, etc.).
- Useful for reducing the number of necessary elements of the susceptibility tensors (using Neumann's principle).

### Optical properties in rotated coordinate frames

Consider two coordinate systems described by Cartesian coordinates $x_\alpha$ and $x'_\alpha$, respectively. The coordinate systems are rotated with respect to each other, and the relation between the coordinates are described by the $[3 \times 3]$ transformation matrix $R_{ab}$ as

$$\mathbf{x}' = \mathbf{R}\mathbf{x} \qquad \Leftrightarrow \qquad x'_\alpha = R_{\alpha\beta}x_\beta, \qquad \text{[B.\,\&\,C. (5.40)]}$$

where $\mathbf{x} = (x, y, z)^{\mathrm{T}}$ and $\mathbf{x}' = (x', y', z')^{\mathrm{T}}$ are column vectors. The inverse transformation between the coordinate systems is similarly given as

$$\mathbf{x} = \mathbf{R}^{-1}\mathbf{x}' \qquad \Leftrightarrow \qquad x_\beta = R_{\alpha\beta}x'_\alpha. \qquad \text{[B.\,\&\,C. (5.41)]}$$

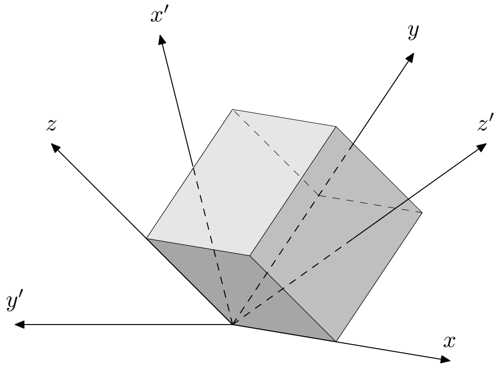


Figure 3. Illustration of proper rotation of the crystal frame $(x, y, z)$ relative to the laboratory reference frame $(x', y', z')$, by means of $x'_\alpha = R_{\alpha\beta} x_\beta$ with $\det(\mathbf{R}) = 1$.

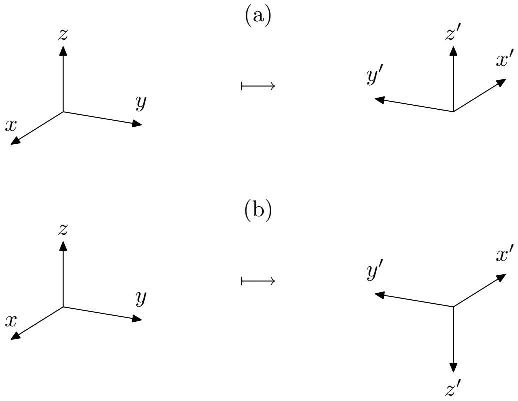


Figure 4. The coordinate transformations (a) $\mathbf{x} = (x, y, z) \mapsto \mathbf{x}' = (-x, -y, z)$, constituting a proper rotation around the $z$-axis, and (b) the space inversion $\mathbf{x} \mapsto \mathbf{x}' = -\mathbf{x}$, an improper rotation corresponding to, for example, a rotation around the $z$-axis followed by an inversion in the $xy$-plane.

We should notice that there are two types of rotations that are encountered as transformations:

- Proper rotations, for which $\det(\mathbf{R}) = 1$. (Righthanded systems keep being righthanded, and lefthanded systems keep being lefthanded.)
- Improper rotations, for which $\det(\mathbf{R}) = -1$. (Righthanded systems are transformed into lefthanded systems, and vice versa.)

The electric field $\mathbf{E}(\mathbf{r}, t)$ and electric polarization density $\mathbf{P}(\mathbf{r}, t)$ are both polar quantities that transform in the same way as regular Cartesian coordinates, and hence we have descriptions of these quantities in coordinate systems $(x, y, z)$ and $(x', y', z')$ related to each other as

$$E'_\mu(\mathbf{r}, t) = R_{\mu u} E_u(\mathbf{r}, t) \qquad \Leftrightarrow \qquad E_u(\mathbf{r}, t) = R_{\mu u} E'_\mu(\mathbf{r}, t),$$

and

$$P'_\mu(\mathbf{r}, t) = R_{\mu u} P_u(\mathbf{r}, t) \qquad \Leftrightarrow \qquad P_u(\mathbf{r}, t) = R_{\mu u} P'_\mu(\mathbf{r}, t),$$

respectively. Using these transformation rules, we will now derive the form of the susceptibilities in rotated coordinate frames.

*First order polarization density in rotated coordinate frames*

From the transformation rule for the electric polarization density above, using the standard form as we previously have expressed the electric field dependence, we have for the first order polarization density in the primed coordinate system

$$\begin{aligned} P^{(1)\prime}_{\mu}(\mathbf{r},t) &= R_{\mu u}P^{(1)}_{u}(\mathbf{r},t)\\ &= R_{\mu u}\varepsilon_0\int_{-\infty}^{\infty}\chi^{(1)}_{ua}(-\omega;\omega)E_a(\omega)\exp(-i\omega t)\,d\omega\\ &= R_{\mu u}\varepsilon_0\int_{-\infty}^{\infty}\chi^{(1)}_{ua}(-\omega;\omega)R_{\alpha a}E'_{\alpha}(\omega)\exp(-i\omega t)\,d\omega\\ &= \varepsilon_0\int_{-\infty}^{\infty}\chi^{(1)\prime}_{\mu\alpha}(-\omega;\omega)E'_{\alpha}(\omega)\exp(-i\omega t)\,d\omega \end{aligned}$$

where

$$\chi^{(1)\prime}_{\mu\alpha}(-\omega;\omega) = R_{\mu u}R_{\alpha a}\chi^{(1)}_{ua}(-\omega;\omega) \qquad \text{[B. \& C. (5.45)]}$$

is the linear electric susceptibility taken in the primed coordinate system.

*Second order polarization density in rotated coordinate frames*

Similarly, we have the second order polarization density in the primed coordinate system as

$$\begin{aligned} P^{(2)\prime}_{\mu}(\mathbf{r},t) &= R_{\mu u}P^{(2)}_{u}(\mathbf{r},t)\\ &= R_{\mu u}\varepsilon_0\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\chi^{(2)}_{uab}(-\omega_\sigma;\omega_1,\omega_2)E_a(\omega_1)E_b(\omega_2)\exp[-i(\omega_1+\omega_1)t]\,d\omega_2\,d\omega_1\\ &= R_{\mu u}\varepsilon_0\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\chi^{(2)}_{uab}(-\omega_\sigma;\omega_1,\omega_2)R_{\alpha a}E'_{\alpha}(\omega_1)R_{\beta b}E'_{\beta}(\omega_2)\exp[-i(\omega_1+\omega_1)t]\,d\omega_2\,d\omega_1\\ &= \varepsilon_0\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\chi^{(2)}_{\mu\alpha\beta}{}'(-\omega_\sigma;\omega_1,\omega_2)E'_{\alpha}(\omega_1)E'_{\beta}(\omega_2)\exp[-i(\omega_1+\omega_1)t]\,d\omega_2\,d\omega_1 \end{aligned}$$

where

$$\chi^{(2)}_{\mu\alpha\beta}{}'(-\omega_\sigma;\omega_1,\omega_2) = R_{\mu u}R_{\alpha a}R_{\beta b}\chi^{(2)}_{uab}(-\omega_\sigma;\omega_1,\omega_2) \qquad \text{[B. \& C. (5.46)]}$$

is the second order electric susceptibility taken in the primed coordinate system.

*Higher order polarization densities in rotated coordinate frames*

In a manner completely analogous to the second order susceptibility, the transformation rule between the primed and unprimed coordinate systems can be obtained for the $n$th order elements of the electric susceptibility tensor as

$$\chi^{(n)}_{\mu\alpha_1\cdots\alpha_n}{}'(-\omega_\sigma;\omega_1,\dots,\omega_n) = R_{\mu u}R_{\alpha_1 a_1}\cdots R_{\alpha_n a_n}\chi^{(n)}_{ua_1\cdots a_n}(-\omega_\sigma;\omega_1,\dots,\omega_n). \qquad \text{[B. \& C. (5.47)]}$$

## Crystallographic point symmetry groups

Typically, a particular point symmetry group of the medium can be described by the *generating matrices* that describe the minimal set of transformation matrices (describing a set of symmetry operations) that will be necessary for the reduction of the constitutive tensors. Two systems are widely used for the description of point symmetry groups:[2]

- The International system, e. g. $\bar{4}3m$, $m3m$, 422, etc.
- The Schönflies system, e. g. $T_d$, $O_h$, $D_4$, etc.

The crystallographic point symmetry groups may contain any of the following symmetry operations:

*1. Rotations through integral multiples of* $2\pi/n$ *about some axis.* The axis is called the $n$-fold rotation axis. It is in solid state physics shown [1–3] that a Bravais lattice can contain only 2-, 3-, 4-, or 6-fold axes, and since the crystallographic point symmetry groups are contained in the Bravais lattice point groups, they too can only have these axes.

---

[2] C. f. Table 2 of the handed out Hartmann's *An Introduction to Crystal Physics*.

*2. Rotation-reflections.* Even when a rotation through $2\pi/n$ is not a symmetry element, sometimes such a rotation followed by a reflection in a plane perpendicular to the axis may be a symmetry operation. The axis is then called an $n$-fold rotation-reflection axis. For example, the groups $S_6$ and $S_4$ have 6- and 4-fold rotation-reflection axes.

*3. Rotation-inversions.* Similarly, sometimes a rotation through $2\pi/n$ followed by an inversion in a point lying on the rotation axis is a symmetry element, even though such a rotation by itself is not. The axis is then called an $n$-fold rotation-inversion axis. However, the axis in $S_6$ is only a 3-fold rotation-inversion axis.

*4. Reflections.* A reflection takes every point into its mirror image in a plane, known as a mirror plane.

*5. Inversions.* An inversion has a single fixed point. If that point is taken as the origin, then every other point $\mathbf{r}$ is taken into $-\mathbf{r}$.

### Schönflies notation for the non-cubic crystallographic point groups

The twenty-seven *non-cubic* crystallographic point symmetry groups may contain any of the following symmetry operations, here given in Schönflies notation[3]:

| | |
|---|---|
| $C_n$ | These groups contain only an $n$-fold rotation axis. |
| $C_{n\mathrm{v}}$ | In addition to the $n$-fold rotation axis, these groups have a mirror plane that contains the axis of rotation, plus as many additional mirror planes as the existence of the $n$-fold axis requires. |
| $C_{n\mathrm{h}}$ | These groups contain in addition to the $n$-fold rotation axis a single mirror plane that is perpendicular to the axis. |
| $S_n$ | These groups contain only an $n$-fold rotation-reflection axis. |
| $D_n$ | In addition to the $n$-fold rotation axis, these groups contain a 2-fold axis perpendicular to the $n$-fold rotation axis, plus as many additional 2-fold axes as are required by the existence of the $n$-fold axis. |
| $D_{n\mathrm{h}}$ | These (the most symmetric groups) contain all the elements of $D_n$ plus a mirror plane perpendicular to the $n$-fold axis. |
| $D_{n\mathrm{d}}$ | These contain the elements of $D_n$ plus mirror planes containing the $n$-fold axis, which bisect the angles between the 2-fold axes. |

### Neumann's principle

Neumann's principle simply states that *any type of symmetry which is exhibited by the point symmetry group of the medium is also possessed by every physical property of the medium.*

In other words, we can reformulate this for the optical properties as: *the susceptibility tensors of the medium must be left invariant under any transformation that also is a point symmetry operation of the medium*, or

$$\chi^{(n)'}_{\mu\alpha_1\cdots\alpha_n}(-\omega_\sigma;\omega_1,\ldots,\omega_n)=\chi^{(n)}_{\mu\alpha_1\cdots\alpha_n}(-\omega_\sigma;\omega_1,\ldots,\omega_n),$$

where the tensor elements in the primed coordinate system are transformed according to

$$\chi^{(n)'}_{\mu\alpha_1\cdots\alpha_n}(-\omega_\sigma;\omega_1,\ldots,\omega_n)=R_{\mu u}R_{\alpha_1 a_1}\cdots R_{\alpha_n a_n}\chi^{(n)}_{ua_1\cdots a_n}(-\omega_\sigma;\omega_1,\ldots,\omega_n),$$

where the $[3\times 3]$ matrix $\mathbf{R}$ describes a point symmetry operation of the system.

---

[3] In Schönflies notation, $C$ stands for "cyclic", $D$ for "dihedral", and $S$ for "spiegel". The subscripts h, v, and d stand for "horizontal", "vertical", and "diagonal", respectively, and refer to the placement of the placement of the mirror planes with respect to the $n$-fold axis, always considered to be vertical. (The "diagonal" planes in $D_{n\mathrm{d}}$ are vertical and bisect the angles between the 2-fold axes)

### Inversion properties

If the *coordinate inversion* $R_{\alpha\beta} = -\delta_{\alpha\beta}$, is a symmetry operation of the medium (i. e. if the medium possess so-called *inversion symmetry*), then it turns out that

$$\chi^{(n)}_{\mu\alpha_1\cdots\alpha_n} = 0$$

for all *even* numbers $n$. (Question: Is this symmetry operation a proper or an improper rotation?)

### Euler angles

As a convenient way of expressing the matrix of proper rotations, one may use the *Euler angles* of classical mechanics,[4]

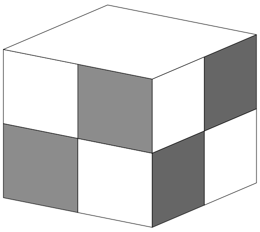
$$\mathbf{R}(\varphi, \vartheta, \psi) = \mathbf{A}(\psi)\mathbf{B}(\vartheta)\mathbf{C}(\varphi),$$

where

$$\mathbf{A}(\psi) = \begin{pmatrix} \cos\psi & \sin\psi & 0 \\ -\sin\psi & \cos\psi & 0 \\ 0 & 0 & 1 \end{pmatrix}, \ \mathbf{B}(\vartheta) = \begin{pmatrix} 1 & 0 & 0 \\ 0 & \cos\vartheta & \sin\vartheta \\ 0 & -\sin\vartheta & \cos\vartheta \end{pmatrix}, \ \mathbf{C}(\varphi) = \begin{pmatrix} \cos\varphi & \sin\varphi & 0 \\ -\sin\varphi & \cos\varphi & 0 \\ 0 & 0 & 1 \end{pmatrix}.$$

### Example of the direct inspection technique applied to tetragonal media

Neumann's principle is a highly useful technique, with applications in a wide range of disciplines in physics. In order to illustrate this, we will now apply Neumann's principle to a particular problem, namely the reduction of the number of elements of the second order electric susceptibility tensor, in a tetragonal medium belonging to point symmetry group 422.



422

Figure 5. An object[5] possessing the symmetries of point symmetry group 422.

By inspecting Tables 2 and 3 of Hartmann's *An introduction to Crystal Physics*[6] one find that the point symmetry group 422 of tetragonal media is described by the generating matrices

$$\mathbf{M}_4 = \begin{pmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & -1 \end{pmatrix}, \qquad \begin{bmatrix} \text{twofold rotation} \\ \text{about } x_1 \text{ axis} \end{bmatrix}$$

and

$$\mathbf{M}_7 = \begin{pmatrix} 0 & -1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix}. \qquad \begin{bmatrix} \text{fourfold rotation} \\ \text{about } x_3 \text{ axis} \end{bmatrix}$$

---

[4] C. f. Herbert Goldstein, *Classical Mechanics* (Addison-Wesley, London, 1980).

[5] The figure illustrating the point symmetry group 422 is taken from N. W. Ashcroft and N. D. Mermin, *Solid state physics* (Saunders College Publishing, Orlando, 1976), page 122.

[6] Ervin Hartmann, *An Introduction to Crystal Physics* (University of Cardiff Press, International Union of Crystallography, 1984), ISBN 0-906449-72-3. Notice that there is a printing error in Table 3, where the twofold rotation about the $x_3$-axis should be described by a matrix denoted "$M_2$", and not "$M_1$" as written in the table.

*Does the 422 point symmetry group possess inversion symmetry?*

In Fig. 6, the steps involved for transformation of the object into an inverted coordinate frame are shown.

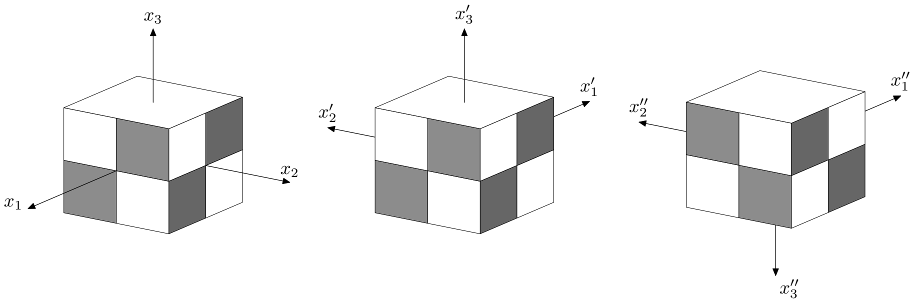


Figure 6. Transformation into an inverted coordinate system $(x'', y'', z'') = (-x, -y, -z)$.

The result of the sequence in Fig. 6 is an object which cannot be reoriented in such a way that one obtains the same shape as we started with for the non-inverted coordinate system, and hence the object of point symmetry group 422 does not possess inversion symmetry.

*Step one – Point symmetry under twofold rotation around the $x_1$-axis*

Considering the point symmetry imposed by the $\mathbf{R} = \mathbf{M}_4$ matrix, we find that (for simplicity omitting the frequency arguments of the susceptibility tensor) the second order susceptibility in the rotated coordinate frame is described by the diagonal elements

$$\begin{aligned}\chi^{(2)'}_{111} &= R_{1\mu}R_{1\alpha}R_{1\beta}\chi^{(2)}_{\mu\alpha\beta}\\ &= \sum_{\mu=1}^{3}\sum_{\alpha=1}^{3}\sum_{\beta=1}^{3} R_{1\mu}R_{1\alpha}R_{1\beta}\chi^{(2)}_{\mu\alpha\beta}\\ &= \sum_{\mu=1}^{3}\sum_{\alpha=1}^{3}\sum_{\beta=1}^{3} \delta_{1\mu}\delta_{1\alpha}\delta_{1\beta}\chi^{(2)}_{\mu\alpha\beta} = \chi^{(2)}_{111}, \qquad \text{(identity)}\end{aligned}$$

and

$$\begin{aligned}\chi^{(2)'}_{222} &= R_{2\mu}R_{2\alpha}R_{2\beta}\chi^{(2)}_{\mu\alpha\beta}\\ &= \sum_{\mu=1}^{3}\sum_{\alpha=1}^{3}\sum_{\beta=1}^{3} R_{2\mu}R_{2\alpha}R_{2\beta}\chi^{(2)}_{\mu\alpha\beta}\\ &= \sum_{\mu=1}^{3}\sum_{\alpha=1}^{3}\sum_{\beta=1}^{3} (-\delta_{2\mu})(-\delta_{2\alpha})(-\delta_{2\beta})\chi^{(2)}_{\mu\alpha\beta} = -\chi^{(2)}_{222}\\ &= \{\text{Neumann's principle}\} = \chi^{(2)}_{222} = 0\end{aligned}$$

which, by noticing that the similar form $R_{3\alpha} = -\delta_{3\alpha}$ holds for the 333-component (i. e. the $zzz$-component), also gives $\chi_{333} = -\chi_{333} = 0$. Further we have for the 231-component

$$\begin{aligned}\chi^{(2)'}_{231} &= R_{2\mu}R_{3\alpha}R_{1\beta}\chi^{(2)}_{\mu\alpha\beta}\\ &= \sum_{\mu=1}^{3}\sum_{\alpha=1}^{3}\sum_{\beta=1}^{3} R_{2\mu}R_{3\alpha}R_{1\beta}\chi^{(2)}_{\mu\alpha\beta}\\ &= \sum_{\mu=1}^{3}\sum_{\alpha=1}^{3}\sum_{\beta=1}^{3} (-\delta_{2\mu})(-\delta_{3\alpha})\delta_{1\beta}\chi^{(2)}_{\mu\alpha\beta} = \chi^{(2)}_{231}, \qquad \text{(identity)}\end{aligned}$$

etc., and by continuing in this manner for all 27 elements of $\chi^{(2)'}_{\mu\alpha\beta}$, one finds that the symmetry operation $\mathbf{R} = \mathbf{M}_4$ leaves us with the tensor elements listed in Table 1.

| Zero elements | Identities (no further info) |
|---|---|
| $\chi^{(2)}_{112}$, $\chi^{(2)}_{113}$, $\chi^{(2)}_{121}$, $\chi^{(2)}_{131}$,<br>$\chi^{(2)}_{211}$, $\chi^{(2)}_{222}$, $\chi^{(2)}_{223}$, $\chi^{(2)}_{232}$,<br>$\chi^{(2)}_{233}$, $\chi^{(2)}_{311}$, $\chi^{(2)}_{322}$, $\chi^{(2)}_{323}$,<br>$\chi^{(2)}_{332}$, $\chi^{(2)}_{333}$ | (all other 13 elements) |

Table 1. Reduced set of tensor elements after the symmetry operation $\mathbf{R} = \mathbf{M}_4$.

*Step two – Point symmetry under fourfold rotation around the $x_3$-axis*

Proceeding with the next point symmetry operation, described by $\mathbf{R} = \mathbf{M}_7$, one finds for the remaining 13 elements that, for example, for the 123-element

$$\begin{aligned}
\chi^{(2)'}_{123} &= R_{1\mu}R_{2\alpha}R_{3\beta}\chi^{(2)}_{\mu\alpha\beta} \\
&= \sum_{\mu=1}^{3}\sum_{\alpha=1}^{3}\sum_{\beta=1}^{3} R_{1\mu}R_{2\alpha}R_{3\beta}\chi^{(2)}_{\mu\alpha\beta} \\
&= \sum_{\mu=1}^{3}\sum_{\alpha=1}^{3}\sum_{\beta=1}^{3} (-\delta_{2\mu})\delta_{1\alpha}\delta_{3\beta}\chi^{(2)}_{\mu\alpha\beta} = -\chi^{(2)}_{213} \\
&= \{\text{Neumann's principle}\} = \chi^{(2)}_{123},
\end{aligned}$$

and for the 132-element

$$\begin{aligned}
\chi^{(2)'}_{132} &= R_{1\mu}R_{3\alpha}R_{2\beta}\chi^{(2)}_{\mu\alpha\beta} \\
&= \sum_{\mu=1}^{3}\sum_{\alpha=1}^{3}\sum_{\beta=1}^{3} R_{1\mu}R_{3\alpha}R_{2\beta}\chi^{(2)}_{\mu\alpha\beta} \\
&= \sum_{\mu=1}^{3}\sum_{\alpha=1}^{3}\sum_{\beta=1}^{3} (-\delta_{2\mu})\delta_{3\alpha}\delta_{1\beta}\chi^{(2)}_{\mu\alpha\beta} = -\chi^{(2)}_{231} \\
&= \{\text{Neumann's principle}\} = \chi^{(2)}_{132},
\end{aligned}$$

while the 111-element (which previously, by using the $\mathbf{R} = \mathbf{M}_4$ point symmetry, just gave an identity with no further information) now gives

$$\begin{aligned}
\chi^{(2)'}_{111} &= R_{1\mu}R_{1\alpha}R_{1\beta}\chi^{(2)}_{\mu\alpha\beta} \\
&= \sum_{\mu=1}^{3}\sum_{\alpha=1}^{3}\sum_{\beta=1}^{3} R_{1\mu}R_{1\alpha}R_{1\beta}\chi^{(2)}_{\mu\alpha\beta} \\
&= \sum_{\mu=1}^{3}\sum_{\alpha=1}^{3}\sum_{\beta=1}^{3} (-\delta_{2\mu})(-\delta_{2\alpha})(-\delta_{2\beta})\chi^{(2)}_{\mu\alpha\beta} = -\chi^{(2)}_{222} \\
&= \{\text{from previous result for } \chi^{(2)}_{222}\} = 0 \\
&= \{\text{Neumann's principle}\} = \chi^{(2)}_{111}.
\end{aligned}$$

By (again) proceeding for all 27 elements of $\chi^{(2)'}_{\mu\alpha\beta}$, one finds the set of tensor elements as listed in

Table 2. (See also the tabulated set in Butcher and Cotter's book, Table A3.2, page 299.)

| Zero elements | Nonzero elements |
|---|---|
| $\chi^{(2)}_{111}, \chi^{(2)}_{112}, \chi^{(2)}_{113}, \chi^{(2)}_{121}, \chi^{(2)}_{122},$ | $\chi^{(2)}_{123} = -\chi^{(2)}_{213},$ |
| $\chi^{(2)}_{131}, \chi^{(2)}_{133}, \chi^{(2)}_{211}, \chi^{(2)}_{212}, \chi^{(2)}_{221},$ | $\chi^{(2)}_{132} = -\chi^{(2)}_{231},$ |
| $\chi^{(2)}_{222}, \chi^{(2)}_{223}, \chi^{(2)}_{232}, \chi^{(2)}_{233}, \chi^{(2)}_{311},$ | $\chi^{(2)}_{321} = -\chi^{(2)}_{312},$ |
| $\chi^{(2)}_{313}, \chi^{(2)}_{322}, \chi^{(2)}_{323}, \chi^{(2)}_{331}, \chi^{(2)}_{332}, \chi^{(2)}_{333}$ | (6 nonzero, 3 independent) |

Table 2. Reduced set of tensor elements after symmetry operations $\mathbf{R} = \mathbf{M}_4$ and $\mathbf{R} = \mathbf{M}_7$.

# Lecture VIII

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*Lecture notes*

# LECTURE 8

In this lecture, the electric polarisation density of the medium is finally inserted into Maxwell's equations, and the wave propagation properties of electromagnetic waves in nonlinear optical media is for the first time in this course analysed. As an example of wave propagation in nonlinear optical media, the optical Kerr effect is analysed for infinite plane continuous waves.

The outline for this lecture is:

- Maxwells equations (general electromagnetic wave propagation)
- Time dependent processes (envelopes slowly varying in space and time)
- Time independent processes (envelopes slowly varying in space but constant in time)
- Examples (optical Kerr-effect $\Leftrightarrow \chi^{(3)}_{\mu\alpha\beta\gamma}(-\omega;\omega,\omega,-\omega)$)

## Wave propagation in nonlinear media

*Maxwell's equations*

The propagation of electromagnetic waves are, from a first principles approach, governed by the *Maxwell's equations* (here listed in their real-valued form in SI units),

$$\begin{aligned}
\nabla\times\mathbf{E}(\mathbf{r},t) &= -\frac{\partial\mathbf{B}(\mathbf{r},t)}{\partial t}, && \text{(Faraday's law)}\\
\nabla\times\mathbf{H}(\mathbf{r},t) &= \mathbf{J}(\mathbf{r},t)+\frac{\partial\mathbf{D}(\mathbf{r},t)}{\partial t}, && \text{(Ampere's law)}\\
\nabla\cdot\mathbf{D}(\mathbf{r},t) &= \rho(\mathbf{r},t),\\
\nabla\cdot\mathbf{B}(\mathbf{r},t) &= 0,
\end{aligned}$$

where $\rho(\mathbf{r},t)$ is the density of free charges, and $\mathbf{J}(\mathbf{r},t)$ the corresponding current density of free charges.

*Constitutive relations*

The constitutive relations are in SI units formulated as

$$\begin{aligned}
\mathbf{D}(\mathbf{r},t) &= \varepsilon_0\mathbf{E}(\mathbf{r},t)+\mathbf{P}(\mathbf{r},t),\\
\mathbf{B}(\mathbf{r},t) &= \mu_0[\mathbf{H}(\mathbf{r},t)+\mathbf{M}(\mathbf{r},t)],
\end{aligned}$$

where $\mathbf{P}(\mathbf{r},t)=\mathbf{P}[\mathbf{E}(\mathbf{r},t),\mathbf{B}(\mathbf{r},t)]$ is the macroscopic polarization density (electric dipole moment per unit volume), and $\mathbf{M}(\mathbf{r},t)=\mathbf{M}[\mathbf{E}(\mathbf{r},t),\mathbf{B}(\mathbf{r},t)]$ the magnetization (magnetic dipole moment per unit volume) of the medium.

Here $\mathbf{E}(\mathbf{r},t)$ and $\mathbf{B}(\mathbf{r},t)$ are considered as the fundamental macroscopic electric and magnetic field quantities; $\mathbf{D}(\mathbf{r},t)$ and $\mathbf{H}(\mathbf{r},t)$ are the corresponding derived fields associated with the state of matter, connected to $\mathbf{E}(\mathbf{r},t)$ and $\mathbf{B}(\mathbf{r},t)$ through the electric polarization density $\mathbf{P}(\mathbf{r},t)$ and magnetization (magnetic polarization density) $\mathbf{M}(\mathbf{r},t)$ through the basic constitutive relations. In fact, the constitutive equations above form the very definitions[1] of the electric polarization density and magnetization.

---

[1] J. D. Jackson, *Classical Electrodynamics*, 2nd ed. (Wiley, New York, 1975); J. A. Stratton, *Electromagnetic Theory* (McGraw-Hill, New York, 1941).

**Two frequent assumptions in nonlinear optics**

- No free charges present,

$$\rho(\mathbf{r},t)=0, \qquad \mathbf{J}(\mathbf{r},t)=\mathbf{0}.$$

(Any relaxation processes etc. are included in imaginary parts of the terms of the electric susceptibility.)

- No magnetization of the medium,

$$\mathbf{M}(\mathbf{r},t)=\mathbf{0}.$$

**The wave equation**

By taking the cross product with the nabla operator and Faraday's law, one obtains

$$\begin{aligned}\nabla\times\nabla\times\mathbf{E}(\mathbf{r},t)&=-\frac{\partial}{\partial t}\nabla\times\mathbf{B}(\mathbf{r},t)\\&=-\mu_0\frac{\partial}{\partial t}\nabla\times\mathbf{H}(\mathbf{r},t)\\&=-\mu_0\frac{\partial}{\partial t}\frac{\partial\mathbf{D}(\mathbf{r},t)}{\partial t}\\&=-\mu_0\Big(\varepsilon_0\frac{\partial^2\mathbf{E}(\mathbf{r},t)}{\partial t^2}+\frac{\partial^2\mathbf{P}(\mathbf{r},t)}{\partial t^2}\Big).\end{aligned}$$

Since now $\mu_0\varepsilon_0=1/c^2$ in SI units, with $c$ being the speed of light in vacuum, one hence obtains the basic wave equation, taken in time domain, as

$$\nabla\times\nabla\times\mathbf{E}(\mathbf{r},t)+\frac{1}{c^2}\frac{\partial^2\mathbf{E}(\mathbf{r},t)}{\partial t^2}=-\mu_0\frac{\partial^2\mathbf{P}(\mathbf{r},t)}{\partial t^2}, \tag{1}$$

where, as in the previous lectures of this course, the polarization density can be written in terms of the perturbation series as

$$\mathbf{P}(\mathbf{r},t)=\sum_{k=1}^{\infty}\mathbf{P}^{(k)}(\mathbf{r},t)=\underbrace{\varepsilon_0\int_{-\infty}^{\infty}\chi^{(1)}_{\mu\alpha}(-\omega;\omega)E_\alpha(\mathbf{r},\omega)\exp(-i\omega t)\,d\omega}_{=\mathbf{P}^{(1)}(\mathbf{r},t)}+\underbrace{\sum_{k=2}^{\infty}\mathbf{P}^{(k)}(\mathbf{r},t)}_{=\mathbf{P}^{(\mathrm{NL})}(\mathbf{r},t)}$$

In the left hand side of Eq. (1), we find the part of the homogeneous wave equation for propagation of electromagnetic waves in vacuum, while the right hand side described the modifications to the vacuum propagation due to the interaction between light and matter. In this respect, it is now clear that the electric polarisation effectively acts as a source term in the mathematical description of electromagnetic wave propagation, making the otherwise homogeneous vacuum problem an inhomogeneous problem (though with known source terms).

It should be noticed that whenever the polarization density is calculated from the Bloch equations (formulated later on, in lecture 10 of this course), instead of by means of a perturbation series as above, the Maxwell equations and the wave equation (1) above are denoted *Maxwell-Bloch equations*. In some sense, we can therefore see the choice of method for the calculation of the polarization density as a switch point not only for using the susceptibility formalism or not for the description of interaction between light and matter, but also for the form of the wave propagation problem in nonlinear media, which mathematically significantly differ between the "pure" Maxwell's equations with susceptibilities and the Maxwell-Bloch equations.

## The wave equation in frequency domain (optional)

Frequently in this course, we have rather been studying the electric fields and polarisation densities in frequency domain, since many phenomena in optics are properly and conveniently described as static (in which case the frequency dependence is simply reduced to the interaction between discrete frequencies in the spectrum). By using the Fourier integral identity[2]

$$E_\alpha(t) = \int_{-\infty}^{\infty} E_\alpha(\omega)\exp(-i\omega t)\,d\omega = \mathfrak{F}^{-1}[E_\alpha](t),$$

with inverse relation

$$E_\alpha(\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} E_\alpha(\tau)\exp(i\omega\tau)\,d\tau = \mathfrak{F}[E_\alpha](\omega),$$

we obtain the wave equation (1) as

$$\nabla\times\nabla\times\mathbf{E}(\mathbf{r},\omega) - \frac{\omega^2}{c^2}\mathbf{E}(\mathbf{r},\omega) = \mu_0\omega^2\mathbf{P}(\mathbf{r},\omega).$$

## Quasimonochromatic light - Time dependent problems

By inserting the perturbation series for the electric polarisation density into the general wave equation (1), which apply to arbitrary electric field distributions and field intensities of the light, one obtains the equation

$$\nabla\times\nabla\times\mathbf{E}(\mathbf{r},t) + \underbrace{\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\int_{-\infty}^{\infty}\mathbf{e}_\mu\varepsilon_{\mu\alpha}(\omega)E_\alpha(\mathbf{r},\omega)\exp(-i\omega t)\,d\omega}_{\text{(denote this integral as } I \text{ for later use)}} = -\mu_0\frac{\partial^2\mathbf{P}^{(\mathrm{NL})}(\mathbf{r},t)}{\partial t^2}, \qquad (2)$$

where

$$\varepsilon_{\mu\alpha}(\omega) = \delta_{\mu\alpha} + \chi^{(1)}_{\mu\alpha}(-\omega;\omega)$$

is a parameter commonly denoted as the *relative electrical permittivity*.[3] This wave equation is identical to Eq. (7.14) in Butcher and Cotter's book. (Notice though the printing error in Butcher and Cotter's Eq. (7.14), where the first $\mu_0$ should be replaced by $1/c^2$.)

The second term of the left hand side of Eq. (2) gives all first order optical contributions to the wave propagation, as well as all linear optical dispersion effects. This terms deserves some extra attention, and we will now proceed with deriving the effect of the frequency dependence of the relative permittivity upon the wave equation. First of all, we notice that since $E_\alpha(\mathbf{r},-\omega) = E^*_\alpha(\mathbf{r},\omega)$, which simply is a consequence of the choice of complex Fourier transform of a real valued field, the reality condition of Eq. (2) requires that

$$\varepsilon_{\mu\alpha}(-\omega) = \varepsilon^*_{\mu\alpha}(\omega).$$

---

[2] From the inverse Fourier integral identity, it follows that the Fourier transform of a derivative of a function $f(t)$ is

$$\mathfrak{F}[f'(t)](\omega) = -i\omega\,\mathfrak{F}[f(t)](\omega) \qquad \Rightarrow \qquad \mathfrak{F}[f''(t)](\omega) = -\omega^2\,\mathfrak{F}[f(t)](\omega).$$

[3] Notice that for isotropic media, $\chi^{(1)}_{\mu\alpha}(-\omega;\omega) = \chi^{(1)}_{xx}(-\omega;\omega)\delta_{\mu\alpha}$, which leads to the simplified form

$$\mathbf{e}_\mu\varepsilon_{\mu\alpha}(\omega)E_\alpha(\mathbf{r},\omega) = \varepsilon(\omega)\mathbf{E}(\mathbf{r},\omega).$$

We will here, however, continue with the general form, in order not to loose generality in discussion that is to follow.

It should be emphasized that this property of the relative electrical permittivity merely is a convenient mathematical construction, since we in regular physical terms only consider positive angular frequencies as argument for the refractive index, etc.

For quasimonochromatic light, the electric field and polarisation density are taken as

$$\begin{aligned}
\mathbf{E}(\mathbf{r},t) &= \sum_{\omega_\sigma\geq 0} \mathrm{Re}[\mathbf{E}_{\omega_\sigma}(\mathbf{r},t)\exp(-i\omega_\sigma t)],\\
\mathbf{P}(\mathbf{r},t) &= \sum_{\omega_\sigma\geq 0} \mathrm{Re}[\mathbf{P}_{\omega_\sigma}(\mathbf{r},t)\exp(-i\omega_\sigma t)],
\end{aligned}$$

where $\mathbf{E}_{\omega_\sigma}(\mathbf{r},t)$ and $\mathbf{P}_{\omega_\sigma}(\mathbf{r},t)$ are slowly varying envelopes of the fields. In the frequency domain, the quasimonochromatic fields are expressed as

$$\begin{aligned}
\mathbf{E}(\mathbf{r},\omega) &= \frac{1}{2}\sum_{\omega_\sigma\geq 0} [\mathbf{E}_{\omega_\sigma}(\mathbf{r},\omega-\omega_\sigma)+\mathbf{E}^*_{\omega_\sigma}(\mathbf{r},-\omega-\omega_\sigma)],\\
\mathbf{P}(\mathbf{r},\omega) &= \frac{1}{2}\sum_{\omega_\sigma\geq 0} [\mathbf{P}_{\omega_\sigma}(\mathbf{r},\omega-\omega_\sigma)+\mathbf{P}^*_{\omega_\sigma}(\mathbf{r},-\omega-\omega_\sigma)],
\end{aligned}$$

where the envelopes have some limited extent around the carrier frequencies at $\pm\omega_\sigma$. Notice that the fields taken in the frequency domain are expressed entirely in terms of their respective temporal envelope, that is to say, without the exponential functions that appear in their counterparts in time domain.

For simplicity considering a medium that in the linear optical domain is isotropic, with the relative electrical permittivity

$$\varepsilon_{\mu\alpha}(\omega)=\varepsilon(\omega)\delta_{\mu\alpha}=n_0^2(\omega)\delta_{\mu\alpha},$$

where $n_0(\omega)$ is the first order contribution to the refractive index of the medium, this leads to the middle term of the wave equation (1) in the form

$$\begin{aligned}
I &\equiv \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\int_{-\infty}^{\infty}\mathbf{e}_\mu\varepsilon_{\mu\alpha}(\omega)E_\alpha(\mathbf{r},\omega)\exp(-i\omega t)\,d\omega\\
&= -\int_{-\infty}^{\infty}\frac{\omega^2 n^2(\omega)}{c^2}\underbrace{\frac{1}{2}\sum_{\omega_\sigma\geq 0}[\mathbf{E}_{\omega_\sigma}(\mathbf{r},\omega-\omega_\sigma)+\mathbf{E}^*_{\omega_\sigma}(\mathbf{r},-\omega-\omega_\sigma)]}_{\text{quasimonochromatic form of }\mathbf{E}(\mathbf{r},\omega)}\exp(-i\omega t)\,d\omega\\
&= \{\text{denote } \omega^2\varepsilon(\omega)/c^2\equiv\omega^2 n_0^2(\omega)/c^2\equiv k^2(\omega)\}\\
&= -\frac{1}{2}\sum_{\omega_\sigma\geq 0}\int_{-\infty}^{\infty}k^2(\omega)[\mathbf{E}_{\omega_\sigma}(\mathbf{r},\omega-\omega_\sigma)+\mathbf{E}^*_{\omega_\sigma}(\mathbf{r},-\omega-\omega_\sigma)]\exp(-i\omega t)\,d\omega.\\
&= -\frac{1}{2}\sum_{\omega_\sigma\geq 0}\int_{-\infty}^{\infty}k^2(\omega)\mathbf{E}_{\omega_\sigma}(\mathbf{r},\omega-\omega_\sigma)\exp(-i\omega t)\,d\omega+\text{c. c.}
\end{aligned}$$

If now the field envelopes decay to zero rapidly enough in the vicinity of the carrier frequencies (as we would expect for quasimonochromatic light, with a strong spectral confinement around the carrier frequency of the light), then we may expect that a good approximation is to make a Taylor expansion of $k^2(\omega)$, in the neighbourhood of respective carrier frequency of the light, as

$$\begin{aligned}
k^2(\omega) &\approx \Big(k(\omega_\sigma)+\frac{dk}{d\omega}\Big|_{\omega_\sigma}(\omega-\omega_\sigma)+\frac{1}{2!}\frac{d^2k}{d\omega^2}\Big|_{\omega_\sigma}(\omega-\omega_\sigma)^2\Big)^2\\
&\approx k_\sigma^2+2k_\sigma\frac{dk}{d\omega}\Big|_{\omega_\sigma}(\omega-\omega_\sigma)+k_\sigma\frac{d^2k}{d\omega^2}\Big|_{\omega_\sigma}(\omega-\omega_\sigma)^2,
\end{aligned}$$

where the notation $k_\sigma = k(\omega_\sigma)$ was introduced, and hence[4]

$$\begin{aligned}
I &\approx -\frac{1}{2}\sum_{\omega_\sigma\geq 0}\int_{-\infty}^{\infty}\Big(k_\sigma^2 + 2k_\sigma\frac{dk}{d\omega}\Big|_{\omega_\sigma}(\omega-\omega_\sigma) + k_\sigma\frac{d^2k}{d\omega^2}\Big|_{\omega_\sigma}(\omega-\omega_\sigma)^2\Big)\\
&\qquad\qquad\qquad\qquad \times \mathbf{E}_{\omega_\sigma}(\mathbf{r},\omega-\omega_\sigma)\exp(-i\omega t)\,d\omega + \text{c. c.}\\
&= \{\text{change variable of integration } \omega' = \omega-\omega_\sigma\}\\
&= -\frac{1}{2}\sum_{\omega_\sigma\geq 0}\int_{-\infty}^{\infty}\Big(k_\sigma^2 + 2k_\sigma\frac{dk}{d\omega}\Big|_{\omega_\sigma}\omega' + k_\sigma\frac{d^2k}{d\omega^2}\Big|_{\omega_\sigma}\omega'^2\Big)\\
&\qquad\qquad\qquad\qquad \times \mathbf{E}_{\omega_\sigma}(\mathbf{r},\omega')\exp(-i\omega' t)\,d\omega'\,\exp(-i\omega_\sigma t) + \text{c. c.}\\
&= \Big\{\text{use } \int_{-\infty}^{\infty}\omega^n f(\omega)\exp(-i\omega t)\,d\omega = \mathfrak{F}^{-1}[\omega^n f(\omega)](t) = i^n\frac{d^n f(t)}{dt^n}\Big\}\\
&= -\frac{1}{2}\sum_{\omega_\sigma\geq 0}\exp(-i\omega_\sigma t)\Big(k_\sigma^2 + i2k_\sigma\frac{dk}{d\omega}\Big|_{\omega_\sigma}\frac{\partial}{\partial t} - k_\sigma\frac{d^2k}{d\omega^2}\Big|_{\omega_\sigma}\frac{\partial^2}{\partial t^2}\Big)\mathbf{E}_{\omega_\sigma}(\mathbf{r},t) + \text{c. c.}
\end{aligned}$$

As this result is inserted back into the wave equation (2), one obtains

$$\begin{aligned}
&\frac{1}{2}\sum_{\omega_\sigma\geq 0}\exp(-i\omega_\sigma t)\Big[\nabla\times\nabla\times\mathbf{E}_{\omega_\sigma}(\mathbf{r},t)\\
&\qquad\qquad - \Big(k_\sigma^2 + 2ik_\sigma\frac{dk}{d\omega}\Big|_{\omega_\sigma}\frac{\partial}{\partial t} - k_\sigma\frac{d^2k}{d\omega^2}\Big|_{\omega_\sigma}\frac{\partial^2}{\partial t^2}\Big)\mathbf{E}_{\omega_\sigma}(\mathbf{r},t)\Big] + \text{c. c.}\\
&\qquad = -\mu_0\frac{\partial^2\mathbf{P}^{(\text{NL})}(\mathbf{r},t)}{\partial t^2}\\
&\qquad = -\mu_0\frac{\partial^2}{\partial t^2}\frac{1}{2}\sum_{\omega_\sigma\geq 0}\mathbf{P}^{(\text{NL})}_{\omega_\sigma}(\mathbf{r},t)\exp(-i\omega_\sigma t) + \text{c. c.}\\
&\qquad \approx \mu_0\frac{1}{2}\sum_{\omega_\sigma\geq 0}\omega_\sigma^2\mathbf{P}^{(\text{NL})}_{\omega_\sigma}(\mathbf{r},t)\exp(-i\omega_\sigma t) + \text{c. c.}
\end{aligned}$$

As we separate out the respective frequency components at $\omega = \omega_\sigma$ of this equation, one obtains the time dependent wave equation for the *temporal envelope components* of the electric field as

$$\nabla\times\nabla\times\mathbf{E}_{\omega_\sigma}(\mathbf{r},t) - \Big(k_\sigma^2 + i2k_\sigma\frac{1}{v_\text{g}}\frac{\partial}{\partial t} - k_\sigma\frac{d^2k}{d\omega^2}\Big|_{\omega_\sigma}\frac{\partial^2}{\partial t^2}\Big)\mathbf{E}_{\omega_\sigma}(\mathbf{r},t) = \mu_0\omega_\sigma^2\mathbf{P}^{(\text{NL})}_{\omega_\sigma}(\mathbf{r},t), \qquad (3)$$

where

$$v_\text{g} = \Big(\frac{dk}{d\omega}\Big|_{\omega_\sigma}\Big)^{-1}.$$

---

[4] Notice that unless we apply the second approximation in the Taylor expansion of $k^2(\omega)$, terms containing the *squares* of the derivatives will appear, which will lead to wave equations that differ from the ones given by Butcher and Cotter. In particular, this situation will arise even if one uses the suggested expansion given by Eq. (7.23) in Butcher and Cotter's book, which hence should be taken with some care if one wish to build a strict foundation for the time-dependent wave equation.

**Three practical approximations**

[1] The infinite plane wave approximation,

$$\mathbf{E}_{\omega_\sigma}(\mathbf{r},t)=\mathbf{E}_{\omega_\sigma}(z,t)\perp\mathbf{e}_z \qquad \Rightarrow \qquad \nabla\times\nabla\times\rightarrow-\frac{\partial^2}{\partial z^2}.$$

[2] Unidirectional propagation,

$$\begin{gathered}\mathbf{E}_{\omega_\sigma}(z,t)=\mathbf{A}_{\omega_\sigma}(z,t)\exp(\pm ik_\sigma z)\\ \Downarrow\\ \nabla\times\nabla\times\mathbf{E}_{\omega_\sigma}(z,t)=-[\frac{\partial^2\mathbf{A}_{\omega_\sigma}}{\partial z^2}\pm 2ik_\sigma\frac{\partial\mathbf{A}_{\omega_\sigma}}{\partial z}-k_\sigma^2\mathbf{A}_{\omega_\sigma}]\exp(\pm ik_\sigma z),\end{gathered}$$

for waves propagating in the positive/negative $z$-direction. In this case, the real-valued electric field hence takes the form

$$\begin{aligned}\mathbf{E}(\mathbf{r},t)&=\sum_{\omega_\sigma\geq 0}\mathrm{Re}[\mathbf{E}_{\omega_\sigma}(\mathbf{r},t)\exp(-i\omega_\sigma t)]\\ &=\sum_{\omega_\sigma\geq 0}\mathrm{Re}[\mathbf{A}_{\omega_\sigma}(z,t)\exp(\pm ik_\sigma z-i\omega_\sigma t)]\\ &=\sum_{\omega_\sigma\geq 0}|\mathbf{A}_{\omega_\sigma}(z,t)|\,\mathrm{Re}\{\exp[ik_\sigma z\mp i\omega_\sigma t+i\phi(z)]\}\\ &=\sum_{\omega_\sigma\geq 0}|\mathbf{A}_{\omega_\sigma}(z,t)|\cos(k_\sigma z\mp\omega_\sigma t+\phi(z)),\end{aligned}$$

where $\phi(z,t)$ describes the spatially and temporally varying phase of the complex-valued slowly varying envelope function $\mathbf{A}_{\omega_\sigma}(z,t)$ of the electric field.

[3] The slowly varying envelope approximation,

$$\Big|\frac{\partial^2\mathbf{A}_{\omega_\sigma}}{\partial z^2}\Big|\ll\Big|k_\sigma\frac{\partial\mathbf{A}_{\omega_\sigma}}{\partial z}\Big|.$$

These approximations, whenever applicable, further reduce the time dependent wave equation to

$$\Big(\pm i\frac{\partial}{\partial z}+i\frac{1}{v_{\mathrm{g}}}\frac{\partial}{\partial t}-\frac{1}{2}\frac{d^2k}{d\omega^2}\Big|_{\omega_\sigma}\frac{\partial^2}{\partial t^2}\Big)\mathbf{A}_{\omega_\sigma}(z,t)=-\frac{\mu_0\omega_\sigma^2}{2k_\sigma}\mathbf{P}^{(\mathrm{NL})}_{\omega_\sigma}(\mathbf{r},t)\exp(\mp ik_\sigma z). \tag{4}$$

This form of the wave equation is identical to Butcher and Cotter's Eq. (7.24), with the exception that here waves propagating in positive (upper signs) as well as negative (lower signs) $z$-direction are considered.

**Monochromatic light**

*Monochromatic optical field*

$$\mathbf{E}(\mathbf{r},t) = \sum_{\sigma} \mathrm{Re}[\mathbf{E}_{\omega_\sigma}(\mathbf{r}) \exp(-i\omega_\sigma t)], \qquad \omega_\sigma \geq 0$$

$$\mathbf{E}(\mathbf{r},\omega) = \frac{1}{2} \sum_{\sigma} [\mathbf{E}_{\omega_\sigma}(\mathbf{r})\delta(\omega - \omega_\sigma) + \mathbf{E}^*_{\omega_\sigma}(\mathbf{r})\delta(\omega + \omega_\sigma)]$$

*Polarization density induced by monochromatic optical field*

$$\mathbf{P}^{(n)}(\mathbf{r},t) = \sum_{\omega_\sigma \geq 0} \mathrm{Re}[\mathbf{P}^{(n)}_{\omega_\sigma} \exp(-i\omega_\sigma t)], \qquad \omega_\sigma = \omega_1 + \omega_2 + \ldots + \omega_n$$

(For construction of $\mathbf{P}^{(n)}_{\omega_\sigma}$, see notes on the Butcher and Cotter convention handed out during the third lecture.)

**Monochromatic light - Time independent problems**

For strictly monochromatic light, as for example the output light of continuous wave lasers, the temporal field envelopes are constants in time, and the wave equation (3) is reduced to

$$\nabla \times \nabla \times \mathbf{E}_{\omega_\sigma}(\mathbf{r}) - k_\sigma^2 \mathbf{E}_{\omega_\sigma}(\mathbf{r}) = \mu_0 \omega_\sigma^2 \mathbf{P}^{(\mathrm{NL})}_{\omega_\sigma}(\mathbf{r}). \tag{5}$$

By applying the above listed approximations, one immediately finds the monochromatic, time independent form of Eq. (3) in the infinite plane wave limit and slowly varying approximation as

$$\frac{\partial}{\partial z}\mathbf{A}_{\omega_\sigma} = \pm i \frac{\mu_0 \omega_\sigma^2}{2k_\sigma} \mathbf{P}^{(\mathrm{NL})}_{\omega_\sigma} \exp(\mp i k_\sigma z), \tag{6}$$

where the upper/lower sign correspond to a wave propagating in the positive/negative $z$-direction. This equation corresponds to Butcher and Cotter's Eq. (7.17).

**Example I: Optical Kerr-effect - Time independent case**

In this example, we consider continuous wave propagation[5] in optical Kerr-media, using light polarized in the $x$-direction and propagating along the positive direction of the $z$-axis,

$$\mathbf{E}(\mathbf{r},t)=\mathrm{Re}[\mathbf{E}_\omega(z)\exp(-i\omega t)],\qquad \mathbf{E}_\omega(z)=\mathbf{A}_\omega(z)\exp(ikz)=\mathbf{e}_x A^x_\omega(z)\exp(ikz),$$

where, as previously, $k=\omega n_0/c$. From material handed out during the third lecture (notes on the Butcher and Cotter convention), the nonlinear polarization density for $x$-polarized light is given as $\mathbf{P}^{(\mathrm{NL})}_\omega=\mathbf{P}^{(3)}_\omega$, with

$$\begin{aligned}\mathbf{P}^{(3)}_\omega&=\varepsilon_0(3/4)\mathbf{e}_x\chi^{(3)}_{xxxx}(-\omega;\omega,\omega,-\omega)|E^x_\omega|^2E^x_\omega\\&=\varepsilon_0(3/4)\chi^{(3)}_{xxxx}|\mathbf{E}_\omega|^2\mathbf{E}_\omega\\&=\varepsilon_0(3/4)\chi^{(3)}_{xxxx}|\mathbf{A}_\omega|^2\mathbf{A}_\omega\exp(ikz),\end{aligned}$$

and the time independent wave equation for the field envelope $\mathbf{A}_\omega$, using Eq. (6), becomes

$$\begin{aligned}\frac{\partial}{\partial z}\mathbf{A}_\omega&=i\frac{\mu_0\omega^2}{2k}\underbrace{\varepsilon_0(3/4)\chi^{(3)}_{xxxx}|\mathbf{A}_\omega|^2\mathbf{A}_\omega\exp(ikz)}_{=\mathbf{P}^{(\mathrm{NL})}_\omega(z)}\exp(-ikz)\\&=i\frac{3\omega^2}{8c^2k}\chi^{(3)}_{xxxx}|\mathbf{A}_\omega|^2\mathbf{A}_\omega\\&=\{\text{since } k=\omega n_0(\omega)/c\}\\&=i\frac{3\omega}{8cn_0}\chi^{(3)}_{xxxx}|\mathbf{A}_\omega|^2\mathbf{A}_\omega\\&=\{\text{in analogy with Butcher and Cotter Eq. (6.63), } n_2=(3/8n_0)\chi^{(3)}_{xxxx}\}\\&=i\frac{\omega n_2}{c}|\mathbf{A}_\omega|^2\mathbf{A}_\omega,\end{aligned}$$

or, equivalently, in its scalar form

$$\frac{\partial}{\partial z}A^x_\omega=i\frac{\omega n_2}{c}|A^x_\omega|^2A^x_\omega.$$

If the medium of interest now is analyzed at an angular frequency far from any resonance, we may look for solutions to this equation with $|\mathbf{A}_\omega|$ being constant (for a lossless medium). For such a case it is straightforward to integrate the final wave equation to yield the general solution

$$\mathbf{A}_\omega(z)=\mathbf{A}_\omega(z_0)\exp[i\omega n_2|\mathbf{A}_\omega(z_0)|^2z/c],$$

or, again equivalently, in the scalar form

$$A^x_\omega(z)=A^x_\omega(z_0)\exp[i\omega n_2|A^x_\omega(z_0)|^2z/c],$$

which hence gives the solution for the real-valued electric field $\mathbf{E}(\mathbf{r},t)$ as

$$\begin{aligned}\mathbf{E}(\mathbf{r},t)&=\mathrm{Re}[\mathbf{E}_\omega(z)\exp(-i\omega t)]\\&=\mathrm{Re}\{\mathbf{A}_\omega(z)\exp[i(kz-\omega t)]\}\\&=\mathrm{Re}\{\mathbf{A}_\omega(z_0)\exp[i(kz+\omega n_2|\mathbf{A}_\omega(z_0)|^2z/c-\omega t)]\}.\end{aligned}$$

From this solution, one immediately finds that the wave propagates with an effective propagation constant

$$k+\omega n_2|\mathbf{A}_\omega(z_0)|^2/c=(\omega/c)(n_0+n_2|\mathbf{A}_\omega(z_0)|^2),$$

that is to say, experiencing the intensity dependent refractive index

$$n_{\mathrm{eff}}=n_0+n_2|\mathbf{A}_\omega(z_0)|^2.$$

---

[5] That is to say, a time independent problem with the temporal envelope of the electrical field being constant in time.

**Example II: Optical Kerr-effect - Time dependent case**

We now consider a *time dependent* envelope $\mathbf{E}_\omega(z,t)$, of an optical wave propagating in the same medium and geometry as in the previous example, for which now

$$\mathbf{E}(\mathbf{r},t)=\mathrm{Re}[\mathbf{E}_\omega(z,t)\exp(-i\omega t)],\qquad \mathbf{E}_\omega(z,t)=\mathbf{A}_\omega(z,t)\exp(ikz)=\mathbf{e}_x A^x_\omega(z,t)\exp(ikz).$$

The proper wave equation to apply for this case is the time dependent wave equation (4), and since the nonlinear polarization density of the medium still is given by the optical Kerr-effect, we obtain

$$\begin{aligned}
\Big(i\frac{\partial}{\partial z}&+i\frac{1}{v_{\mathrm{g}}}\frac{\partial}{\partial t}-\frac{1}{2}\frac{d^2k}{d\omega^2}\Big|_{\omega_\sigma}\frac{\partial^2}{\partial t^2}\Big)\mathbf{A}_\omega(z,t)\\
&=-\frac{\mu_0\omega^2}{2k}\underbrace{\varepsilon_0(3/4)\chi^{(3)}_{xxxx}|\mathbf{A}_\omega(z,t)|^2\mathbf{A}_\omega(z,t)\exp(ikz)}_{=\mathbf{P}^{(\mathrm{NL})}_\omega(z,t)}\exp(-ikz)\\
&=-\frac{3\omega^2}{8c^2k}\chi^{(3)}_{xxxx}|\mathbf{A}_\omega(z,t)|^2\mathbf{A}_\omega(z,t)\\
&=\{\text{as in previous example}\}\\
&=-\frac{\omega n_2}{c}|\mathbf{A}_\omega(z,t)|^2\mathbf{A}_\omega(z,t).
\end{aligned}$$

The resulting wave equation

$$\Big(i\frac{\partial}{\partial z}+i\frac{1}{v_{\mathrm{g}}}\frac{\partial}{\partial t}-\frac{1}{2}\frac{d^2k}{d\omega^2}\Big|_{\omega_\sigma}\frac{\partial^2}{\partial t^2}\Big)\mathbf{A}_\omega=-\frac{\omega n_2}{c}|\mathbf{A}_\omega|^2\mathbf{A}_\omega$$

is the starting point for analysis of solitons and solitary waves in optical Kerr-media. The obtained equation is the non-normalized form of the in nonlinear physics (not only nonlinear optics!) often encountered *nonlinear Schrödinger equation* (or NLSE, as its common acronym yields).

For a discussion on the transformation that cast the nonlinear Schrödinger equation into the *normalized nonlinear Schrödinger equation*, see Butcher and Cotter's book, page 240.

# Lecture IX

**Lecture Notes on Nonlinear Optics**
Nonlinear Optics (5A5513, 5p for advanced undergraduate and doctoral students)
Course given at the Royal Institute of Technology,
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## LECTURE 9

In this lecture, we will focus on examples of electromagnetic wave propagation in nonlinear optical media, by applying the forms of Maxwell's equations as obtained in the eighth lecture to a set of particular nonlinear interactions as described by the previously formulated nonlinear susceptibility formalism.

The outline for this lecture is:

- General process for solving problems in nonlinear optics
- Second harmonic generation (SHG)
- Optical Kerr-effect

### General process for solving problems in nonlinear optics

The typical steps in the process of solving a theoretical problem in nonlinear optics typically involve:

$$\left\{\begin{matrix}\text{define the optical interaction of interest}\\ \text{(identifying the susceptibility)}\end{matrix}\right\}$$

$$\Downarrow$$

$$\left\{\begin{matrix}\text{define in which medium the interaction take place}\\ \text{(identify crystallographic point symmetry group)}\end{matrix}\right\}$$

$$\Downarrow$$

$$\left\{\begin{matrix}\text{consider eventual additional symmetries and constraints}\\ \text{(e. g. intrinsic, overall, or Kleinman symmetries)}\end{matrix}\right\}$$

$$\Downarrow$$

$$\left\{\begin{matrix}\text{construct the polarization density}\\ \text{(the Butcher and Cotter convention)}\end{matrix}\right\}$$

$$\Downarrow$$

$$\left\{\begin{matrix}\text{formulate the proper wave equation}\\ \text{(e. g. taking dispersion or diffraction into account)}\end{matrix}\right\}$$

$$\Downarrow$$

$$\left\{\begin{matrix}\text{formulate the proper boundary}\\ \text{conditions for the wave equation}\end{matrix}\right\}$$

$$\Downarrow$$

$$\left\{\begin{matrix}\text{solve the wave equation under}\\ \text{the boundary conditions}\end{matrix}\right\}$$

### Formulation of the exercises in this lecture

In order to illustrate the scheme as previously outlined, the following exercises serve as to give the connection between the susceptibilities, as extensively analysed from a quantum-mechanical basis in earlier lectures of this course, and the wave equation, derived from Maxwell's equations of motion for electromagnetic fields.

**Exercise 1.** (*Second harmonic generation in negative uniaxial media*) Consider a continuous pump wave at angular frequency $\omega$, initially polarized in the $y$-direction and propagating in the positive $x$-direction of a negative uniaxial crystal of crystallographic point symmetry group $3m$. (Examples of crystals belonging to this class: beta-$BaB_2O_4$/BBO, $LiNbO_3$.)

**1a.** Formulate the polarization density of the medium for the pump and second harmonic wave.

**1b.** Formulate the system of equations of motion for the electromagnetic fields.

**1c.** Assuming no second harmonic signal present at the input, solve the equations of motion for the second harmonic field, using the non-depleted pump approximation, and derive an expression for the conversion efficiency of the second harmonic generation.

**Exercise 2.** (*Optical Kerr-effect – continuous wave case*) In this setup, a monochromatic optical wave is propagating in the positive $z$-direction of an isotropic optical Kerr-medium.

**2a.** Formulate the polarization density of the medium for a wave polarized in the $xy$-plane.

**2b.** Formulate the polarization density of the medium for a wave polarized in the $x$-direction.

**2c.** Formulate the wave equation for continuous wave propagation in optical Kerr-media. The continuous wave is $x$-polarized and propagates in the positive $z$-direction.

**2d.** For lossless media, solve the wave equation and give an expression for the nonlinear, intensity-dependent refractive index $n = n_0 + n_2|\mathbf{E}_\omega|^2$.

## Second harmonic generation

*The optical interaction*

In the case of second harmonic generation (SHG), two photons at angular frequency $\omega$ combine to a photon at twice the angular frequency,

$$\hbar\omega + \hbar\omega \rightarrow \hbar(2\omega).$$

This interaction is for the second harmonic wave (at angular frequency $\omega$)described by the second order susceptibility

$$\chi^{(2)}_{\mu\alpha\beta}(-\omega_\sigma;\omega,\omega),$$

where $\omega_\sigma = 2\omega$ is the generated second harmonic frequency of the light.

*Symmetries of the medium*

In this example we consider second harmonic generation in trigonal media of crystallographic point symmetry group $3m$. (Example: $\text{LiNbO}_3$)

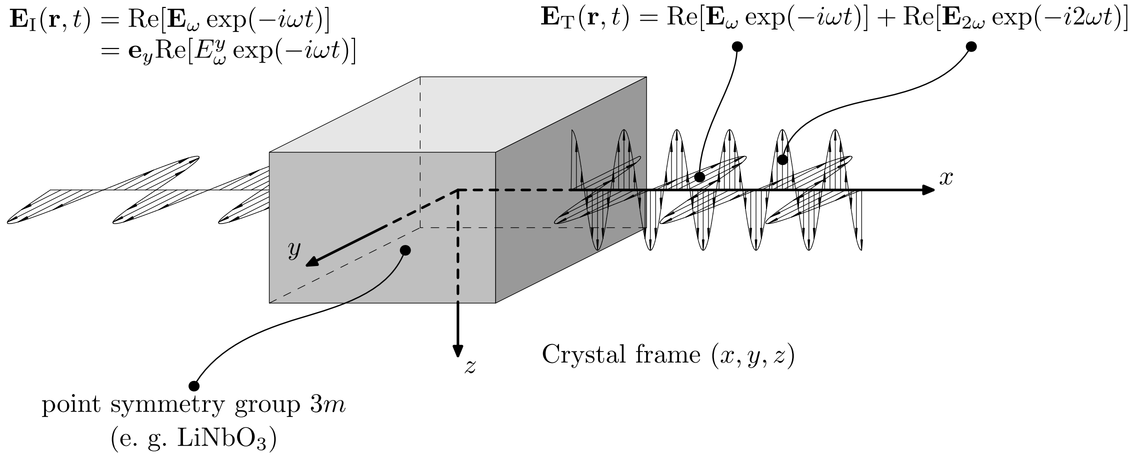

Figure 1. The setup for optical second harmonic generation in $\text{LiNbO}_3$.

For this point symmetry group, the nonzero tensor elements of the first order susceptibility are (for example according to Table A3.1 in *The Elements of Nonlinear Optics*)

$$\chi^{(1)}_{xx} = \chi^{(1)}_{yy}, \qquad \chi^{(1)}_{zz},$$

which gives the ordinary refractive indices

$$n_x(\omega) = n_y(\omega) = [1 + \chi^{(1)}_{xx}(-\omega;\omega)]^{1/2} \equiv n_\mathrm{O}(\omega)$$

for waves components polarized in the $x$- or $y$-directions, and the extraordinary refractive index

$$n_z(\omega) = [1 + \chi^{(1)}_{zz}(-\omega;\omega)]^{1/2} \equiv n_\mathrm{E}(\omega)$$

for the wave component polarized in the $z$-direction. Since we here are considering a negatively uniaxial crystal (see Butcher and Cotter, p. 214), these refractive indices satisfy the inequality

$$n_\mathrm{E}(\omega) \le n_\mathrm{O}(\omega).$$

The nonzero tensor elements of the second order susceptibility are (for example according to Table A3.2 in *The Elements of Nonlinear Optics*)

$$\begin{aligned} &\chi^{(2)}_{yxx} = \chi^{(2)}_{xyx} = \chi^{(2)}_{xxy} = -\chi^{(2)}_{yyy}, \qquad \chi^{(2)}_{zzz}, \\ &\chi^{(2)}_{zxx} = \chi^{(2)}_{zyy}, \qquad \chi^{(2)}_{yyz} = \chi^{(2)}_{xxz}, \qquad \chi^{(2)}_{yzy} = \chi^{(2)}_{xzx}, \end{aligned} \tag{1}$$

*Additional symmetries*

Intrinsic permutation symmetry for the case of second harmonic generation gives

$$\chi^{(2)}_{xxz}(-2\omega;\omega,\omega)=\chi^{(2)}_{xzx}(-2\omega;\omega,\omega)=\chi^{(2)}_{yzy}(-2\omega;\omega,\omega)=\chi^{(2)}_{yyz}(-2\omega;\omega,\omega),$$

which reduces the second order susceptibility in Eq. (1) to a set of 11 tensor elements, of which only 4 are independent. (We recall that the intrinsic permutation symmetry is always applicable, as being a consequence of the symmetrization described in lectures two and five.) Whenever Kleinman symmetry holds, the susceptibility is in addition symmetric under any permutation of the indices, which hence gives the additional relation

$$\chi^{(2)}_{zxx}(-2\omega;\omega,\omega)=\chi^{(2)}_{xzx}(-2\omega;\omega,\omega)=\chi^{(2)}_{xxz}(-2\omega;\omega,\omega),$$

i. e. reducing the second order susceptibility to a set of 11 tensor elements, of which only 3 are independent.

To summarize, the set of nonzero tensor elements describing second harmonic generation under Kleinman symmetry is

$$\begin{aligned}&\chi^{(2)}_{yxx}=\chi^{(2)}_{xyx}=\chi^{(2)}_{xxy}=-\chi^{(2)}_{yyy}, \qquad \chi^{(2)}_{zzz},\\ &\chi^{(2)}_{zxx}=\chi^{(2)}_{zyy}=\chi^{(2)}_{yyz}=\chi^{(2)}_{xxz}=\chi^{(2)}_{yzy}=\chi^{(2)}_{xzx}.\end{aligned} \tag{2}$$

For the pump field at angular frequency $\omega$, the relevant susceptibility describing the interaction with the second harmonic wave is[1]

$$\chi^{(2)}_{\mu\alpha\beta}(-\omega;2\omega,-\omega).$$

For an arbitrary frequency argument, this is the proper form of the susceptibility to use for the fundamental field, and this form generally differ from that of the susceptibilities for the second harmonic field. However, whenever Kleinman symmetry holds, the susceptibility for the fundamental field can be cast into the same parameters as for the second harmonic field, since

$$\begin{aligned}\chi^{(2)}_{\mu\alpha\beta}(-\omega;2\omega,-\omega)&=\big\{\text{Apply overall permutation symmetry}\big\}\\ &=\chi^{(2)}_{\alpha\mu\beta}(2\omega;-\omega,-\omega)\\ &=\big\{\text{Apply Kleinman symmetry}\big\}\\ &=\chi^{(2)}_{\mu\alpha\beta}(2\omega;-\omega,-\omega)\\ &=\big\{\text{Apply reality condition [B. \&C. Eq. (2.43)]}\big\}\\ &=[\chi^{(2)}_{\mu\alpha\beta}(-2\omega;\omega,\omega)]^*\\ &=\chi^{(2)}_{\mu\alpha\beta}(-2\omega;\omega,\omega).\end{aligned}$$

Hence the second order interaction is described by the same set of tensor elements for the fundamental as well as the second harmonic optical wave whenever Kleinman symmetry applies.

---

[1] Keep in mind that in the convention of Butcher and Cotter, the frequency arguments to the right of the semicolon may be writen in arbitrary order, hence we may in an equal description instead use

$$\chi^{(2)}_{xxz}(-\omega;-\omega,2\omega)$$

for the description of the second order interaction between light and matter.

*The polarization density*

Following the convention of Butcher and Cotter,[2] the degeneracy factor for the second harmonic signal at $2\omega$ is

$$K(-2\omega;\omega,\omega) = 2^{l+m-n}p,$$

where

$$\begin{aligned} p &= \{\text{the number of } \textit{distinct} \text{ permutations of } \omega,\omega\} = 1,\\ n &= \{\text{the order of the nonlinearity}\} = 2,\\ m &= \{\text{the number of angular frequencies } \omega_k \text{ that are zero}\} = 0,\\ l &= \left\{ \begin{array}{ll} 1, & \text{if } 2\omega \neq 0,\\ 0, & \text{otherwise.} \end{array} \right\} = 1, \end{aligned}$$

i. e.

$$K(-2\omega;\omega,\omega) = 2^{1+0-2} \times 1 = 1/2.$$

For the fundamental optical field at $\omega$, one might be mislead to assume that since the second order interaction for this field is described by an identical set of tensor elements as for the second harmonic wave, the degeneracy factor must also be identical to the previously derived one. This is, however, *a very wrong assumption*, and one can easily verify that the proper degeneracy factor for the fundamental field instead is given as

$$K(-\omega;2\omega,-\omega) = 2^{l+m-n}p,$$

where

$$\begin{aligned} p &= \{\text{the number of } \textit{distinct} \text{ permutations of } 2\omega,-\omega\} = 2,\\ n &= \{\text{the order of the nonlinearity}\} = 2,\\ m &= \{\text{the number of angular frequencies } \omega_k \text{ that are zero}\} = 0,\\ l &= \left\{ \begin{array}{ll} 1, & \text{if } \omega \neq 0,\\ 0, & \text{otherwise.} \end{array} \right\} = 1, \end{aligned}$$

i. e.

$$K(-\omega;2\omega,-\omega) = 2^{1+0-2} \times 2 = 1.$$

The general second harmonic polarization density of the medium is hence given as

$$\begin{aligned} [\mathbf{P}^{(\mathrm{NL})}_{2\omega}]_z = [\mathbf{P}^{(2)}_{2\omega}]_z &= \varepsilon_0 \underbrace{K(-2\omega;\omega,\omega)\chi^{(2)}_{z\alpha\beta}(-2\omega;\omega,\omega)}_{=\frac{1}{2}\chi^{(2)}_{z\alpha\beta}(-2\omega;\omega,\omega)} E^{\alpha}_{\omega}E^{\beta}_{\omega}\\ &= (\varepsilon_0/2)[\chi^{(2)}_{zxx}E^x_{\omega}E^x_{\omega} + \chi^{(2)}_{zyy}E^y_{\omega}E^y_{\omega} + \chi^{(2)}_{zzz}E^z_{\omega}E^z_{\omega}]\\ &= (\varepsilon_0/2)[\chi^{(2)}_{zxx}(E^x_{\omega}E^x_{\omega} + E^y_{\omega}E^y_{\omega}) + \chi^{(2)}_{zzz}E^z_{\omega}E^z_{\omega}],\\ [\mathbf{P}^{(\mathrm{NL})}_{2\omega}]_y &= (\varepsilon_0/2)[\chi^{(2)}_{yxx}E^x_{\omega}E^x_{\omega} + \chi^{(2)}_{yyy}E^y_{\omega}E^y_{\omega} + \chi^{(2)}_{yyz}E^y_{\omega}E^z_{\omega} + \chi^{(2)}_{yzy}E^z_{\omega}E^y_{\omega}]\\ &= (\varepsilon_0/2)[\chi^{(2)}_{yxx}(E^x_{\omega}E^x_{\omega} - E^y_{\omega}E^y_{\omega}) + \chi^{(2)}_{zxx}(E^y_{\omega}E^z_{\omega} + E^z_{\omega}E^y_{\omega})],\\ [\mathbf{P}^{(\mathrm{NL})}_{2\omega}]_x &= (\varepsilon_0/2)[\chi^{(2)}_{xxy}E^x_{\omega}E^y_{\omega} + \chi^{(2)}_{xyx}E^y_{\omega}E^x_{\omega} + \chi^{(2)}_{xxz}E^x_{\omega}E^z_{\omega} + \chi^{(2)}_{xzx}E^z_{\omega}E^x_{\omega}]\\ &= (\varepsilon_0/2)[\chi^{(2)}_{yxx}(E^x_{\omega}E^y_{\omega} + E^y_{\omega}E^x_{\omega}) + \chi^{(2)}_{zxx}(E^x_{\omega}E^z_{\omega} + E^z_{\omega}E^x_{\omega})], \end{aligned}$$

---

[2] See course material on the Butcher and Cotter convention handed out during the third lecture. Notice that for the first order polarization density, one at optical frequencies *always* has the trivial degeneracy factor

$$K(-2\omega;\omega) = 2^{l+m-n}p = 2^{1+0-1} \times 1 = 1.$$

while the general polarization density at the angular frequency of the pump field becomes[3]

$$
\begin{aligned}
[\mathbf{P}^{(\mathrm{NL})}_{\omega}]_z &= [\mathbf{P}^{(2)}_{\omega}]_z = \varepsilon_0 \underbrace{K(-\omega;2\omega,-\omega)\chi^{(2)}_{z\alpha\beta}(-\omega;2\omega,-\omega)}_{=\chi^{(2)}_{z\alpha\beta}(-2\omega;\omega,\omega)} E^{\alpha}_{2\omega}E^{\beta}_{-\omega}\\
&= \varepsilon_0[\chi^{(2)}_{zxx}E^{x}_{2\omega}E^{x*}_{\omega} + \chi^{(2)}_{zyy}E^{y}_{2\omega}E^{y*}_{\omega} + \chi^{(2)}_{zzz}E^{z}_{2\omega}E^{z*}_{\omega}]\\
&= \varepsilon_0[\chi^{(2)}_{zxx}(E^{x}_{2\omega}E^{x*}_{\omega} + E^{y}_{2\omega}E^{y*}_{\omega}) + \chi^{(2)}_{zzz}E^{z}_{2\omega}E^{z*}_{\omega}],\\
[\mathbf{P}^{(\mathrm{NL})}_{\omega}]_y &= \varepsilon_0[\chi^{(2)}_{yxx}E^{x}_{2\omega}E^{x*}_{\omega} + \chi^{(2)}_{yyy}E^{y}_{2\omega}E^{y*}_{\omega} + \chi^{(2)}_{yyz}E^{y}_{2\omega}E^{z*}_{\omega} + \chi^{(2)}_{yzy}E^{z}_{2\omega}E^{y*}_{\omega}]\\
&= \varepsilon_0[\chi^{(2)}_{yxx}(E^{x}_{2\omega}E^{x*}_{\omega} - E^{y}_{2\omega}E^{y*}_{\omega}) + \chi^{(2)}_{zxx}(E^{y}_{2\omega}E^{z*}_{\omega} + E^{z}_{2\omega}E^{y*}_{\omega})],\\
[\mathbf{P}^{(\mathrm{NL})}_{\omega}]_x &= \varepsilon_0[\chi^{(2)}_{xxy}E^{x}_{2\omega}E^{y*}_{\omega} + \chi^{(2)}_{xyx}E^{y}_{2\omega}E^{x*}_{\omega} + \chi^{(2)}_{xxz}E^{x}_{2\omega}E^{z*}_{\omega} + \chi^{(2)}_{xzx}E^{z}_{2\omega}E^{x*}_{\omega}]\\
&= \varepsilon_0[\chi^{(2)}_{yxx}(E^{x}_{2\omega}E^{y*}_{\omega} + E^{y}_{2\omega}E^{x*}_{\omega}) + \chi^{(2)}_{zxx}(E^{x}_{2\omega}E^{z*}_{\omega} + E^{z}_{2\omega}E^{x*}_{\omega})].
\end{aligned}
$$

For a pump wave polarized in the $yz$-plane of the crystal frame, the polarization density of the medium hence becomes

$$
\begin{aligned}
[\mathbf{P}^{(\mathrm{NL})}_{2\omega}]_z &= (\varepsilon_0/2)[\chi^{(2)}_{zxx}E^{y}_{\omega}E^{y}_{\omega} + \chi^{(2)}_{zzz}E^{z}_{\omega}E^{z}_{\omega}],\\
[\mathbf{P}^{(\mathrm{NL})}_{2\omega}]_y &= (\varepsilon_0/2)[-\chi^{(2)}_{yxx}E^{y}_{\omega}E^{y}_{\omega} + \chi^{(2)}_{zxx}(E^{y}_{\omega}E^{z}_{\omega} + E^{z}_{\omega}E^{y}_{\omega})],\\
[\mathbf{P}^{(\mathrm{NL})}_{2\omega}]_x &= 0,
\end{aligned}
$$

and

$$
\begin{aligned}
[\mathbf{P}^{(\mathrm{NL})}_{\omega}]_z &= \varepsilon_0[\chi^{(2)}_{zxx}E^{y}_{2\omega}E^{y*}_{\omega} + \chi^{(2)}_{zzz}E^{z}_{2\omega}E^{z*}_{\omega}],\\
[\mathbf{P}^{(\mathrm{NL})}_{\omega}]_y &= \varepsilon_0[-\chi^{(2)}_{yxx}E^{y}_{2\omega}E^{y*}_{\omega} + \chi^{(2)}_{zxx}(E^{y}_{2\omega}E^{z*}_{\omega} + E^{z}_{2\omega}E^{y*}_{\omega})],\\
[\mathbf{P}^{(\mathrm{NL})}_{\omega}]_x &= 0.
\end{aligned}
$$

*The wave equation*

Strictly speaking, the previously formulated polarization density gives a coupled system between the polarization states of both the fundamental and second harmonic waves, since both the $y$- and $z$-components of the polarization densities at $\omega$ and $2\omega$ contain components of all other field components. However, for simplicity we will here restrict the continued analysis to the case of a $y$-polarized input pump wave, which through the $\chi^{(2)}_{zyy} = \chi^{(2)}_{zxx}$ elements give rise to a $z$-polarized second harmonic frequency component at $2\omega$.

The electric fields of the fundamental and second harmonic optical waves are for the forward propagating configuration expressed in their *spatial envelopes* $\mathbf{A}_{\omega}$ and $\mathbf{A}_{2\omega}$ as

$$
\begin{aligned}
\mathbf{E}_{\omega}(x) &= \mathbf{e}_y A^{y}_{\omega}(x)\exp(ik_{\omega_y}x), &\qquad k_{\omega_y} &\equiv \omega n_{\omega_y}/c \equiv \omega n_{\mathrm{O}}(\omega)/c\\
\mathbf{E}_{2\omega}(x) &= \mathbf{e}_z A^{z}_{2\omega}(x)\exp(ik_{2\omega_z}x), &\qquad k_{2\omega_z} &\equiv 2\omega n_{2\omega_z}/c \equiv 2\omega n_{\mathrm{E}}(2\omega)/c
\end{aligned}
$$

Using the above separation of the natural, spatial oscillation of the light, in the infinite plane wave approximation and by using the slowly varying envelope approximation, the wave equation for the envelope of the second harmonic optical field becomes (see Eq. (6) in the notes from lecture eight)

$$
\begin{aligned}
\frac{\partial A^{z}_{2\omega}}{\partial x} &= i\frac{\mu_0(2\omega)^2}{2k_{2\omega_z}}[\mathbf{P}^{(\mathrm{NL})}_{2\omega}]_z\exp(-ik_{2\omega_z}x)\\
&= i\frac{\mu_0(2\omega)^2}{2(2\omega n_{2\omega_z}/c)}\underbrace{\frac{\varepsilon_0}{2}\chi^{(2)}_{zxx}A^{y\,2}_{\omega}\exp(2ik_{\omega_y}x)}_{=[\mathbf{P}^{(\mathrm{NL})}_{2\omega}]_z}\exp(-ik_{2\omega_z}x)\\
&= i\frac{\omega\chi^{(2)}_{zxx}}{2n_{2\omega_z}c}A^{y\,2}_{\omega}\exp[i(2k_{\omega_y} - k_{2\omega_z})x],
\end{aligned}
$$

---

[3] Keep in mind that a negative frequency argument to the right of the semicolon in the susceptibility is to be associated with the complex conjugate of the respective electric field; see Butcher and Cotter, section 2.3.2.

while for the fundamental wave,

$$\begin{aligned}\frac{\partial A_{\omega}^{y}}{\partial x} &= i\frac{\mu_0\omega^2}{2k_{\omega_y}}[\mathbf{P}_{\omega}^{(\mathrm{NL})}]_y \exp(-ik_{\omega_y}x)\\
&= i\frac{\mu_0\omega^2}{2(\omega n_{\omega_y}/c)}\underbrace{\varepsilon_0\chi_{zxx}^{(2)}A_{2\omega}^{z}\exp(ik_{2\omega_z}x)A_{\omega}^{y*}\exp(-ik_{\omega_y}x)}_{=[\mathbf{P}_{\omega}^{(\mathrm{NL})}]_y}\exp(-ik_{\omega_y}x)\\
&= i\frac{\omega\chi_{zxx}^{(2)}}{2n_{\omega_y}c}A_{2\omega}^{z}A_{\omega}^{y*}\exp[-i(2k_{\omega_y}-k_{2\omega_z})x].\end{aligned}$$

These equations can hence be summarized by the coupled system

$$\frac{\partial A_{2\omega}^{z}}{\partial x} = i\frac{\omega\chi_{zxx}^{(2)}}{2n_{2\omega_z}c}A_{\omega}^{y\,2}\exp(i\Delta kx), \tag{3a}$$

$$\frac{\partial A_{\omega}^{y}}{\partial x} = i\frac{\omega\chi_{zxx}^{(2)}}{2n_{\omega_y}c}A_{2\omega}^{z}A_{\omega}^{y*}\exp(-i\Delta kx). \tag{3b}$$

where

$$\begin{aligned}\Delta k &= 2k_{\omega_y}-k_{2\omega_z}\\
&= 2\omega n_{\omega_y}/c - 2\omega n_{2\omega_z}/c\\
&= (2\omega/c)(n_{\omega_y}-\omega n_{2\omega_z})\end{aligned}$$

is the so-called phase mismatch between the pump and second harmonic wave.

*Boundary conditions*

Here the boundary conditions are simply that no second harmonic signal is present at the input,

$$A_{2\omega}^{z}(0) = 0,$$

together with a known input field at the fundamental frequency,

$$A_{\omega}^{y}(0) = \{\text{known}\}.$$

*Solving the wave equation*

Considering a nonzero $\Delta k$, the conversion efficiency is regularly quite small, and one may approximately take the spatial distribution of the pump wave to be constant, $A_{\omega}^{y}(x) \approx A_{\omega}^{y}(0)$. Using this approximation[4], and by applying the initial condition $A_{2\omega}^{z}(0) = 0$ of the second harmonic signal, one finds

$$\begin{aligned}A_{2\omega}^{z}(L) &= \int_0^L \frac{\partial A_{2\omega}^{z}(z)}{\partial x}\,dx\\
&= \int_0^L i\frac{\omega\chi_{zxx}^{(2)}}{2n_{2\omega_z}c}A_{\omega}^{y\,2}(0)\exp(i\Delta kx)\,dx\\
&= \frac{\omega\chi_{zxx}^{(2)}}{2n_{2\omega_z}c}A_{\omega}^{y\,2}(0)\frac{1}{\Delta k}[\exp(i\Delta kL)-1]\\
&= \big\{\text{Use }[\exp(i\Delta kL)-1]/\Delta k = iL\exp(i\Delta kL/2)\,\mathrm{sinc}(\Delta kL/2)\big\}\\
&= i\frac{\omega\chi_{zxx}^{(2)}L}{2n_{2\omega_z}c}A_{\omega}^{y\,2}(0)\exp(i\Delta kL/2)\,\mathrm{sinc}(\Delta kL/2).\end{aligned}$$

---

[4] For an outline of the method of solving the coupled system (1) exactly in terms of Jacobian elliptic functions (thus allowing for a depleted pump as well), see J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, Phys. Rev. **127**, 1918–1939, (1962).

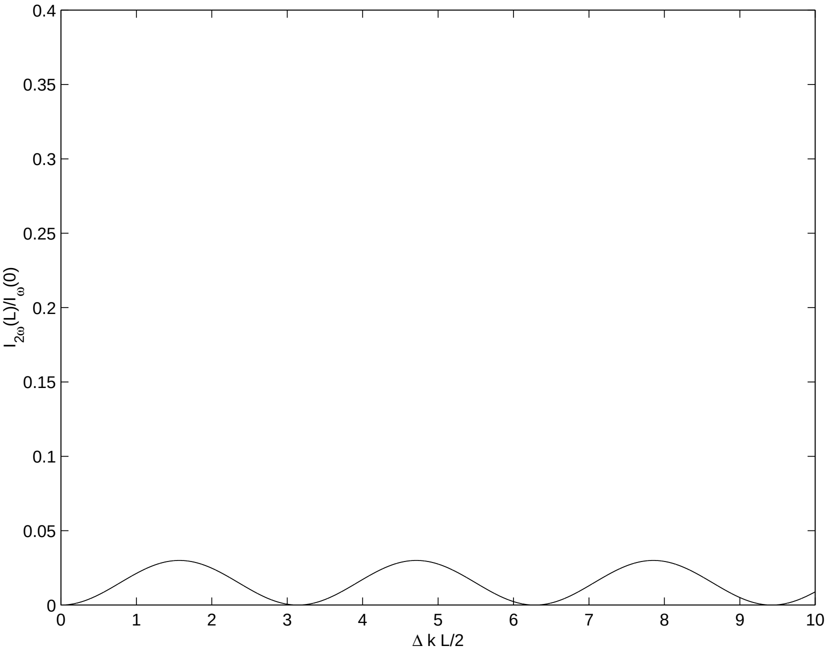

Figure 2. Conversion efficiency $I_{2\omega}(L)/I_\omega(0)$ as function of normalized crystal length $\Delta kL/2$. The conversion efficiency is in the phase mismatched case ($\Delta k \neq 0$) a periodic function, with period $2L_{\rm c}$, with $L_{\rm c} = \pi/\Delta k$ being the *coherence length*.

In terms if the light intensities of the waves, one after a propagation distance $x = L$ hence has the second harmonic signal with intensity $I_{2\omega}(L)$ expressed in terms of the input intensity $I_\omega$ as

$$\begin{aligned}
I_{2\omega}(L) &= \frac{1}{2}\varepsilon_0 c n_{2\omega_z}|A^z_{2\omega}(L)|^2\\
&= \frac{1}{2}\varepsilon_0 c n_{2\omega_z}\Big|i\frac{\omega\chi^{(2)}_{zxx}L}{2n_{2\omega_z}c}A^{y\,2}_\omega(0)\exp(i\Delta kL/2)\,\mathrm{sinc}(\Delta kL/2)\Big|^2\\
&= \varepsilon_0\frac{\omega^2L^2}{8n_{2\omega_z}c}|\chi^{(2)}_{zxx}|^2|A^y_\omega(0)|^4\,\mathrm{sinc}^2(\Delta kL/2)\\
&= \left\{\text{Use } |A^y_\omega(0)|^2 = \frac{2I_\omega(0)}{\varepsilon_0 c n_{\omega_y}}\right\}\\
&= \varepsilon_0\frac{\omega^2L^2}{8n_{2\omega_z}c}|\chi^{(2)}_{zxx}|^2\frac{4I^2_\omega(0)}{\varepsilon_0^2c^2n^2_{\omega_y}}\,\mathrm{sinc}^2(\Delta kL/2)\\
&= \frac{\omega^2L^2}{2\varepsilon_0c^3}\frac{|\chi^{(2)}_{zxx}(-2\omega;\omega,\omega)|^2}{n_{2\omega_z}n^2_{\omega_y}}I^2_\omega(0)\,\mathrm{sinc}^2(\Delta kL/2),
\end{aligned}$$

i. e. with the conversion efficiency

$$\frac{I_{2\omega}(L)}{I_\omega(0)} = \frac{\omega^2L^2}{2\varepsilon_0c^3}\frac{|\chi^{(2)}_{zxx}(-2\omega;\omega,\omega)|^2}{n_{2\omega_z}n^2_{\omega_y}}I_\omega(0)\,\mathrm{sinc}^2(\Delta kL/2).$$

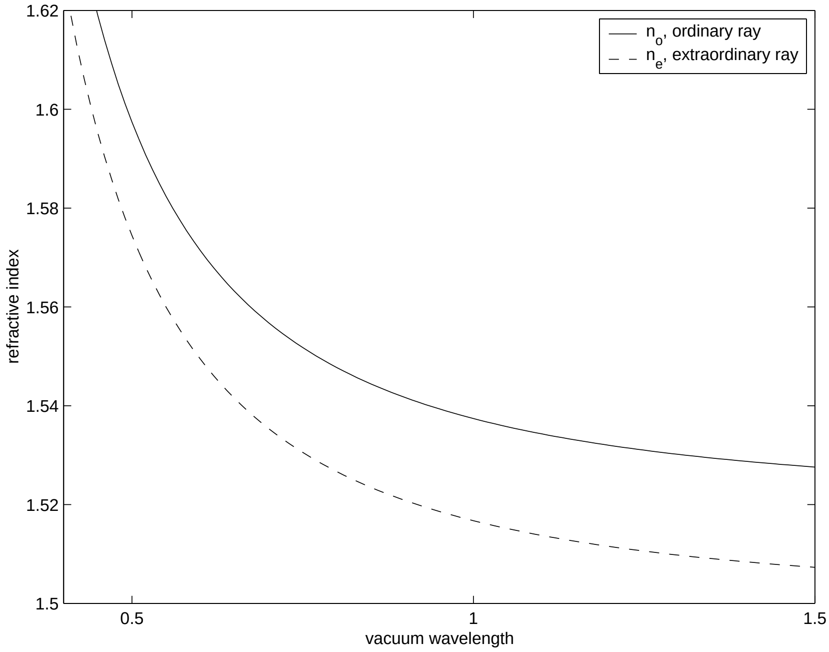

Figure 3. Ordinary and extraordinary refractive indices of a negative uniaxial crystal as function of vacuum wavelength of the light, in the case of normal dispersion. Phase matching between the pump and second harmonic wave is obtained whenever $n_{\omega_y} \equiv n_{\mathrm{O}}(\omega) = n_{\mathrm{E}}(2\omega) \equiv n_{2\omega_z}$.

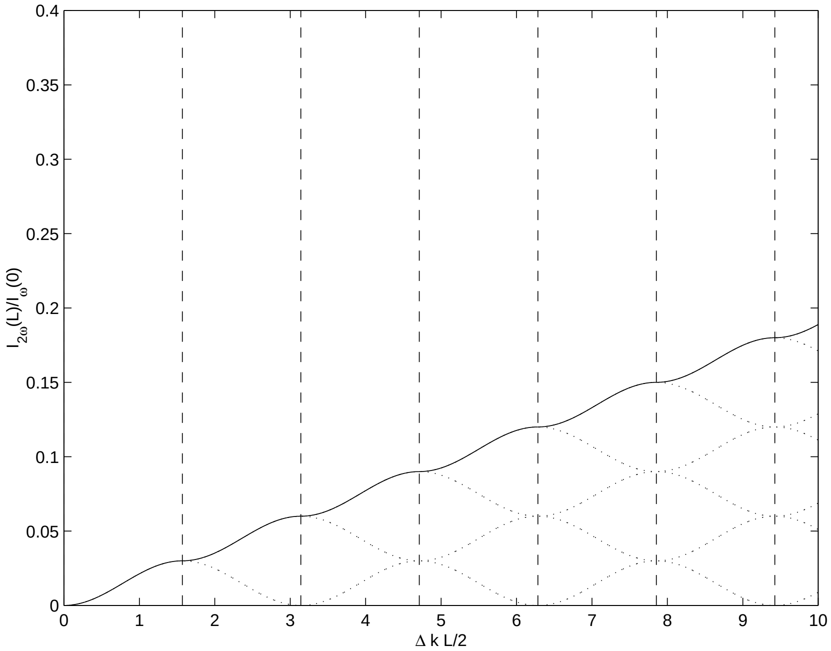

Figure 4. Conversion efficiency $I_{2\omega}(L)/I_{\omega}(0)$ as function of normalized crystal length $\Delta kL/2$ when the material properties are periodically reversed, with a half-period of $L_{\mathrm{c}}$.

**Optical Kerr-effect - Field corrected refractive index**

As a start, we assume a monochromatic optical wave (containing forward and/or backward propagating components) polarized in the $xy$-plane,

$$\mathbf{E}(z,t) = \mathrm{Re}[\mathbf{E}_\omega(z)\exp(-i\omega t)] \in \mathbb{R}^3,$$

with all spatial variation of the field contained in

$$\mathbf{E}_\omega(z) = \mathbf{e}_x E^x_\omega(z) + \mathbf{e}_y E^y_\omega(z) \in \mathbb{C}^3.$$

*The optical interaction*

Optical Kerr-effect is in isotropic media described by the third order susceptibility[5]

$$\chi^{(3)}_{\mu\alpha\beta\gamma}(-\omega;\omega,\omega,-\omega).$$

*Symmetries of the medium*

The general set of nonzero components of $\chi^{(3)}_{\mu\alpha\beta\gamma}$ for isotropic media are from Appendix A3.3 of Butcher and Cotters book given as

$$\begin{aligned}
&\chi^{(3)}_{xxxx} = \chi^{(3)}_{yyyy} = \chi^{(3)}_{zzzz},\\
&\chi^{(3)}_{yyzz} = \chi^{(3)}_{zzyy} = \chi^{(3)}_{zzxx} = \chi^{(3)}_{xxzz} = \chi^{(3)}_{xxyy} = \chi^{(3)}_{yyxx}\\
&\chi^{(3)}_{yzyz} = \chi^{(3)}_{zyzy} = \chi^{(3)}_{zxzx} = \chi^{(3)}_{xzxz} = \chi^{(3)}_{xyxy} = \chi^{(3)}_{yxyx}\\
&\chi^{(3)}_{yzzy} = \chi^{(3)}_{zyyz} = \chi^{(3)}_{zxxz} = \chi^{(3)}_{xzzx} = \chi^{(3)}_{xyyx} = \chi^{(3)}_{yxxy}
\end{aligned} \tag{4}$$

with

$$\chi^{(3)}_{xxxx} = \chi^{(3)}_{xxyy} + \chi^{(3)}_{xyxy} + \chi^{(3)}_{xyyx},$$

i. e. a general set of 21 elements, of which only 3 are independent.

*Additional symmetries*

By applying the intrinsic permutation symmetry in the middle indices for optical Kerr-effect, one generally has

$$\chi^{(3)}_{\mu\alpha\beta\gamma}(-\omega;\omega,\omega,-\omega) = \chi^{(3)}_{\mu\beta\alpha\gamma}(-\omega;\omega,\omega,-\omega),$$

which hence slightly reduce the set (4) to still 21 nonzero elements, but of which now only two are independent. For a beam polarized in the $xy$-plane, the elements of interest are only those which only contain $x$ or $y$ in the indices, i. e. the subset

$$\chi^{(3)}_{xxxx} = \chi^{(3)}_{yyyy}, \quad \chi^{(3)}_{xxyy} = \chi^{(3)}_{yyxx} = \left\{ \begin{array}{c} \text{intr. perm. symm.} \\ (\alpha,\omega) \rightleftharpoons (\beta,\omega) \end{array} \right\} = \chi^{(3)}_{xyxy} = \chi^{(3)}_{yxyx}, \quad \chi^{(3)}_{xyyx} = \chi^{(3)}_{yxxy},$$

with

$$\chi^{(3)}_{xxxx} = \chi^{(3)}_{xxyy} + \chi^{(3)}_{xyxy} + \chi^{(3)}_{xyyx},$$

i. e. a set of eight elements, of which only two are independent.

---

[5] Again, keep in mind that in the convention of Butcher and Cotter, the frequency arguments to the right of the semicolon may be writen in arbitrary order, hence we may in an equal description instead use

$$\chi^{(3)}_{\mu\alpha\beta\gamma}(-\omega;\omega,-\omega,\omega)$$

or

$$\chi^{(3)}_{\mu\alpha\beta\gamma}(-\omega;-\omega,\omega,\omega)$$

for this description of the third order interaction between light and matter.

*The polarization density*

The degeneracy factor $K(-\omega;\omega,\omega,-\omega)$ is calculated as

$$K(-\omega;\omega,\omega,-\omega) = 2^{l+m-n}p = 2^{1+0-3}3 = 3/4.$$

From the reduced set of nonzero susceptibilities for the beam polarized in the $xy$-plane, and by using the calculated value of the degeneracy factor in the convention of Butcher and Cotter, we hence have the third order electric polarization density at $\omega_\sigma = \omega$ given as $\mathbf{P}^{(n)}(\mathbf{r},t) = \mathrm{Re}[\mathbf{P}^{(n)}_\omega \exp(-i\omega t)]$, with

$$\begin{aligned}
\mathbf{P}^{(3)}_\omega &= \sum_\mu \mathbf{e}_\mu (P^{(3)}_\omega)_\mu \\
&= \{\text{Using the convention of Butcher and Cotter}\} \\
&= \sum_\mu \mathbf{e}_\mu \left[\varepsilon_0 \frac{3}{4} \sum_\alpha \sum_\beta \sum_\gamma \chi^{(3)}_{\mu\alpha\beta\gamma}(-\omega;\omega,\omega,-\omega)(E_\omega)_\alpha (E_\omega)_\beta (E_{-\omega})_\gamma\right] \\
&= \{\text{Evaluate the sums over } (x,y,z) \text{ for field polarized in the } xy \text{ plane}\} \\
&= \varepsilon_0 \frac{3}{4}\{\mathbf{e}_x[\chi^{(3)}_{xxxx}E^x_\omega E^x_\omega E^x_{-\omega} + \chi^{(3)}_{xyyx}E^y_\omega E^y_\omega E^x_{-\omega} + \chi^{(3)}_{xyxy}E^y_\omega E^x_\omega E^y_{-\omega} + \chi^{(3)}_{xxyy}E^x_\omega E^y_\omega E^y_{-\omega}] \\
&\qquad + \mathbf{e}_y[\chi^{(3)}_{yyyy}E^y_\omega E^y_\omega E^y_{-\omega} + \chi^{(3)}_{yxxy}E^x_\omega E^x_\omega E^y_{-\omega} + \chi^{(3)}_{yxyx}E^x_\omega E^y_\omega E^x_{-\omega} + \chi^{(3)}_{yyxx}E^y_\omega E^x_\omega E^x_{-\omega}]\} \\
&= \{\text{Make use of } \mathbf{E}_{-\omega} = \mathbf{E}^*_\omega \text{ and relations } \chi^{(3)}_{xxyy} = \chi^{(3)}_{yyxx}, \text{ etc.}\} \\
&= \varepsilon_0 \frac{3}{4}\{\mathbf{e}_x[\chi^{(3)}_{xxxx}E^x_\omega |E^x_\omega|^2 + \chi^{(3)}_{xyyx}E^{y\,2}_\omega E^{x*}_\omega + \chi^{(3)}_{xyxy}|E^y_\omega|^2 E^x_\omega + \chi^{(3)}_{xxyy}E^x_\omega |E^y_\omega|^2] \\
&\qquad + \mathbf{e}_y[\chi^{(3)}_{xxxx}E^y_\omega |E^y_\omega|^2 + \chi^{(3)}_{xyyx}E^{x\,2}_\omega E^{y*}_\omega + \chi^{(3)}_{xyxy}|E^x_\omega|^2 E^y_\omega + \chi^{(3)}_{xxyy}E^y_\omega |E^x_\omega|^2]\} \\
&= \{\text{Make use of the intrinsic permutation symmetry}\} \\
&= \varepsilon_0 \frac{3}{4}\{\mathbf{e}_x[(\chi^{(3)}_{xxxx}|E^x_\omega|^2 + 2\chi^{(3)}_{xxyy}|E^y_\omega|^2)E^x_\omega + (\chi^{(3)}_{xxxx} - 2\chi^{(3)}_{xxyy})E^{y\,2}_\omega E^{x*}_\omega \\
&\qquad \mathbf{e}_y[(\chi^{(3)}_{xxxx}|E^y_\omega|^2 + 2\chi^{(3)}_{xxyy}|E^x_\omega|^2)E^y_\omega + (\chi^{(3)}_{xxxx} - 2\chi^{(3)}_{xxyy})E^{x\,2}_\omega E^{y*}_\omega.
\end{aligned}$$

For the optical field being linearly polarized, say in the $x$-direction, the expression for the polarization density is significantly simplified, to yield

$$\mathbf{P}^{(3)}_\omega = \varepsilon_0(3/4)\mathbf{e}_x \chi^{(3)}_{xxxx}|E^x_\omega|^2 E^x_\omega,$$

i. e. taking a form that can be interpreted as an intensity-dependent ($\sim |E^x_\omega|^2$) contribution to the refractive index (cf. Butcher and Cotter §6.3.1).

*The wave equation – Time independent case*

In this example, we consider continuous wave propagation[6] in optical Kerr-media, using light polarized in the $x$-direction and propagating along the positive direction of the $z$-axis,

$$\mathbf{E}(\mathbf{r},t) = \mathrm{Re}[\mathbf{E}_\omega(z)\exp(-i\omega t)], \qquad \mathbf{E}_\omega(z) = \mathbf{A}_\omega(z)\exp(ikz) = \mathbf{e}_x A^x_\omega(z)\exp(ikz),$$

where, as previously, $k = \omega n_0/c$. From material handed out during the third lecture (notes on the Butcher and Cotter convention), the nonlinear polarization density for $x$-polarized light is given as $\mathbf{P}^{(\mathrm{NL})}_\omega = \mathbf{P}^{(3)}_\omega$, with

$$\begin{aligned}
\mathbf{P}^{(3)}_\omega &= \varepsilon_0(3/4)\mathbf{e}_x \chi^{(3)}_{xxxx}(-\omega;\omega,\omega,-\omega)|E^x_\omega|^2 E^x_\omega \\
&= \varepsilon_0(3/4)\chi^{(3)}_{xxxx}|\mathbf{E}_\omega|^2 \mathbf{E}_\omega \\
&= \varepsilon_0(3/4)\chi^{(3)}_{xxxx}|\mathbf{A}_\omega|^2 \mathbf{A}_\omega \exp(ikz),
\end{aligned}$$

---

[6] That is to say, a time independent problem with the temporal envelope of the electrical field being constant in time.

and the time independent wave equation for the field envelope $\mathbf{A}_\omega$, using Eq. (6), becomes

$$\begin{aligned}\frac{\partial}{\partial z}\mathbf{A}_\omega &= i\frac{\mu_0\omega^2}{2k}\underbrace{\varepsilon_0(3/4)\chi^{(3)}_{xxxx}|\mathbf{A}_\omega|^2\mathbf{A}_\omega\exp(ikz)}_{=\mathbf{P}^{(\mathrm{NL})}_\omega(z)}\exp(-ikz)\\ &= i\frac{3\omega^2}{8c^2k}\chi^{(3)}_{xxxx}|\mathbf{A}_\omega|^2\mathbf{A}_\omega\\ &= \{\text{since } k=\omega n_0(\omega)/c\}\\ &= i\frac{3\omega}{8cn_0}\chi^{(3)}_{xxxx}|\mathbf{A}_\omega|^2\mathbf{A}_\omega,\end{aligned}$$

or, equivalently, in its scalar form

$$\frac{\partial}{\partial z}A^x_\omega = i\frac{3\omega}{8cn_0}\chi^{(3)}_{xxxx}|A^x_\omega|^2A^x_\omega.$$

*Boundary conditions – Time independent case*

For this special case of unidirectional wave propagation, the boundary condition is simply a known optical field at the input,

$$A^x_\omega(0) = \{\text{known}\}.$$

*Solving the wave equation – Time independent case*

If the medium of interest now is analyzed at an angular frequency far from any resonance, we may look for solutions to this equation with $|\mathbf{A}_\omega(z)|$ being constant (for a lossless medium). For such a case it is straightforward to integrate the final wave equation to yield the general solution

$$\mathbf{A}_\omega(z) = \mathbf{A}_\omega(0)\exp[i\frac{3\omega}{8cn_0}\chi^{(3)}_{xxxx}|\mathbf{A}_\omega(0)|^2z],$$

or, again equivalently, in the scalar form

$$A^x_\omega(z) = A^x_\omega(0)\exp[i\frac{3\omega}{8cn_0}\chi^{(3)}_{xxxx}|A^x_\omega(0)|^2z],$$

which hence gives the solution for the real-valued electric field $\mathbf{E}(\mathbf{r},t)$ as

$$\begin{aligned}\mathbf{E}(\mathbf{r},t) &= \mathrm{Re}[\mathbf{E}_\omega(z)\exp(-i\omega t)]\\ &= \mathrm{Re}\{\mathbf{A}_\omega(z)\exp[i(kz-\omega t)]\}\\ &= \mathrm{Re}\{\mathbf{A}_\omega(0)\exp[i(\underbrace{\frac{\omega n_0}{c}z+\frac{3\omega}{8cn_0}\chi^{(3)}_{xxxx}|A^x_\omega(0)|^2z}_{\equiv k_{\mathrm{eff}}z}-\omega t)]\}.\end{aligned}$$

From this solution, one immediately finds that the wave propagates with an effective propagation constant

$$k_{\mathrm{eff}} = \frac{\omega}{c}[n_0+\frac{3}{8n_0}\chi^{(3)}_{xxxx}|A^x_\omega(0)|^2],$$

that is to say, experiencing the intensity dependent refractive index

$$\begin{aligned}n_{\mathrm{eff}} &= n_0+\frac{3}{8n_0}\chi^{(3)}_{xxxx}|A^x_\omega(0)|^2\\ &= n_0+n_2|A^x_\omega(0)|^2,\end{aligned}$$

with

$$n_2 = \frac{3}{8n_0}\chi^{(3)}_{xxxx}.$$

# Lecture X

**Lecture Notes on Nonlinear Optics**
Nonlinear Optics (5A5513, 5p for advanced undergraduate and doctoral students)
Course given at the Royal Institute of Technology,
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## LECTURE 10

In this lecture, we will focus on examples of electromagnetic wave propagation in nonlinear optical media, by applying the forms of Maxwell's equations as obtained in the eighth lecture to a set of particular nonlinear interactions as described by the previously formulated nonlinear susceptibility formalism.

The outline for this lecture is:
- What are solitons?
- Basics of soliton theory
- Spatial and temporal solitons
- The mathematical equivalence between spatial and temporal solitons
- The creation of temporal and spatial solitons

### What are solitons?

The first reported observation of solitons was made in 1834 by John Scott Russell, a Scottish scientist and later famous Victorian engineer and shipbuilder, while studying water waves in the Glasgow-Edinburgh channel. As part of this investigation, he was observing a boat being pulled along, rapidly, by a pair of horses. For some reason, the horses stopped the boat rather suddenly, and the stopping of the boat caused a verystrong wave to be generated. This wave, in fact, a significant hump of water stretching across the rather narrow canal, rose up at the front of the boat and proceeded to travel, quite rapidly down the canal. Russell, immediately, realised that the wave was something very special. It was "alone", in the sense that it sat on the canal with no disturbance to the front or the rear, nor did it die away until he had followed it for quite a long way. The word "alone" is synonymous with "solitary", and Russell soon referred to his observation as the Great Solitary Wave.

The word "solitary" is now routinely used, indeed even the word "solitary" tends to be replaced by the more generic word "soliton". Once the physics behind Russell's wave is understood, however, solitons, of one kind or another, appear to be everywhere but it is interesting that the underlying causes of soliton generation were not understood by Russell, and only partially by his contemporaries.

### Classes of solitons

*Bright temporal envelope solitons*

Pulses of light with a certain shape and energy that can propagate unchanged over large distances. This is the class of solitons which we will focus on in this lecture.

*Dark temporal envelope solitons*

Pulses of "darkness" within a continuous wave, where the pulses are of a certain shape, and possess propagation properties similar to the bright solitons.

*Spatial solitons*

Continuous wave beams or pulses, with a transverse extent of the beam that via the refractive index change due to optical Kerr-effect can compensate for the diffraction of the beam. The optically induced change of refractive index works as an effective waveguide for the light.

**The normalized nonlinear Schrödinger equation for temporal solitons**

The starting point for the analysis of temporal solitons is the time-dependent wave equation for the spatial envelopes of the electromagnetic fields in optical Kerr-media, here for simplicity taken for linearly polarized light in isotropic media,

$$\Big(i\frac{\partial}{\partial z}+i\frac{1}{v_{\mathrm{g}}}\frac{\partial}{\partial t}-\frac{\beta}{2}\frac{\partial^2}{\partial t^2}\Big)\mathbf{A}_\omega(z,t)=-\frac{\omega n_2}{c}|\mathbf{A}_\omega(z,t)|^2\mathbf{A}_\omega(z,t), \tag{1}$$

where, as previously, $v_{\mathrm{g}}=(dk/d\omega)^{-1}$ is the linear group velocity, and where we introduced the notation

$$\beta=\frac{d^2k}{d\omega^2}\Big|_{\omega_\sigma}$$

for the second order linear dispersion of the medium, and (in analogy with Butcher and Cotter Eq. (6.63)),

$$n_2=(3/8n_0)\chi^{(3)}_{xxxx}$$

for the intensity-dependent refractive index $n=n_0+n_2|\mathbf{E}_\omega|^2$. Since we here are considering wave propagation in isotropic media, with linearly polarized light (for which no polarization state cross-talk occur), the wave equation (1) is conveniently taken in a scalar form as

$$\Big(i\frac{\partial}{\partial z}+i\frac{1}{v_{\mathrm{g}}}\frac{\partial}{\partial t}-\frac{\beta}{2}\frac{\partial^2}{\partial t^2}\Big)A_\omega(z,t)=-\frac{\omega n_2}{c}|A_\omega(z,t)|^2A_\omega(z,t). \tag{2}$$

Equation (2) consists of three terms that interact. The first two terms contain first order derivatives of the envelope, and these terms can be seen as the homogeneous part of a wave equation for the envelope, giving travelling wave solutions that depend on the other two terms, which rather act like source terms.

The third term contains a second order derivative of the envelope, and this terms is also linearly dependent on the dispersion $\beta$ of the medium, that is to say, the change of the group velocity of the medium with respect to the angular frequency $\omega$ of the light. This term is generally responsible for smearing out a short pulse as it traverses a dispersive medium.

Finally, the fourth term is a nonlinear source term, which depending on the sign of $n_2$ will concentrate higher frequency components either at the leading or trailing edge of the pulse, as soon will be shown.

*The effect of dispersion*

The *group velocity dispersion* $dv_{\mathrm{g}}/d\omega$ is related to the introduced dispersion parameter $\beta\equiv d^2k/d\omega^2$ as

$$\frac{dv_{\mathrm{g}}}{d\omega}=\frac{d}{d\omega}\left[\left(\frac{dk(\omega)}{d\omega}\right)^{-1}\right]=-\underbrace{\left(\frac{dk(\omega)}{d\omega}\right)^{-2}}_{\equiv v_{\mathrm{g}}^2}\underbrace{\frac{d^2k(\omega)}{d\omega^2}}_{\equiv\beta}=-v_{\mathrm{g}}^2\beta,$$

and hence the sign of the group velocity dispersion is the opposite of the sign of the dispersion parameter $\beta$. In order to get a qualitative picture of the effect of linear dispersion, let us consider the effect of the sign of $\beta$:

- $\beta>0$:
  For this case, the group velocity dispersion is negative, since

  $$\frac{dv_{\mathrm{g}}}{d\omega}=-v_{\mathrm{g}}^2\beta<0.$$

  This implies that the *group velocity decreases* with an increasing angular frequency $\omega$. In other words, the "blue" frequency components of the pulse travel slower than the "red" components. Considering the effects on the pulse as it propagates, the leading edge of the pulse will after some distance contain a higher concentration of low ("red") frequencies, while the trailing edge rather will contain a higher concentration of high ("blue") frequencies. This effect is illustrated in Fig. 1.

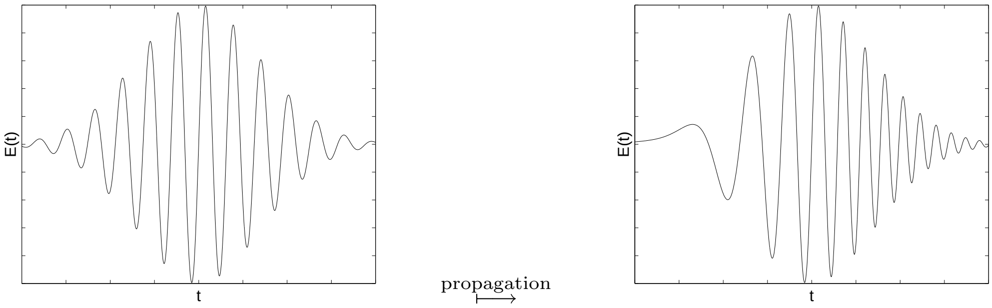

Figure 1. Pulse propagation in a linearly dispersive medium with $\beta > 0$.

Whenever "red" frequency components travel faster than "blue" components, we usually associate this with so-called *normal dispersion*.

- $\beta < 0$:
  For this case, the group velocity dispersion is instead positive, since now

$$\frac{dv_{\mathrm{g}}}{d\omega} = -v_{\mathrm{g}}^2 \beta > 0.$$

This implies that the *group velocity increases* with an increasing angular frequency $\omega$. In other words, the "blue" frequency components of the pulse now travel *faster* than the "red" components. Considering the effects on the pulse as it propagates, the leading edge of the pulse will after some distance hence contain a higher concentration of high ("blue") frequencies, while the trailing edge rather will contain a higher concentration of low ("red") frequencies. This effect, being the inverse of the one described for a negative group velocity dispersion, is illustrated in Fig. 2.

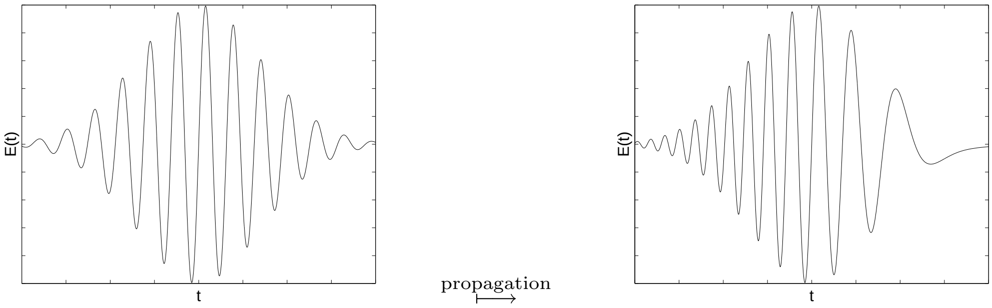

Figure 2. Pulse propagation in a linearly dispersive medium with $\beta < 0$.

Whenever "blue" frequency components travel faster than "red" components, we usually associate this with so-called *anomalous dispersion*.

Notice that depending on the distribution of the frequency components of the pulse as it enters a dispersive medium, the pulse may for some propagation distance actually undergo *pulse compression*. For $\beta > 0$, this occurs if the leading edge of the pulse contain a higher concentration of "blue" frequencies, while for $\beta < 0$, this occurs if the leading edge of the pulse instead contain a higher concentration of "red" frequencies.

*The effect of a nonlinear refractive index*

Having sorted out the effects of the sign of $\beta$ on the pulse propagation, we will now focus on the effects of a nonlinear, optical field dependent refractive index of the medium.

In order to extract the effect of the nonlinear refractive index, we will here go to the very definition of the instantaneous angular frequency of the light from its real-valued electric field,

$$\mathbf{E}(\mathrm{r}, t) = \mathrm{Re}[\mathrm{E}_{\omega}(\mathbf{r}, t)\exp(-i\omega t)].$$

For light propagating in a medium where the refractive index depend on the intensity as

$$n(t) = n_0 + n_2 I(t),$$

the spatial envelope will typically be described by an effective propagation constant (see lecture notes as handed out during lecture nine)

$$k_{\mathrm{eff}}(\omega, I(t)) = (\omega/c)(n_0 + n_2 I(t)),$$

and the local, instantaneous angular frequency becomes

$$\begin{aligned}\omega_{\mathrm{loc}} &= -\frac{d}{dt}\Big\{\text{phase of the light}\Big\}\\ &= -\frac{d}{dt}\left[k_{\mathrm{eff}}(\omega, I(t)) - \omega t\right]\\ &= -\frac{d}{dt}\left[\frac{\omega}{c}(n_0(\omega) + n_2(\omega)I(t))\right] + \omega\\ &= \omega - \frac{\omega n_2(\omega)}{c}\frac{dI(t)}{dt}.\end{aligned}$$

The typical behaviour of the instantaneous angular frequancy $\omega_{\mathrm{loc}}(t)$ on a typical pulse shape is shown in Fig. 3, for the case of $n_2 > 0$ and a Gaussian pulse.

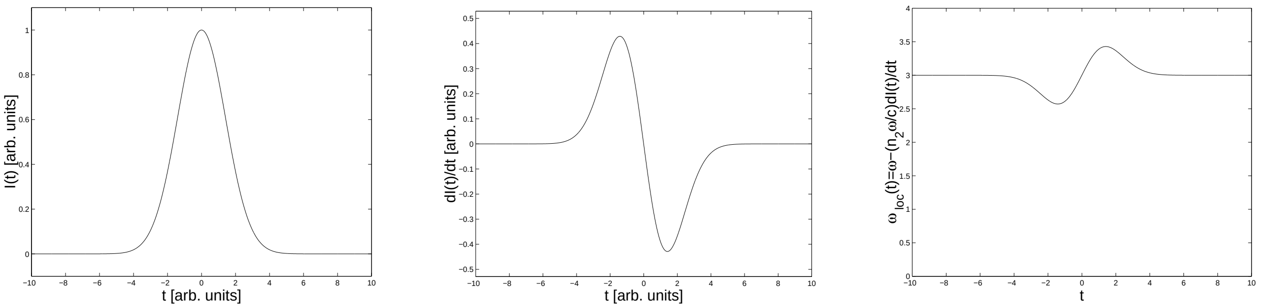

Figure 3. Effect of a intensity dependent refractive index $n = n_0 + n_2 I(t)$ on frequency content of the pulse.

As seen in the figure, the leading edge of the pulse has a slight decrease in angular frequency, while the trailing edge has a slight increase. This means that in the presence of an intensity dependent refractive index, for $n_2 > 0$, the pulse will have a concentration of "red" frequencies at the leading edge, while the trailing edge will have a concentration of "blue" frequencies. This is illustrated in Fig. 4.

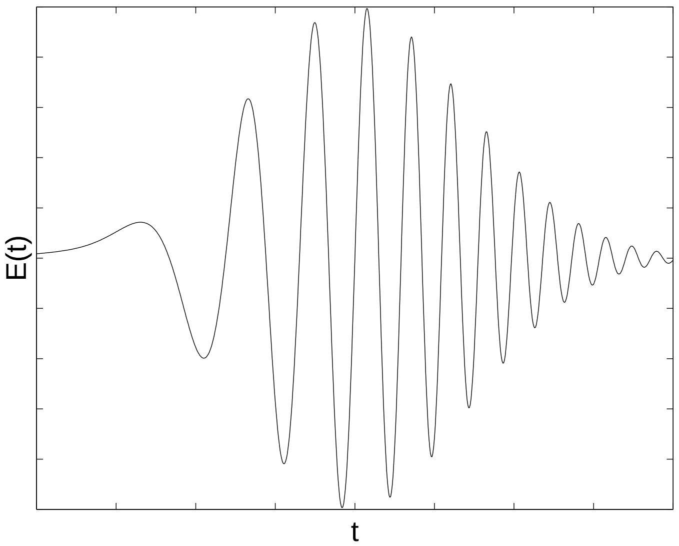

Figure 4. Typical frequency chirp of an optical pulse in a nonlinear medium with $n_2 > 0$.

If instead $n_2 < 0$, i. e. for an intensity dependent refractive index that decrease with an increasing intensity, the roles of the "red" and "blue" edges of the pulse are reversed.

*The basic idea behind temporal solitons*

As seen from Figs. 2 and 4, the effect of anomalous dispersion (with $\beta < 0$) and the effect of a nonlinear, intensity dependent refractive index (with $n_2 > 0$) are opposite of each other. When combined, that is to say, considering pulse propagation in a medium which simultaneously possesses anomalous dispersion and $n_2 > 0$, these effects can combine, *giving a pulse that can propagate without altering its shape.* This is the basic principle of the *temporal soliton.*

*Normalization of the nonlinear Schrödinger equation*

Equation (2) can now be cast into a normalized form, the so-called *nonlinear Schrödinger equation*, by applying the change of variables[1]

$$u = \tau\sqrt{\frac{n_2\omega}{c|\beta|}}A_\omega, \qquad s = (t - z/v_{\mathrm{g}})/\tau, \qquad \zeta = |\beta|z/\tau^2,$$

where $\tau$ is some characteristic time of the evolution of the pulse, usually taken as the pulse duration time, which gives the normalized form

$$\Big(i\frac{\partial}{\partial\zeta} - \frac{1}{2}\,\mathrm{sgn}(\beta)\frac{\partial^2}{\partial s^2}\Big)u(\zeta, s) + |u(\zeta, s)|^2 u(\zeta, s) = 0. \tag{3}$$

This normalized equation has many interesting properties, and for some cases even analytical solutions exist, as we will see in the following sections. Before actually solving the equation, however, we will consider another mechanism for the generation of solitons.

---

[1] Please note that there is a printing error in Butcher and Cotter's book in the section that deals with the normalization of the nonlinear Schrödinger equation. In the first line of Eq. (7.55), there is an ambiguity of the denominator, as well as an erroneous dispersion term, and the equation

$$u = \tau\sqrt{n_2\omega/c|d^2k/d\omega^2|^2}\widehat{E}$$

should be replaced by

$$u = \tau\sqrt{n_2\omega/(c|d^2k/d\omega^2|)}\widehat{E}.$$

(The other lines of Eq. (7.55) in Butcher and Cotter are correct.)

Before leaving the temporal pulse propagation, a few remarks on the signs of the dispersion term $\beta$ and the nonlinear refractive index $n_2$ should be made. Whenever $\beta > 0$, the group velocity dispersion

$$\frac{dv_{\rm g}}{d\omega} \equiv \frac{d}{d\omega}\left[\left(\frac{dk}{d\omega}\right)^{-1}\right] = -\left(\frac{dk}{d\omega}\right)^{-2}\frac{d^2k}{d\omega^2} = -v_{\rm g}^2\frac{d^2k}{d\omega^2}$$

will be negative, and the pulse will experience what we call a *normal* dispersion, for which the refractive index of the medium decrease with an increasing wavelength of the light. This is the "common" way dispersion enters in optical processes, where the pulse is broadened as it traverses the medium.

### Spatial solitons

As a light beam with some limited spatial extent in the transverse direction enter an optical Kerr media, the intensity variation across the beam will via the intensity dependent refractive index $n = n_0 + n_2 I$ form a lensing through the medium. Depending on the sign of the coefficient $n_2$ (the "nonlinear refractive index"), the beam will either experience a defocusing lensing effect (if $n_2 < 0$) or a focusing lensing effect (if $n_2 > 0$); in the latter case the beam itself will create a self-induced waveguide in the medium (see Fig. 5).

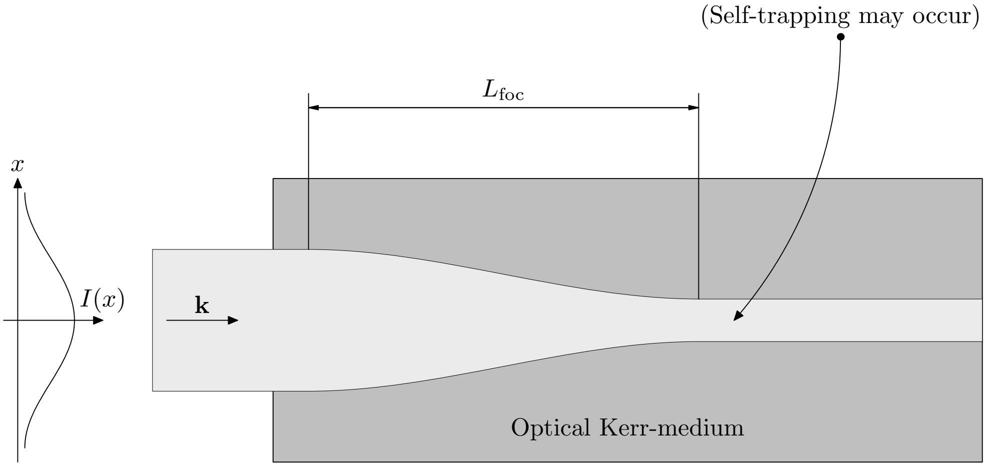

Figure 5. An illustration of the effect of self-focusing.

As being the most important case for beams with maximum intensity in the middle of the beam (as we usually encounter them in most situations), we will focus on the case $n_2 > 0$. For this case, highly intense beams may cause such a strong focusing that the beam eventually break up again, due to strong diffraction effects for very narrow beams, or even due to material damage in the nonlinear crystal.

For some situations, however, there exist stationary solutions to the spatial light distribution that exactly balance between the self-focusing and the diffraction of the beam. We can picture this as a balance between two lensing effects, with the first one due to self-focusing, with an effective focal length $f_{\rm foc}$ (see Fig. 6), and the second one due to diffraction, with an effective focal length of $f_{\rm defoc}$ (see Fig. 7).

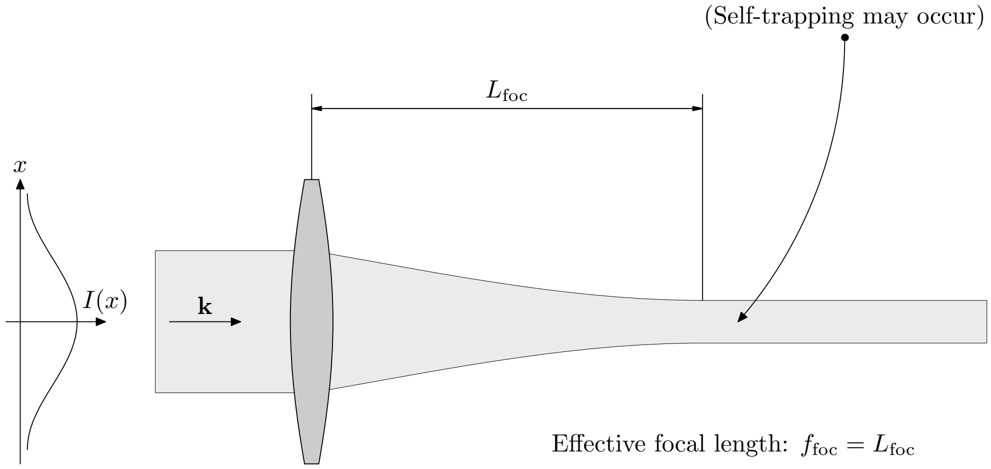

Figure 6. Self-focusing seen as an effective lensing of the optical beam.

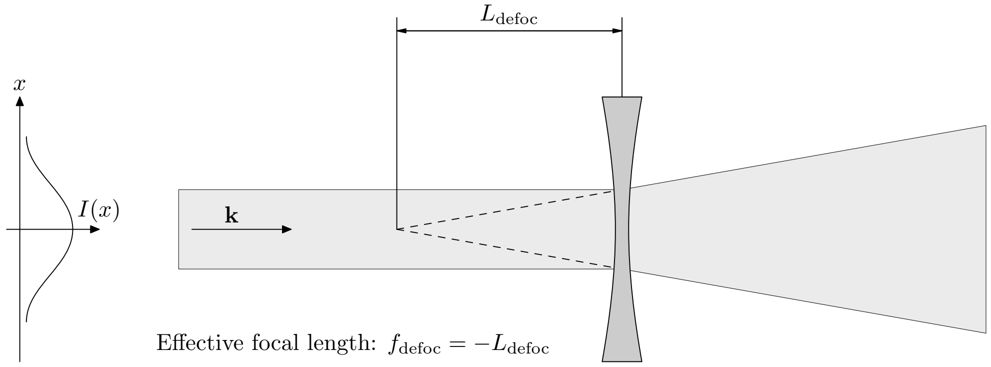

Figure 7. Diffraction seen as an effective defocusing of the optical beam.

Whenever these effects balance each other, we in this picture have the effective focal length $f_{\rm foc} + f_{\rm defoc} = 0$.

In the electromagnetic wave picture, the propagation of an optical continuous wave in optical Kerr-media is governed by the wave equation

$$\begin{aligned}\nabla\times\nabla\times\mathbf{E}_\omega(\mathbf{r}) - k^2\mathbf{E}_\omega(\mathbf{r}) &= \mu_0\omega^2\mathbf{P}^{(\rm NL)}_\omega(\mathbf{r})\\ &= \frac{3}{4}\frac{\omega^2}{c^2}\chi^{(3)}_{xxxx}|\mathbf{E}_\omega(\mathbf{r})|^2\mathbf{E}_\omega(\mathbf{r}),\end{aligned} \tag{4}$$

with $k = \omega n_0/c$, using notations as previously introduced in this course. For simplicity we will from now on consider the spatial extent of the beam in only one transverse Cartesian coordinate $x$.

By introducing the spatial envelope $\mathbf{A}_\omega(x,z)$ according to

$$\mathbf{E}_\omega(\mathbf{r}) = \mathbf{A}_\omega(x,z)\exp(ikz),$$

and using the slowly varying envelope approximation in the direction of propagation $z$, the wave equation (4) takes the form

$$i\frac{\partial\mathbf{A}_\omega(x,z)}{\partial z} + \frac{1}{2k}\frac{\partial^2\mathbf{A}_\omega(x,z)}{\partial x^2} = -\frac{\omega n_2}{c}|\mathbf{A}_\omega(x,z)|^2\mathbf{A}_\omega(x,z). \tag{5}$$

Notice the strong similarity between this equation for continuous wave propagation and the equation (3) for the envelope of a infinite plane wave pulse. The only significant difference, apart from the physical dimensions of the involved parameters, is that here nu additional first order derivative with respect to $x$ is present. In all other respects, Eqs. (3) and (5) are identical, if we interchange the roles of the time $t$ in Eq. (3) with the transverse spatial coordinate $x$ in Eq. (5).

While the sign of the dispersion parameter $\beta$ occurring in Eq. (3) has significance for the compression or broadening of the pulse, no such sign option appear in Eq. (5) for the spatial envelope of the continuous wave beam. This follows naturally, since the spatial broadening mechanism (in contrary to the temporal compression or broadening of the pulse) is due to diffraction, a non-reversible process which in nature always tend to broaden a collimated light beam.

As with Eq. (3) for the temporal pulse propagation, we may now for the continuous wave case cast Eq. (5) into a normalized form, by applying the change of variables

$$u = L\sqrt{\frac{n_2\omega k}{c}}A_\omega, \qquad s = x/L, \qquad \zeta = z/(kL^2),$$

where $L$ is some characteristic length of the evolution of the beam, usually taken as the transverse beam width, which gives the normalized form

$$\Big(i\frac{\partial}{\partial\zeta} + \frac{1}{2}\frac{\partial^2}{\partial s^2}\Big)u(\zeta,s) + |u(\zeta,s)|^2 u(\zeta,s) = 0. \qquad (6)$$

### Mathematical equivalence between temporal and spatial solitons

As seen in the above derivation of the normalized forms of the equations governing wave propagation of temporal and spatial solitons, they are described by exactly the same normalized nonlinear Schrödinger equation. The only difference between the two cases are the ways the normalization is being carried out. In the interpretation of the solutions to the nonlinear Schrödinger equation, the $s$ variable could for the temporal solitons be taken as a normalized time variable, while for the spatial solitons, the $s$ variable could instead be taken as a normalized transverse coordinate.

### Soliton solutions

The nonlinear Schrödinger equations given by Eqs. (3) and (6) possess infinitely many solutions, of which only a few are possible to obtain analytically. In the regime where $dv_{\rm g}/d\omega > 0$ (i. e. for which $\beta < 0$), an exact temporal soliton solution to Eq. (3) is though obtained when the pulse $u(\zeta,s)$ has the initial shape

$$u(0,s) = N\,\mathrm{sech}(s),$$

where $N \geq 1$ is an integer number. Depending on the value of $N$, solitons of different order can be formed, and the so-called "fundamental soliton" is given for $N = 1$. For higher values of $N$, the solitons are hence called "higher order solitons".

The first analytical solution to the nonlinear Schrödinger equation is given for $N = 1$ as[2]

$$u(\zeta,s) = \mathrm{sech}(s)\exp(i\zeta/2).$$

The shape of this fundamental solution is shown in Fig. 8.

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[2] Please note that there is a printing error in Butcher and Cotter's *The Elements of Nonlinear Optics* in their expression for this solution, on page 241, row 30, where their erroneous equation "$u(\zeta,s) = \mathrm{sech}(s)\exp(-i\zeta/2)$" should be replaced by the proper one, *without* the minus sign in the exponential.

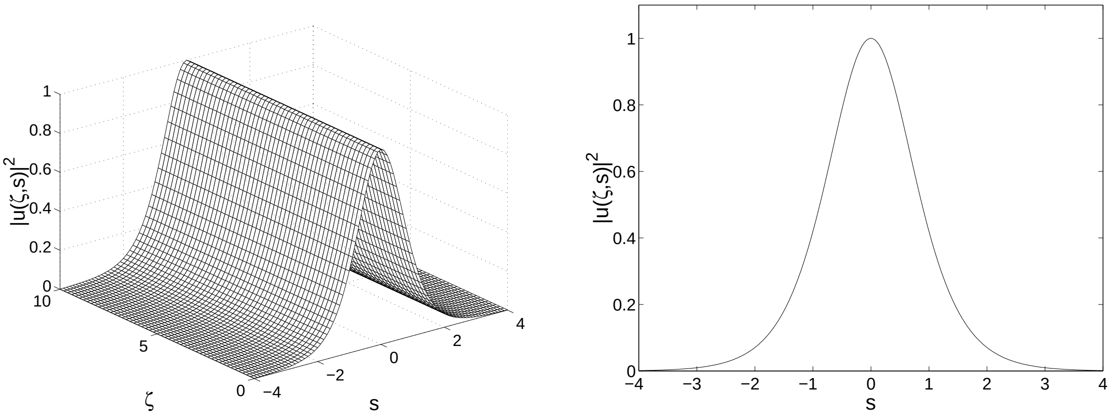

Figure 8. The fundamental bright soliton solution to the NLSE.

For higher order solitons, the behaviour is usually not stable with respect to the normalized distance $\zeta$, but rather of an oscillatory nature, as shown in Fig. 10.1 of the handed out material. (Figure 10.1 is copied from Govind P. Agrawal *Fiber-Optic Communication systems* (Wiley, New York, 1997).) This figure shows the fundamental soliton together with the third order ($N = 3$) soliton, and one can see that there is a continuous, oscillatory energy transfer in the $s$-direction of the pulse. (See also Butcher and Cotter's Fig. 7.8 on page 242, where the $N = 4$ soliton is shown.)

The solutions so far discussed belong to a class called "bright solitons". The reason for using the term "bright soliton" becomes more clear if we consider another type of solutions to the nonlinear Schrdinger equation, namely the "dark" solitons, given as the solutions

$$u(\zeta, s) = [\eta \tanh(\eta(s - \kappa\zeta)) - i\kappa] \exp(iu_0^2\zeta),$$

with $u_0$ being the normalized amplitude of the continuous-wave background, $\phi$ is an internal phase angle in the range $0 \le \phi \le \pi/2$, and

$$\eta = u_0 \cos\phi, \qquad \kappa = u_0 \sin\phi.$$

For the dark solitons, one makes a distinction between the "black" soliton for $\phi = 0$, which drops down to zero intensity in the middle of the pulse, and the "grey" solitons for $\phi \neq 0$, which do not drop down to zero. For the black solitons, the solution for $\phi = 0$ takes ths simpler form

$$u(\zeta, s) = u_0 \tanh(u_0 s) \exp(iu_0^2\zeta).$$

The shape of the black fundamental soliton is shown in Fig. 9.

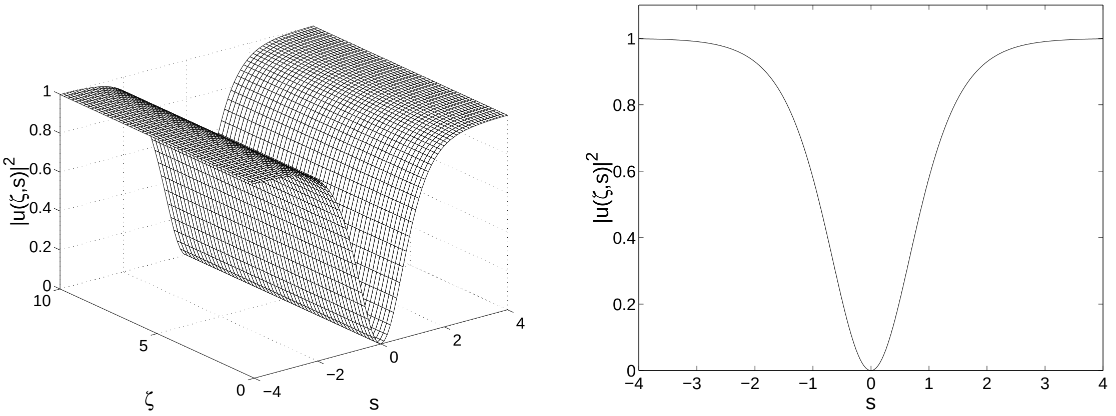

Figure 9. The fundamental dark (black) soliton solution to the NLSE.

Another important difference between the bright and the dark soliton, apart from their obvious difference in appearances, is that the velocity of a dark soliton depends on its amplitude, through the internal phase angle $u_0^2\zeta$. This is not the case for the bright solitons, which propagate with the same velocity irregardless of the amplitude.

The darks soliton is easily pictured as a dark travelling pulse in an otherwise continuous level background intensity. The described dark solitons, however, are equally well applied to spatial solitons as well, for the case $n_2 > 0$, where a dark center of the beam causes a slightly *lower* refractive index than for the illuminated surroundings, hence generating an effective "anti-waveguide" that compensates for the diffraction experianced by the black center.

### General travelling wave solutions

It should be emphasized that the nonlinear Schrödinger equation permits travelling wave solutions as well. On example of such an exact solution is given by

$$u(\zeta, s) = a\,\mathrm{sech}[a(s - c\zeta/\sqrt{2})]\exp[ic(s\sqrt{2} - c\zeta)/2 + in\zeta]$$

where $n = (1/2)(a^2 + c^2/2)$. That this in fact *is* a solution to the nonlinear Schrödinger equation,

$$\Big(i\frac{\partial}{\partial\zeta} + \frac{1}{2}\frac{\partial^2}{\partial s^2}\Big)u(\zeta, s) + |u(\zeta, s)|^2 u(\zeta, s) = 0,$$

(here for simplicity taken for the special case $\mathrm{sgn}(\beta) = -1$) is straightforward to verify by, for example, using the following MapleV blocks:

```
restart:
assume(s,real);
assume(zeta,real);
assume(a,real);
assume(c,real);
n:=(1/2)*(a^2+c^2/2);
u(zeta,s):=a*sech(a*(s*sqrt(2)-c*zeta)/sqrt(2))
            *exp(I*((c/2)*(s*sqrt(2)-c*zeta)+n*zeta));
nlse:=I*diff(u(zeta,s),zeta)+(1/2)*diff(u(zeta,s),s$2)
            +conjugate(u(zeta,s))*u(zeta,s)^2;
simplify(nlse);
```

For further information regarding travelling wave solutions and higher order soliton solutions to the nonlinear Schrödinger equation, see P. G. Johnson and R. S. Drazin, *Solitons: an introduction* (Cambridge Univrsity Press, Cambridge, 1989).

### Soliton interactions

One can understand the implications of soliton interaction by solving the NLSE numerically with the input amplitude consisting of a soliton pair

$$u(0, \tau) = \mathrm{sech}(\tau - q_0) + r\,\mathrm{sech}[r(\tau + q_0)]\exp(i\vartheta)$$

with, as previously, $\mathrm{sech}(x) \equiv 1/\cosh(x)$, and $r$ is the relative amplitude of the second soliton with respect to the other, $\vartheta$ the phase difference, and $2q_0$ the initial, normalized separation between the solitons.

A set of computer generated solutions to this pair of initial soliton shapes are shown in the handed-out Fig. 10.6 of Govind P. Agrawal *Fiber-Optic Communication systems* (Wiley, New York, 1997). In this figure, the upper left graph shows that a pair of solitons may, as a matter of fact, attract each other, forming a soliton pair which oscillate around the center of the moving reference frame.

Another interesting point is that soliton pairs may be formed by spatial solitons as well. In Fig. 9 of the handed-out material, the self-trapping of two spatial solitons, launched with initial trajectories that do not lie in the same plane, are shown. In this experiment, carried out by Mitchell et. al. at Princeton[3], the two solitons start spiraling around each other in a helix, experiencing attractive forces that together with the orbital momentum carried by the pulses form a stable configuration.

### Dependence on initial conditions

For a real situation, one might ask oneself how sensitive the forming of solitons is, depending on perturbations on the preferred sech($s$) initial shape. In a real situation, for example, we will rarely be able to construct the exact pulse form required for launching a pulse that will possess the soliton properties already from the beginning.

As a matter of fact, the soliton formation process accepts quite a broad range of initial pulse shapes, and as long as the initial intensity is sufficiently well matched to the energy content of the propagating soliton, the generated soliton is remarkable stable against perurbations. In a functional theoretical analogy, we may call this the soliton "acceptance angle" of initial functions that will be accepted for soliton formation in a medium.

In order to illustrate the soliton formation, one may study Figs. 10.2 and 10.3 of Govind P. Agrawal *Fiber-Optic Communication systems* (Wiley, New York, 1997)[4] In Fig. 10.2, the input pulse shape is a Gaussian, rather than the natural sech($s$) initial shape. As can be seen in the figure, the pulse shape gradually change towards the fundamental soliton, even though the Gaussian shape is a quite bad approximation to the final sech($s$) form.

The forming of the soliton does not only depend on the initial shape of the pulse, but also on the peak intensity of the pulse. In Fig. 10.3, an ideal sech($s$) pulse shape, though with a 20 percent higher pulse amplitude than the ideal one of unity, is used as input. In this case the pulse slightly oscillate in amplitude during the propagation, but finally approaching the fundamental soliton solution.

Finally, as being an example of an even worse approximation to the sech($s$) shape, a square input pulse can also generate solitons, as shown in the handed-out Fig. 16 of *Beam Shaping and Control with Nonlinear Optics*, Eds. F. Kajzar and R. Reinisch (Plenum Press, New York, 1998).

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[3] M. Mitchell, Z. Chen, M. Shih, and M. Sageev, Phys. Rev. Lett. **77**, 490 (1996).

[4] The same pictures can be found in Govind P. Agrawal *Nonlinear Fiber Optics* (Academic Press, New York, 1989).

# Lecture XI

**Lecture Notes on Nonlinear Optics**
Nonlinear Optics (5A5513, 5p for advanced undergraduate and doctoral students)
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## Lecture 11

In this lecture, we will focus on configurations where the angular frequency of the light is close to some transition frequency of the medium. In particular, we will start with a brief outline of how the non-resonant susceptibilities may be modified in such a way that weakly resonant interactions can be taken into account. Having formulated the susceptibilities at weakly resonant interaction, we will proceed with formulating a non-perturbative approach of calculation of the polarization density of the medium. For the two-level system, this results in the Bloch equations governing resonant interaction between light and matter.

The outline for this lecture is:
- Singularities of the non-resonant susceptibilities
- Alternatives to perturbation analysis of the polarization density
- Relaxation of the medium
- The two-level system and the Bloch equation
- The resulting polarization density of the medium at resonance

### Singularities of non-resonant susceptibilities

In the theory described so far in this course, all interactions have for simplicity been considered as non-resonant. The explicit forms of the susceptibilities, in terms of the electric dipole moments and transition frequencies of the molecules, have been obtained in lecture six, of the forms

$$\chi^{(1)}_{\mu\alpha}(-\omega;\omega) \sim \frac{r^{\mu}_{ab} r^{\alpha}_{ba}}{\Omega_{ba}-\omega} + \{\text{similar terms}\}, \qquad \text{[B.\,\&\,C.\,(4.58)]}$$

$$\chi^{(2)}_{\mu\alpha\beta}(-\omega_{\sigma};\omega_1,\omega_2) \sim \frac{r^{\mu}_{ab} r^{\alpha}_{bc} r^{\beta}_{ca}}{(\Omega_{ba}-\omega_1-\omega_2)(\Omega_{ca}-\omega_2)} + \{\text{similar terms}\}, \qquad \text{[B.\,\&\,C.\,(4.63)]}$$

$$\chi^{(3)}_{\mu\alpha\beta\gamma}(-\omega_{\sigma};\omega_1,\omega_2,\omega_3) \sim \frac{r^{\mu}_{ab} r^{\alpha}_{bc} r^{\beta}_{cd} r^{\gamma}_{da}}{(\Omega_{ba}-\omega_1-\omega_2-\omega_3)(\Omega_{ca}-\omega_2-\omega_3)(\Omega_{da}-\omega_3)} + \{\text{similar terms}\}, \qquad \text{[B.\,\&\,C.\,(4.64)]}$$

$$\vdots$$

To recapitulate, these forms have all been derived under the assumption that the Hamiltonian (which is the general operator which describes the state of the system) consist only of a thermal equilibrium part and an interaction part (in the electric dipolar approximation), of the form

$$\hat{H} = \hat{H}_0 + \hat{H}_{\mathrm{I}}(t).$$

This is a form which clearly does not contain any term related to relaxation effects of the medium, that is to say, it does not contain any term describing any energy flow into thermal heat. As long as we consider the interaction part of the Hamiltonian to be sufficiently strong compared to any relaxation effect of the medium, this is a valid approximation.

However, the problem with the non-resonant forms of the susceptibilities clearly comes into light when we consider an angular frequency of the light that is close to a transition frequency of the system, since for the first order susceptibility,

$$\chi^{(1)}_{\mu\alpha}(-\omega;\omega) \to \infty, \quad \text{when } \omega \to \Omega_{ba},$$

or for the second order susceptibility,

$$\chi^{(2)}_{\mu\alpha\beta}(-\omega;\omega_1,\omega_2)\to\infty,\quad \text{when } \omega_1+\omega_2\to\Omega_{ba} \text{ or } \omega_2\to\Omega_{ca}.$$

This clearly non-physical behaviour is a consequence of that the denominators of the rational expressions for the susceptibilities have singularities at the resonances, and the aim with this lecture is to show how these singularities can be removed.

### Modification of the Hamiltonian for resonant interaction

Whenever we have to consider relaxation effects of the medium, as in the case of resonant interactions, the Hamiltonian should be modified to

$$\hat{H}=\hat{H}_0+\hat{H}_{\text{I}}(t)+\hat{H}_{\text{R}}, \tag{1}$$

where, as previously, $\hat{H}_0$ is the Hamiltonian in the absence of external forces, $\hat{H}_{\text{I}}(t)=-\hat{Q}_\alpha E_\alpha(\mathbf{r},t)$ is the interaction Hamiltonian (here taken in the Schrödinger picture, as described in lecture four), being linear in the applied electric field of the light, and where the new term $\hat{H}_{\text{R}}$ describes the various relaxation processes that brings the system into the thermal equilibrium whenever external forces are absent. The state of the system (atom, molecule, or general ensemble) is then conveniently described by the density operator formalism, from which we can obtain macroscopically observable parameters of the medium, such as the electric polarization density (as frequently encountered in this course), the magnetization of the medium, current densities, etc.

The form (1) of the Hamiltonian is now to be analysed by means of the equation of motion of the density operator $\hat{\rho}$,

$$i\hbar\frac{d\hat{\rho}}{dt}=\hat{H}\hat{\rho}-\hat{\rho}\hat{H}=[\hat{H},\hat{\rho}], \tag{2}$$

and depending on the setup, this equation may be solved by means of perturbation analysis (for non-resonant and weakly resonant interactions), or by means of non-perturbative approaches, such as the Bloch equations (for strongly resonant interactions).

### Phenomenological representation of relaxation processes

In many cases, the relaxation process of the medium towards thermal equilibrium can be described by

$$[\hat{H}_{\text{R}},\hat{\rho}]=-i\hbar\hat{\Gamma}(\hat{\rho}-\hat{\rho}_0),$$

where $\hat{\rho}_0$ is the thermal equilibrium density operator of the system. The here phenomenologically introduced operator $\hat{\Gamma}$ describes the relaxation of the medium, and can can be considered as being independent of the interaction Hamiltonian. Here the operator $\hat{\Gamma}$ has the physical dimension of an angular frequency, and its matrix elements can be considered as giving the time constants of decay for various states of the system.

### Perturbation analysis of weakly resonant interactions

Before entering the formalism of the Bloch equations for strongly resonant interactions, we will outline the weakly resonant interactions in a perturbative analysis for the susceptibilities, as previously developed in lectures three, four, and five.

By taking the perturbation series for the density operator as

$$\hat{\rho}(t)=\underbrace{\hat{\rho}_0}_{\sim[E(t)]^0}+\underbrace{\hat{\rho}_1(t)}_{\sim[E(t)]^1}+\underbrace{\hat{\rho}_2(t)}_{\sim[E(t)]^2}+\ldots+\underbrace{\hat{\rho}_n(t)}_{\sim[E(t)]^n}+\ldots,$$

as we previously did for the strictly non-resonant case, one obtains the system of equations

$$\begin{aligned}
i\hbar\frac{d\hat{\rho}_0}{dt} &= [\hat{H}_0,\hat{\rho}_0],\\
i\hbar\frac{d\hat{\rho}_1(t)}{dt} &= [\hat{H}_0,\hat{\rho}_1(t)]+[\hat{H}_{\mathrm{I}}(t),\hat{\rho}_0]-i\hbar\hat{\Gamma}\hat{\rho}_1(t),\\
i\hbar\frac{d\hat{\rho}_2(t)}{dt} &= [\hat{H}_0,\hat{\rho}_2(t)]+[\hat{H}_{\mathrm{I}}(t),\hat{\rho}_1(t)]-i\hbar\hat{\Gamma}\hat{\rho}_2(t),\\
&\vdots\\
i\hbar\frac{d\hat{\rho}_n(t)}{dt} &= [\hat{H}_0,\hat{\rho}_n(t)]+[\hat{H}_{\mathrm{I}}(t),\hat{\rho}_{n-1}(t)]-i\hbar\hat{\Gamma}\hat{\rho}_n(t),\\
&\vdots
\end{aligned}$$

As in the non-resonant case, one may here start with solving for the zeroth order term $\hat{\rho}_0$, with all other terms obtained by consecutively solving the equations of order $j=1,2,\dots,n$, in that order.

Proceeding in exactly the same path as for the non-resonant case, solving for the density operator in the interaction picture and expressing the various terms of the electric polarization density in terms of the corresponding traces

$$P_\mu(\mathbf{r},t)=\sum_{n=0}^{\infty}P_\mu^{(n)}(\mathbf{r},t)=\frac{1}{V}\sum_{n=0}^{\infty}\mathrm{Tr}[\hat{\rho}_n(t)\hat{Q}_\mu],$$

one obtains the linear, first order susceptibility of the form

$$\chi^{(1)}_{\mu\alpha}(-\omega;\omega)=\frac{Ne^2}{\varepsilon_0\hbar}\sum_a\varrho_0(a)\sum_b\Big(\frac{r^\mu_{ab}r^\alpha_{ba}}{\Omega_{ba}-\omega-i\Gamma_{ba}}+\frac{r^\alpha_{ab}r^\mu_{ba}}{\Omega_{ba}+\omega-i\Gamma_{ba}}\Big).$$

Similarly, the second order susceptibility for weakly resonant interaction is obtained as

$$\begin{aligned}
&\chi^{(2)}_{\mu\alpha\beta}(-\omega_\sigma;\omega_1,\omega_2)\\
&\quad=\frac{Ne^3}{\varepsilon_0\hbar^2}\frac{1}{2!}\mathbf{S}\sum_a\varrho_0(a)\sum_b\sum_c\Big\{\frac{r^\mu_{ab}r^\alpha_{bc}r^\beta_{ca}}{(\Omega_{ac}+\omega_2-i\Gamma_{ac})(\Omega_{ab}+\omega_\sigma-i\Gamma_{ab})}\\
&\qquad-\frac{r^\alpha_{ab}r^\mu_{bc}r^\beta_{ca}}{(\Omega_{ac}+\omega_2-i\Gamma_{ac})(\Omega_{bc}+\omega_\sigma-i\Gamma_{bc})}-\frac{r^\beta_{ab}r^\mu_{bc}r^\alpha_{ca}}{(\Omega_{ba}+\omega_2-i\Gamma_{ba})(\Omega_{bc}+\omega_\sigma-i\Gamma_{bc})}\\
&\qquad+\frac{r^\beta_{ab}r^\alpha_{bc}r^\mu_{ca}}{(\Omega_{ba}+\omega_2-i\Gamma_{ba})(\Omega_{ca}+\omega_\sigma-i\Gamma_{ca})}\Big\}.
\end{aligned}$$

In these expressions for the susceptibilities, the singularities at resonance are removed, and the spectral properties of the absolute values of the susceptibilities are described by regular Lorenzian line shapes.

The values of the matrix elements $\Gamma_{mn}$ are in many cases difficult to derive from a theoretical basis; however, they are often straightforward to obtain by regular curve-fitting and regression analysis of experimental data.

As seen from the expressions for the susceptibilities above, we still have a boosting of them close to resonance (resonant enhancement). However, the values of the susceptibilities reach a plateau at exact resonance, with maximum values determined by the magnitudes of the involved matrix elements $\Gamma_{mn}$ of the relaxation operator.

## Validity of perturbation analysis of the polarization density

Strictly speaking, the perturbative approach is only to be considered as for an infinite series expansion. For a limited number of terms, the perturbative approach is only an approximative method, which though for many cases is sufficient.

The perturbation series, in the form that we have encountered it in this course, defines a power series in the applied electric field of the light, and as long as the lower order terms are dominant in the expansion, we may safely neglect the higher order ones. Whenever we encounter strong fields, however, we may run into trouble with the series expansion, in particular if we are in a resonant optical regime, with a boosting effect of the polarization density of the medium. (This boosting effect can be seen as the equivalent to the close-to-resonance behaviour of the mechanical spring model under influence of externally driving forces.)

As an illustration to this source of failure of the model in the presence of strong electrical fields, we may consider another, more simple example of series expansions, namely the Taylor expansion of the function $\sin(x)$ around $x \approx 0$, as shown in Fig. 1.

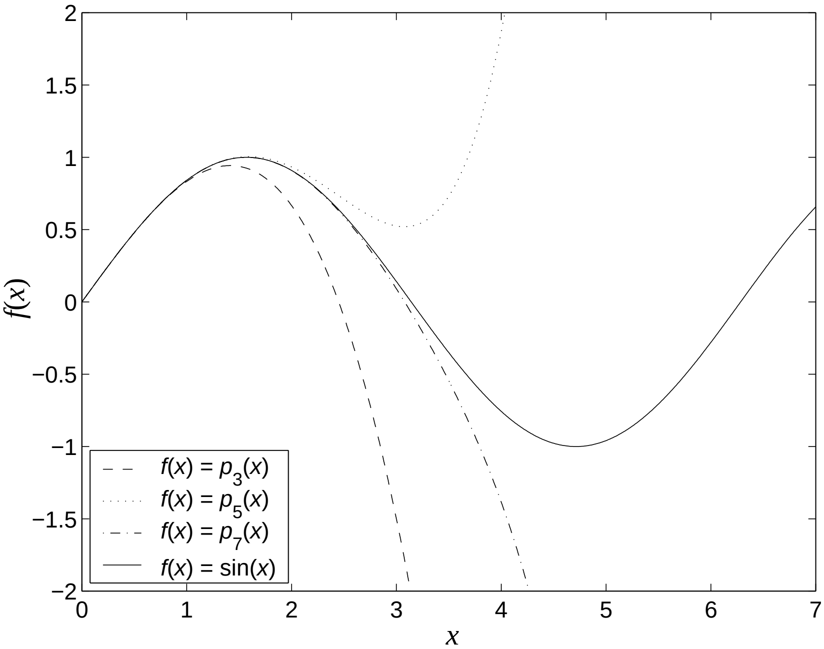

Figure 1. Approximations to $f(x) = \sin(x)$ by means of power series expansions of various degrees.

In analogy to the susceptibility formalism, we may consider $x$ to have the role of the electric field (the variable which we make the power expansion in terms of), and $\sin(x)$ to have the role of the polarization density or the density operator (simply the function we wish to analyze). For low numerical values of $x$, up to about $x \approx 1$, the $\sin(x)$ function is well described by keeping only the first two terms of the expansion, corresponding to a power expansion up to and including order three,

$$\sin(x) \approx p_3(x) = x - \frac{x^3}{3!}.$$

For higher values of $x$, say up to about $x \approx 2$, the expansion is still following the exact function to a good approximation if we include also the third term, corresponding to a power expansion up to and including order five,

$$\sin(x) \approx p_5(x) = x - \frac{x^3}{3!} + \frac{x^5}{5!}.$$

This necessity of including higher and higher order terms goes on as we increase the value of $x$, and we can from the graph also see that the breakdown at a certain level of approximation causes severe difference between the approximate and exact curves. In particular, if one wish to calculate the value of the function $\sin(x)$ for small $x$, it might be a good idea to apply the series expansion.

For greater values of $x$, say $x \approx 10$, the series expansion approach is, however, a bad idea, and an efficient evaluation of $\sin(x)$ requires another approach.

As a matter of fact, the same arguments hold for the more complex case of the series expansion of the density operator[1], for which we for high intensities (high electrical field strengths) must include higher order terms as well.

However, we have seen that even in the non-resonant case, we may encounter great algebraic complexity even in low order nonlinear terms, and since the problem of formulating a proper polarization density is expanding more or less exponentially with the order of the nonlinearity, the usefulness of the susceptibility formalism eventually breaks down. The solution to this problem is to identify the relevant transitions of the ensemble, and to solve the equation of motion (2) exactly instead (or at least within other levels of approximation which do not rely on the perturbative foundation of the susceptibility formalism).

### The two-level system

In many cases, the interaction between light and matter can be reduced to that of a two-level system, consisting of only two energy eigenstates $|a\rangle$ and $|b\rangle$. The equation of motion of the density operator is generally given by Eq. (2) as

$$i\hbar\frac{d\hat{\rho}}{dt} = [\hat{H}, \hat{\rho}],$$

with

$$\hat{H} = \hat{H}_0 + \hat{H}_{\mathrm{I}}(t) + \hat{H}_{\mathrm{R}}.$$

For the two-level system, the equation of motion can be expressed in terms of the matrix elements of the density operator as

$$i\hbar\frac{d\rho_{aa}}{dt} = [\hat{H}_0, \hat{\rho}]_{aa} + [\hat{H}_{\mathrm{I}}(t), \hat{\rho}]_{aa} + [\hat{H}_{\mathrm{R}}, \hat{\rho}]_{aa}, \tag{3a}$$

$$i\hbar\frac{d\rho_{ab}}{dt} = [\hat{H}_0, \hat{\rho}]_{ab} + [\hat{H}_{\mathrm{I}}(t), \hat{\rho}]_{ab} + [\hat{H}_{\mathrm{R}}, \hat{\rho}]_{ab}, \tag{3b}$$

$$i\hbar\frac{d\rho_{bb}}{dt} = [\hat{H}_0, \hat{\rho}]_{bb} + [\hat{H}_{\mathrm{I}}(t), \hat{\rho}]_{bb} + [\hat{H}_{\mathrm{R}}, \hat{\rho}]_{bb}, \tag{3c}$$

where the fourth equation for $\rho_{ba}$ was omitted, since the solution for this element immediately follows from

$$\rho_{ba} = \rho_{ab}^*.$$

*Terms involving the thermal equilibrium Hamiltonian*

The system of Eqs. (3) is the starting point for derivation of the so-called Bloch equations. Starting with the thermal-equilibrium part of the commutators in the right-hand sides of Eqs. (3), we have for the diagonal elements

$$\begin{aligned}
[\hat{H}_0, \hat{\rho}]_{aa} &= \langle a|\hat{H}_0\hat{\rho}|a\rangle - \langle a|\hat{\rho}\hat{H}_0|a\rangle \\
&= \sum_k \underbrace{\langle a|\hat{H}_0|k\rangle}_{=\mathbb{E}_a\delta_{ak}}\langle k|\hat{\rho}|a\rangle - \sum_j \langle a|\hat{\rho}|j\rangle \underbrace{\langle j|\hat{H}_0|a\rangle}_{=\mathbb{E}_j\delta_{ja}} \\
&= \mathbb{E}_a\rho_{aa} - \rho_{aa}\mathbb{E}_a \\
&= 0 \\
&= [\hat{H}_0, \hat{\rho}]_{bb},
\end{aligned}$$

---

[1] We may recall that the series expansion of the density operator is *the* very origin of the expansion of the polarization density of the medium in terms of the electric field, and hence also the very foundation for the whole susceptibility formalism as described in this course.

and for the off-diagonal elements

$$\begin{aligned}
[\hat{H}_0, \hat{\rho}]_{ab} &= \langle a|\hat{H}_0\hat{\rho}|b\rangle - \langle a|\hat{\rho}\hat{H}_0|b\rangle \\
&= \sum_k \underbrace{\langle a|\hat{H}_0|k\rangle}_{=\mathbb{E}_a\delta_{ak}}\langle k|\hat{\rho}|b\rangle - \sum_j \langle a|\hat{\rho}|j\rangle \underbrace{\langle j|\hat{H}_0|b\rangle}_{=\mathbb{E}_j\delta_{jb}} \\
&= \mathbb{E}_a\rho_{ab} - \rho_{ab}\mathbb{E}_b \\
&= -(\mathbb{E}_b - \mathbb{E}_a)\rho_{ab} \\
&= -\hbar\Omega_{ba}\rho_{ab}
\end{aligned}$$

*Terms involving the interaction Hamiltonian*

For the commutators in the right-hand sides of Eqs. (3) involving the interaction Hamiltonian, we similarly have for the diagonal elements

$$\begin{aligned}
[\hat{H}_{\mathrm{I}}(t), \hat{\rho}]_{aa} &= \langle a|(-e\hat{r}_\alpha E_\alpha(\mathbf{r},t))\hat{\rho}|a\rangle - \langle a|\hat{\rho}(-e\hat{r}_\alpha E_\alpha(\mathbf{r},t))|a\rangle \\
&= -eE_\alpha(\mathbf{r},t)\left\{ \sum_k \langle a|\hat{r}_\alpha|k\rangle\langle k|\hat{\rho}|a\rangle - \sum_j \langle a|\hat{\rho}|j\rangle\langle j|\hat{r}_\alpha|a\rangle \right\} \\
&= -eE_\alpha(\mathbf{r},t)\left\{ r^\alpha_{aa}\rho_{aa} + r^\alpha_{ab}\rho_{ba} - \rho_{aa}r^\alpha_{aa} - \rho_{ab}r^\alpha_{ba} \right\} \\
&= -e(r^\alpha_{ab}\rho_{ba} - r^\alpha_{ba}\rho_{ab})E_\alpha(\mathbf{r},t) \\
&= -[\hat{H}_{\mathrm{I}}(t), \hat{\rho}]_{bb},
\end{aligned}$$

and for the off-diagonal elements

$$\begin{aligned}
[\hat{H}_{\mathrm{I}}(t), \hat{\rho}]_{ab} &= \langle a|(-e\hat{r}_\alpha E_\alpha(\mathbf{r},t))\hat{\rho}|b\rangle - \langle a|\hat{\rho}(-e\hat{r}_\alpha E_\alpha(\mathbf{r},t))|b\rangle \\
&= -eE_\alpha(\mathbf{r},t)\left\{ \sum_k \langle a|\hat{r}_\alpha|k\rangle\langle k|\hat{\rho}|b\rangle - \sum_j \langle a|\hat{\rho}|j\rangle\langle j|\hat{r}_\alpha|b\rangle \right\} \\
&= -eE_\alpha(\mathbf{r},t)\left\{ r^\alpha_{aa}\rho_{ab} + r^\alpha_{ab}\rho_{bb} - \rho_{aa}r^\alpha_{ab} - \rho_{ab}r^\alpha_{bb} \right\} \\
&= -er^\alpha_{ab}E_\alpha(\mathbf{r},t)(\rho_{bb} - \rho_{aa}) - e(r^\alpha_{aa} - r^\alpha_{bb})E_\alpha(\mathbf{r},t)\rho_{ab} \\
&= \{\text{Optical Stark shift} : \delta\mathbb{E}_k \equiv -er^\alpha_{kk}E_\alpha(\mathbf{r},t), \quad k = a, b\} \\
&= -er^\alpha_{ab}E_\alpha(\mathbf{r},t)(\rho_{bb} - \rho_{aa}) + (\delta\mathbb{E}_a - \delta\mathbb{E}_b)\rho_{ab}.
\end{aligned}$$

*Terms involving relaxation processes*

For the commutators describing relaxation processes, the diagonal elements are given as

$$\begin{aligned}
[\hat{H}_{\mathrm{R}}, \hat{\rho}]_{aa} &= -i\hbar(\rho_{aa} - \rho_0(a))/T_a, \\
[\hat{H}_{\mathrm{R}}, \hat{\rho}]_{bb} &= -i\hbar(\rho_{bb} - \rho_0(b))/T_b,
\end{aligned}$$

where $T_a$ and $T_b$ are the decay rates towards the thermal equilibrium at respective level, and where $\rho_0(a)$ and $\rho_0(b)$ are the thermal equilibrium values of $\rho_{aa}$ and $\rho_{bb}$, respectively (i. e. the thermal equilibrium population densities of the respective level). The off-diagonal elements are similarly given as

$$\begin{aligned}
[\hat{H}_{\mathrm{R}}, \hat{\rho}]_{ab} &= -i\hbar\rho_{ab}/T_2, \\
[\hat{H}_{\mathrm{R}}, \hat{\rho}]_{ba} &= -i\hbar\rho_{ba}/T_2.
\end{aligned}$$

A common approximation is to consider the two states $|a\rangle$ and $|b\rangle$ to be sufficiently similar in order to approximate their lifetimes as equal, i. e. $T_a \approx T_b \approx T_1$, where $T_1$ for historical reasons is denoted as the *longitudinal relaxation time*. For the same historical reason, the relaxation time $T_2$ is denoted as the *transverse relaxation time*.[2]

As the above matrix elements of the commutators involving the various terms of the Hamiltonian are inserted into the right-hand sides of Eqs. (3), one obtains the following system of equations for the matrix elements of the density operator,

$$i\hbar\frac{d\rho_{aa}}{dt} = -e(r^{\alpha}_{ab}\rho_{ba} - r^{\alpha}_{ba}\rho_{ab})E_{\alpha}(\mathbf{r},t) - i\hbar(\rho_{aa} - \rho_0(a))/T_a, \tag{4a}$$

$$i\hbar\frac{d\rho_{ab}}{dt} = -\hbar\Omega_{ba}\rho_{ab} - er^{\alpha}_{ab}E_{\alpha}(\mathbf{r},t)(\rho_{bb} - \rho_{aa}) + (\delta\mathbb{E}_a - \delta\mathbb{E}_b)\rho_{ab} - i\hbar\rho_{ab}/T_2, \tag{4b}$$

$$i\hbar\frac{d\rho_{bb}}{dt} = e(r^{\alpha}_{ab}\rho_{ba} - r^{\alpha}_{ba}\rho_{ab})E_{\alpha}(\mathbf{r},t) - i\hbar(\rho_{bb} - \rho_0(b))/T_b. \tag{4c}$$

(The system of equations (4) corresponds to Butcher and Cotter's Eqs. (6.35).) So far, the applied electric field of the light is allowed to be of arbitrary form. However, in order to simplify the following analysis, we will assume the light to be linearly polarized and quasimonochromatic, of the form

$$E_{\alpha}(\mathbf{r},t) = \mathrm{Re}[E^{\alpha}_{\omega}(t)\exp(-i\omega t)].$$

We will in addition assume the slowly varying temporal envelope $E^{\alpha}_{\omega}(t)$ to be real-valued, and we will also neglect the optical Stark shifts $\delta\mathbb{E}_a$ and $\delta\mathbb{E}_b$. In the absence of strong static magnetic fields, we may also assume the matrix elements $er^{\alpha}_{ab}$ to be real-valued. When these assumptions and approximations are applied to the equations of motion (4), one obtains

$$\frac{d\rho_{aa}}{dt} = i(\rho_{ba} - \rho_{ab})\beta(t)\cos(\omega t) - (\rho_{aa} - \rho_0(a))/T_a, \tag{5a}$$

$$\frac{d\rho_{ab}}{dt} = i\Omega_{ba}\rho_{ab} + i\beta(t)\cos(\omega t)(\rho_{bb} - \rho_{aa}) - \rho_{ab}/T_2, \tag{5b}$$

$$\frac{d\rho_{bb}}{dt} = -i(\rho_{ba} - \rho_{ab})\beta(t)\cos(\omega t) - (\rho_{bb} - \rho_0(b))/T_b, \tag{5c}$$

where the *Rabi frequency* $\beta(t)$, defined in terms of the spatial envelope of the electrical field and the transition dipole moment as

$$\beta(t) = er^{\alpha}_{ab}E^{\alpha}_{\omega}(t)/\hbar = e\mathbf{r}_{ab}\cdot\mathbf{E}_{\omega}(t)/\hbar,$$

was introduced.

### The rotating-wave approximation

In the middle equation of the system (5), we have a time-derivative of $\rho_{ab}$ in the left-hand side, while we in the right-hand side have a term $i\Omega_{ba}\rho_{ab}$. Seen as the homogeneous part of a linear differential equation, this suggests that we may further simplify the equations of motion by taking a new variable $\rho^{\Omega}_{ab}$ according to the variable substitution

$$\rho_{ab} = \rho^{\Omega}_{ab}\exp[i(\Omega_{ba} - \Delta)t], \tag{6}$$

where $\Delta \equiv \Omega_{ba} - \omega$ is the detuning of the angular frequency of the light from the transition frequency $\Omega_{ba} \equiv (\mathbb{E}_b - \mathbb{E}_a)/\hbar$.

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[2] For a deeper discusssion and explanation of the various mechanisms involved in relaxation, see for example Charles P. Slichter, *Principles of Magnetic Resonance* (Springer-Verlag, Berlin, 1978), available at KTHB. This reference is not mentioned in Butcher and Cotters book, but it is a very good text on relaxation phenomena and how to incorporate them into a density-functional description of interaction between light and matter.

By inserting Eq. (6) into Eqs. (5), keeping in mind that $\rho_{ba} = \rho^*_{ab}$, one obtains the system

$$\frac{d\rho_{aa}}{dt} = i(\rho^{\Omega}_{ba} \exp[-i(\Omega_{ba} - \Delta)t] - \rho^{\Omega}_{ab} \exp[i(\Omega_{ba} - \Delta)t])\beta(t)\cos(\omega t) - (\rho_{aa} - \rho_0(a))/T_a, \tag{6a}$$

$$\frac{d\rho^{\Omega}_{ab}}{dt} = i\Delta\rho^{\Omega}_{ab} + i\beta(t)\cos(\omega t)\exp[-i(\Omega_{ba} - \Delta)t](\rho_{bb} - \rho_{aa}) - \rho^{\Omega}_{ab}/T_2, \tag{6b}$$

$$\frac{d\rho_{bb}}{dt} = -i(\rho^{\Omega}_{ba} \exp[-i(\Omega_{ba} - \Delta)t] - \rho^{\Omega}_{ab} \exp[i(\Omega_{ba} - \Delta)t])\beta(t)\cos(\omega t) - (\rho_{bb} - \rho_0(b))/T_b, \tag{6c}$$

The idea with the rotating-wave approximation is now to separate out rapidly oscillating terms of angular frequencies $\omega + \Omega_{ba}$ and $-(\omega + \Omega_{ba})$, and neglect these terms, compared with more slowly varying terms. The motivation for this approximation is that whenever high-frequency components appear in the equations of motions, the high-frequency terms will when integrated contain large denominators, and will hence be minor in comparison with terms with a slow variation. In some sense we can also see this as a temporal averaging procedure, where rapidly oscillating terms average to zero rapidly compared to slowly varying (or constant) components.

For example, in Eq. (6b), the product of the $\cos(\omega t)$ and the exponential function is approximated as

$$\begin{aligned}\cos(\omega t)\exp[-i(\Omega_{ba} - \Delta)t] &= \frac{1}{2}[\exp(i\omega t) + \exp(-i\omega t)]\exp[-i\underbrace{(\Omega_{ba} - \Delta)}_{=\omega}t]\\ &= \frac{1}{2}[1 + \exp(-i2\omega t)] \to \frac{1}{2},\end{aligned}$$

while in Eqs. (6a) and (6c), the same argument gives

$$\begin{aligned}\exp[i(\Omega_{ba} - \Delta)t]\cos(\omega t) &= \frac{1}{2}[\exp(i\omega t) + \exp(-i\omega t)]\exp[-i\underbrace{(\Omega_{ba} - \Delta)}_{=\omega}t]\\ &= \frac{1}{2}[\exp(i2\omega t) + 1] \to \frac{1}{2}.\end{aligned}$$

By applying this *rotating-wave approximation*, the equations of motion (6) hence take the form

$$\frac{d\rho_{aa}}{dt} = \frac{i}{2}(\rho^{\Omega}_{ba} - \rho^{\Omega}_{ab})\beta(t) - (\rho_{aa} - \rho_0(a))/T_a, \tag{7a}$$

$$\frac{d\rho^{\Omega}_{ab}}{dt} = i\Delta\rho^{\Omega}_{ab} + \frac{i}{2}\beta(t)(\rho_{bb} - \rho_{aa}) - \rho^{\Omega}_{ab}/T_2, \tag{7b}$$

$$\frac{d\rho_{bb}}{dt} = -\frac{i}{2}(\rho^{\Omega}_{ba} - \rho^{\Omega}_{ab})\beta(t) - (\rho_{bb} - \rho_0(b))/T_b. \tag{7c}$$

In this final form, before entering the Bloch vector description of the interaction, these equations correspond to Butcher and Cotter's Eqs. (6.41).

### The Bloch equations

Assuming the two states $|a\rangle$ and $|b\rangle$ to be sufficiently similar in order to approximate $T_a \approx T_b \approx T_1$, where $T_1$ is the longitudinal relaxation time, and by taking new variables $(u, v, w)$ according to

$$\begin{aligned}u &= \rho^{\Omega}_{ba} + \rho^{\Omega}_{ab},\\ v &= i(\rho^{\Omega}_{ba} - \rho^{\Omega}_{ab}),\\ w &= \rho_{bb} - \rho_{aa},\end{aligned}$$

the equations of motion (7) are cast in the *Bloch equations*

$$\frac{du}{dt} = -\Delta v - u/T_2, \tag{8a}$$

$$\frac{dv}{dt} = \Delta u + \beta(t)w - v/T_2, \tag{8b}$$

$$\frac{dw}{dt} = -\beta(t)v - (w - w_0)/T_1. \tag{8c}$$

In these equations, the introduced variable $w$ describes the population inversion of the two-level system, while $u$ and $v$ are related to the dispersive and absorptive components of the polarization density of the medium. In the Bloch equations above, $w_0 = \rho_0(b) - \rho_0(a)$ is the thermal equilibrium inversion of the system with no optical field applied.

**The resulting electric polarization density of the medium**

The so far developed theory of the density matrix under resonant interaction can now be applied to the calculation of the electric polarization density of the medium, consisting of $N$ identical molecules per unit volume, as

$$\begin{aligned}
P_\mu(\mathbf{r},t) &= N\langle e\hat{r}_\mu\rangle \\
&= N\,\mathrm{Tr}[\hat{\rho}e\hat{r}_\mu] \\
&= N\sum_{k=a,b}\langle k|\hat{\rho}e\hat{r}_\mu|k\rangle \\
&= N\sum_{k=a,b}\sum_{j=a,b}\langle k|\hat{\rho}|j\rangle\langle j|e\hat{r}_\mu|k\rangle \\
&= N\sum_{k=a,b}\{\langle k|\hat{\rho}|a\rangle\langle a|e\hat{r}_\mu|k\rangle + \langle k|\hat{\rho}|b\rangle\langle b|e\hat{r}_\mu|k\rangle\} \\
&= N\{\langle a|\hat{\rho}|a\rangle\langle a|e\hat{r}_\mu|a\rangle + \langle b|\hat{\rho}|a\rangle\langle a|e\hat{r}_\mu|b\rangle + \langle a|\hat{\rho}|b\rangle\langle b|e\hat{r}_\mu|a\rangle + \langle b|\hat{\rho}|b\rangle\langle b|e\hat{r}_\mu|b\rangle\} \\
&= N(\rho_{ba}er^\mu_{ab} + \rho_{ab}er^\mu_{ba}) \\
&= \{\text{Make use of } \rho_{ab} = (u+iv)\exp(i\omega t) = \rho^*_{ba}\} \\
&= N[(u-iv)\exp(-i\omega t)er^\mu_{ab} + (u+iv)\exp(i\omega t)er^\mu_{ba}].
\end{aligned}$$

The temporal envelope $P^\mu_\omega$ of the polarization density, throughout this course as well as in Butcher and Cotter's book, is taken as

$$P^\mu(\mathbf{r},t) = \mathrm{Re}[P^\mu_\omega \exp(-i\omega t)],$$

and by identifying this expression with the right-hand side of the result above, we hence finally have obtained the polarization density in terms of the Bloch parameters $(u, v, w)$ as

$$P^\mu_\omega(\mathbf{r},t) = Ner^\mu_{ab}(u - iv).$$

This expression for the temporal envelope of the polarization density is exactly in the same mode of description as the one as previously used in the susceptibility theory, as in the wave equations developed in lecture eight. The only difference is that now we instead consider the polarization density as given by a non-perturbative analysis. Taken together with the Maxwell's equations (or the proper wave equation for the envelopes of the fields), the Bloch equations are known as the *Maxwell–Bloch equations.*

# Lecture XII

**Lecture Notes on Nonlinear Optics**
Nonlinear Optics (5A5513, 5p for advanced undergraduate and doctoral students)
Course given at the Royal Institute of Technology,
Department of Laser Physics and Quantum Optics
SE–106 91, Stockholm, Sweden
January 8 – March 24, 2003

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## LECTURE 12

In this final lecture, we will study the behaviour of the Bloch equations in different regimes of resonance and relaxation. The Bloch equations are formulated as a vector model, and numerical solutions to the equations are discussed.

For steady-state interaction, the polarization density of the medium, as obtained from the Bloch equations, is expressed in a closed form. The closed solution is then expanded in a power series, which when compared with the series obtained from the susceptibility formalism finally tie together the Bloch theory with the susceptibilities.

The outline for this lecture is:

- Recapitulation of the Bloch equations
- The vector model of the Bloch equations
- Special cases and examples
- Steady-state regime
- The intensity dependent refractive index at steady-state
- Comparison with the susceptibility model

### Recapitulation of the Bloch equations for two-level systems

Assuming two states $|a\rangle$ and $|b\rangle$ to be sufficiently similar in order for their respective lifetimes $T_a \approx T_b \approx T_1$ to hold, where $T_1$ is the *longitudinal relaxation time*, the Bloch equations for the two-level are given as

$$\frac{du}{dt} = -\Delta v - u/T_2, \tag{1a}$$

$$\frac{dv}{dt} = \Delta u + \beta(t)w - v/T_2, \tag{1b}$$

$$\frac{dw}{dt} = -\beta(t)v - (w - w_0)/T_1, \tag{1c}$$

where $\beta \equiv er^{\alpha}_{ab}E^{\alpha}_{\omega}(t)/\hbar$ is the Rabi frequency, being a quantity linear in the applied electric field of the light, $\Delta \equiv \Omega_{ba} - \omega$ is the detuning of the angular frequency of the light from the transition frequency $\Omega_{ba} \equiv (\mathbb{E}_b - \mathbb{E}_a)/\hbar$, and where the variables $(u, v, w)$ are related to the matrix elements $\rho_{mn}$ of the density operator as

$$\begin{aligned} u &= \rho^{\Omega}_{ba} + \rho^{\Omega}_{ab}, \\ v &= i(\rho^{\Omega}_{ba} - \rho^{\Omega}_{ab}), \\ w &= \rho_{bb} - \rho_{aa}. \end{aligned}$$

In these equations, $\rho^{\Omega}_{ab}$ is the *temporal envelope of the off-diagonal elements*, given by

$$\rho_{ab} \equiv \rho^{\Omega}_{ab} \exp[i(\Omega_{ba} - \Delta)t].$$

In the Bloch equations (1), the variable $w$ describes the population inversion of the two-level system, while $u$ and $v$ are related to the dispersive and absorptive components of the polarization density of the medium. In the Bloch equations, $w_0 \equiv \rho_0(b) - \rho_0(a)$ is the thermal equilibrium inversion of the system with no optical field applied.

**The resulting electric polarization density of the medium**

The so far developed theory of the density matrix under resonant interaction can now be applied to the calculation of the electric polarization density of the medium, consisting of $N$ identical molecules per unit volume, as

$$\begin{aligned}
P_\mu(\mathbf{r},t) &= N\langle e\hat{r}_\mu\rangle \\
&= N\,\mathrm{Tr}[\hat{\rho}e\hat{r}_\mu] \\
&= N\sum_{k=a,b}\langle k|\hat{\rho}e\hat{r}_\mu|k\rangle \\
&= N\sum_{k=a,b}\sum_{j=a,b}\langle k|\hat{\rho}|j\rangle\langle j|e\hat{r}_\mu|k\rangle \\
&= N\sum_{k=a,b}\{\langle k|\hat{\rho}|a\rangle\langle a|e\hat{r}_\mu|k\rangle + \langle k|\hat{\rho}|b\rangle\langle b|e\hat{r}_\mu|k\rangle\} \\
&= N\{\langle a|\hat{\rho}|a\rangle\langle a|e\hat{r}_\mu|a\rangle + \langle b|\hat{\rho}|a\rangle\langle a|e\hat{r}_\mu|b\rangle + \langle a|\hat{\rho}|b\rangle\langle b|e\hat{r}_\mu|a\rangle + \langle b|\hat{\rho}|b\rangle\langle b|e\hat{r}_\mu|b\rangle\} \\
&= N(\rho_{ba}er^\mu_{ab} + \rho_{ab}er^\mu_{ba}) \\
&= \{\text{Make use of } \rho_{ab} = (u+iv)\exp(i\omega t) = \rho^*_{ba}\} \\
&= N[(u-iv)\exp(-i\omega t)er^\mu_{ab} + (u+iv)\exp(i\omega t)er^\mu_{ba}].
\end{aligned}$$

The temporal envelope $P^\mu_\omega$ of the polarization density is throughout this course as well as in Butcher and Cotter's book taken as

$$P^\mu(\mathbf{r},t) = \mathrm{Re}[P^\mu_\omega \exp(-i\omega t)],$$

and by identifying this expression with the right-hand side of the result above, we hence finally have obtained the polarization density in terms of the Bloch parameters $(u,v,w)$ as

$$P^\mu_\omega(\mathbf{r},t) = Ner^\mu_{ab}(u-iv). \tag{2}$$

This expression for the temporal envelope of the polarization density is exactly in the same mode of description as the one as previously used in the susceptibility theory, as in the wave equations developed in lecture eight. The only difference is that now we instead consider the polarization density as given by a non-perturbative analysis. Taken together with the Maxwell's equations (or the propern wave equation for the envelopes of the fields), the Bloch equations are known as the *Maxwell–Bloch equations*.

From Eq. (2), it should now be clear that the Bloch variable $u$ essentially gives the in-phase part of the polarization density (at least in this case, where we may consider the transition dipole moments to be real-valued), corresponding to the dispersive components of the interaction between light and matter, while the Bloch variable $v$ on the other hand gives terms which are shifted ninety degrees out of phase with the optical field, hence corresponding to absorptive terms.

**The vector model of the Bloch equations**

In the form of Eqs. (1), the Bloch equations can be expressed in the form of an Euler equation as

$$\frac{d\mathbf{R}}{dt} = \mathbf{\Omega} \times \mathbf{R} - \underbrace{(u/T_2, v/T_2, (w - w_0)/T_1)}_{\text{relaxation term}}, \qquad \text{[B. \& C. (6.54)]}$$

where $\mathbf{R} = (u, v, w)$ is the so-called *Bloch vector*, that in the abstract $(\mathbf{e}_u, \mathbf{e}_v, \mathbf{e}_w)$-space describes the state of the medium, and

$$\mathbf{\Omega} = (-\beta(t), 0, \Delta)$$

is the vector that gives the precession of the Bloch vector (see Fig. 1).

This form, originally proposed in 1946 by Felix Bloch[1] for the motion of a nuclear spin in a magnetic field under influence of radio-frequency electromagnetic fields, and later on adopted by Feynman, Vernon, and Hellwarth[2] for solving problems in maser theory[3], corresponds to the motion of a damped gyroscope in the presence of a gravitational field. In this analogy, the vector $\mathbf{\Omega}$ can be considered as the torque vector of the spinning top of the gyroscope.

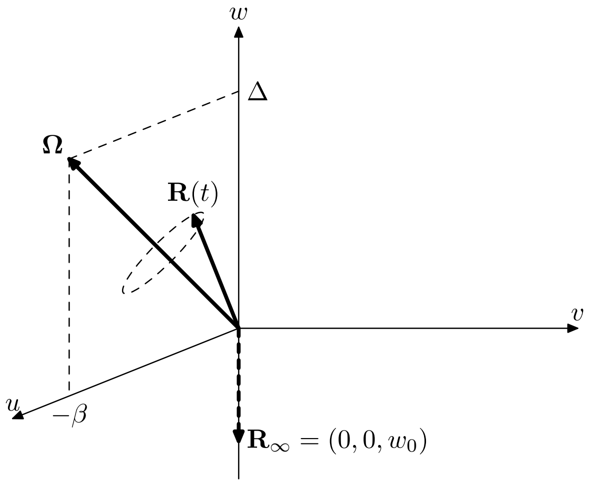

Figure 1. Evolution of the Bloch vector $\mathbf{R}(t) = (u(t), v(t), w(t))$ around the "torque vector" $\mathbf{\Omega} = (-\beta(t), 0, \Delta)$. In the absence of optical fields, the Bloch vector relax towards the thermal equilibrium state $\mathbf{R}_\infty = (0, 0, w_0)$, where $w_0 = \rho(b) - \rho(a)$ is the molecular population inversion at thermal equilibrium. At moderate temperatures, the thermal equilibrium population inversion is very close to $w_0 = -1$.

From the vector form of the Bloch equations, it is found that the Bloch vector rotates around the torque vector $\mathbf{\Omega}$ as the state of matter approaches steady state. For an adiabatically changing applied optical field (i. e. a slowly varying envelope of the field), this precession follows the torque vector.

The relaxation term in the vector Bloch equations also tells us that the relaxation along the $w$-direction is given by the time constant $T_1$, while the relaxation in the $(u, v)$-plane instead is given by the time constant $T_2$. By considering the $w$-axis as the "longitudinal" direction and the $(u, v)$-plane as the "transverse" plane, the terminology for $T_1$ as being the "longitudinal relaxation time" and $T_2$ as being the "transverse relaxation time" should hence be clear.

---

[1] F. Bloch, *Nuclear induction*, Phys. Rev. **70**, 460 (1946). Felix Bloch was in 1952 awarded the Nobel prize in physics, together with Edward Mills Purcell, "for their development of new methods for nuclear magnetic precision measurements and discoveries in connection therewith".

[2] R. P. Feynman, F. L. Vernon, and R. W. Hellwarth, *Geometrical representation of the Schrödinger equation for solving maser problems*, J. Appl. Phys. **28**, 49 (1957).

[3] Microwave Amplification by Stimulated Emission of Radiation, a device for amplification of microwaves, essentially working on the same principle as the laser.

## Transient build-up at exact resonance as the optical field is switched on

*The case $T_1 \gg T_2$ – Longitudinal relaxation slower than transverse relaxation*

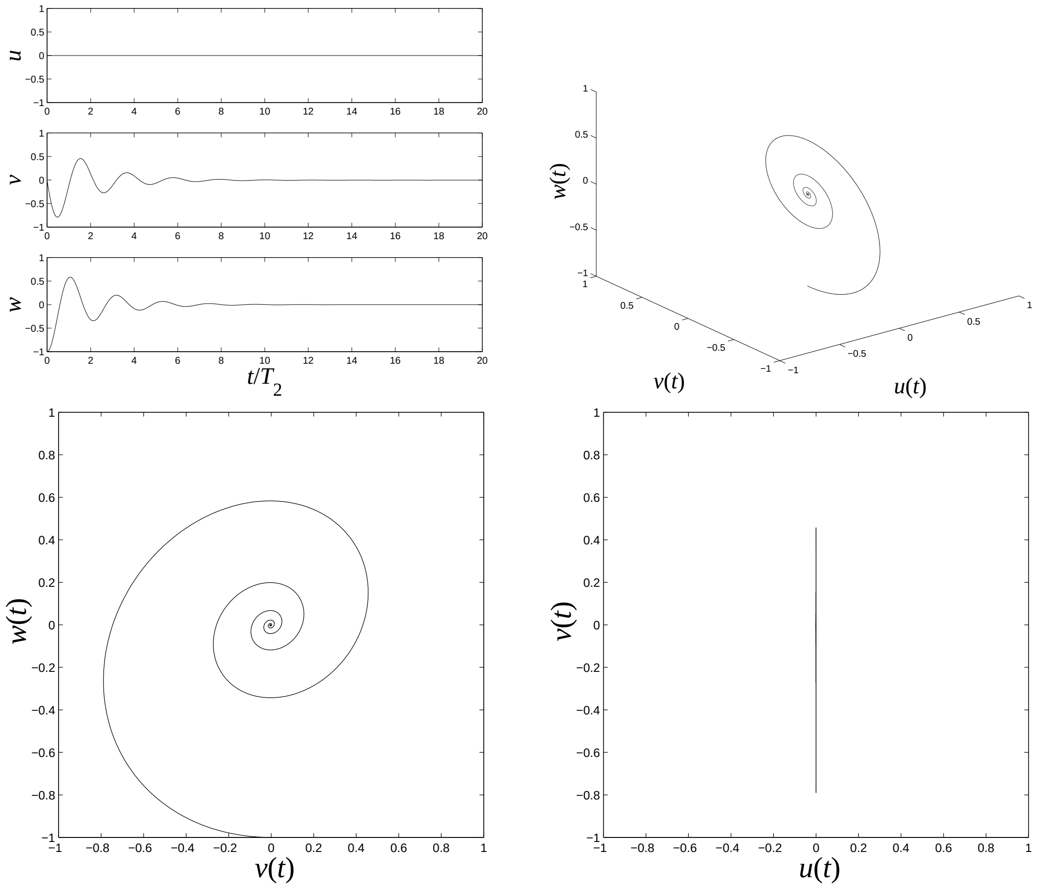

Figure 2a. Evolution of the Bloch vector $(u(t), v(t), w(t))$ as the optical field is switched on, for the exactly resonant case ($\delta = 0$), and with the longitudinal relaxation time being much greater than the transverse relaxation time ($T_1 \gg T_2$). The parameters used in the simulation are $\eta \equiv T_1/T_2 = 100$, $\delta \equiv \Delta T_2 = 0$, $w_0 = -1$, and $\gamma(t) \equiv \beta(t)T_2 = 3$, $t > 0$. The medium was initially at thermal equilibrium, $(u(0), v(0), w(0)) = (0, 0, w_0) = -(0, 0, 1)$.

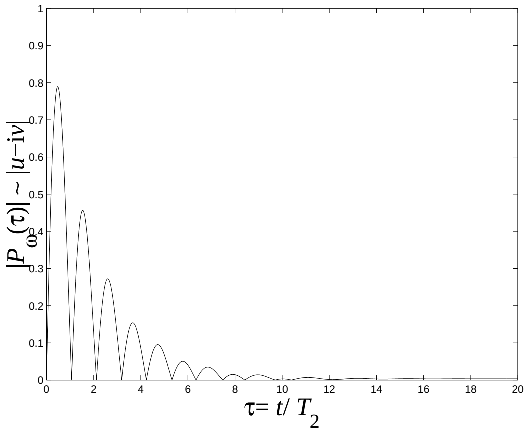

Figure 2b. Evolution of the magnitude of the polarization density $|P_\omega(t)| \sim |u(t) - iv(t)|$ as the optical field is switched on, corresponding to the simulation shown in Fig. 2a.

*The case* $T_1 \approx T_2$ *– Longitudinal relaxation approximately equal to transverse relaxation*

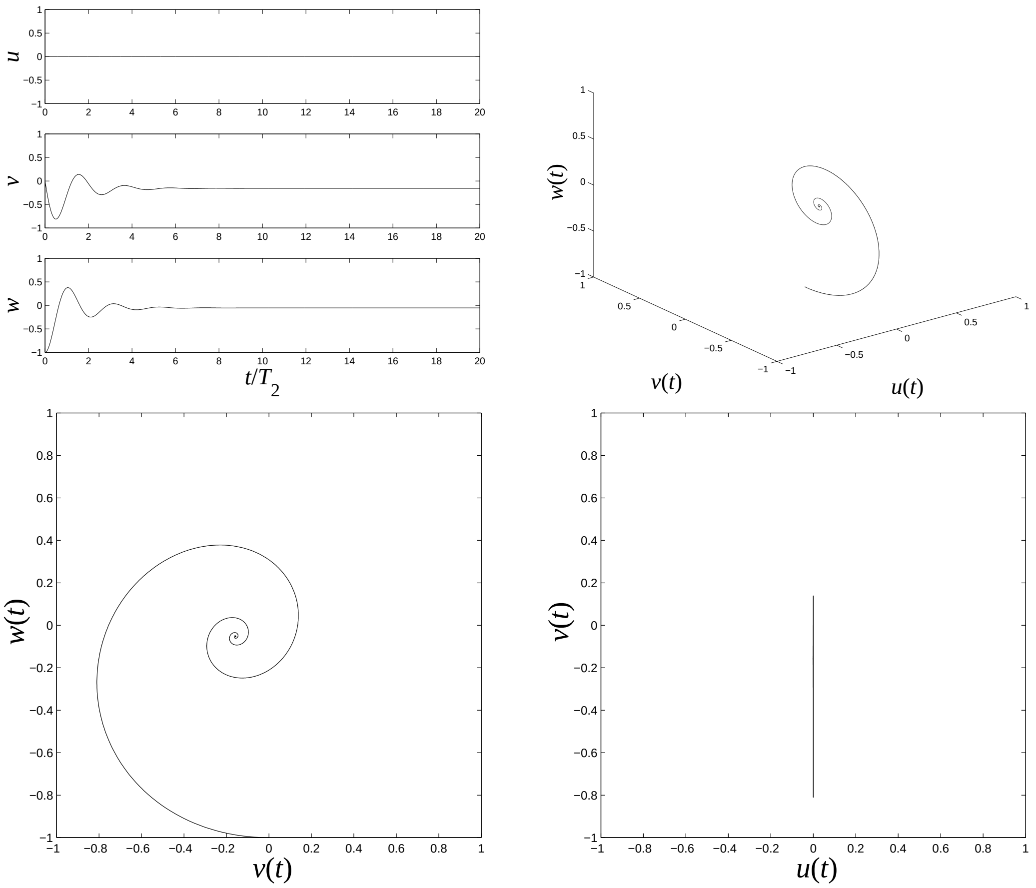

Figure 3a. Evolution of the Bloch vector $(u(t), v(t), w(t))$ as the optical field is switched on, for the exactly resonant case ($\delta = 0$), and with the longitudinal relaxation time being approximately equal to the transverse relaxation time ($T_1 \approx T_2$). The parameters used in the simulation are $\eta \equiv T_1/T_2 = 2$, $\delta \equiv \Delta T_2 = 0$, $w_0 = -1$, and $\gamma(t) \equiv \beta(t)T_2 = 3$, $t > 0$. The medium was initially at thermal equilibrium, $(u(0), v(0), w(0)) = (0, 0, w_0) = -(0, 0, 1)$.

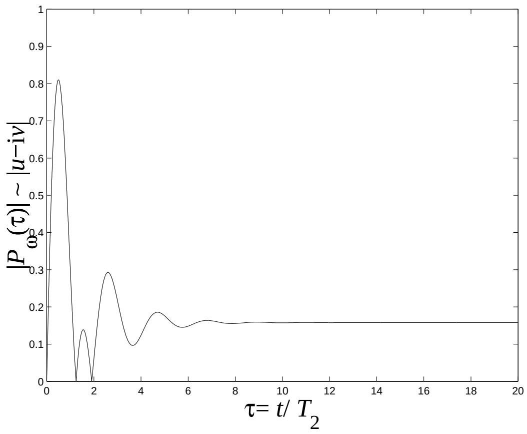

Figure 3b. Evolution of the magnitude of the polarization density $|P_\omega(t)| \sim |u(t) - iv(t)|$ as the optical field is switched on, corresponding to the simulation shown in Fig. 3a.

**Transient build-up at off-resonance as the optical field is switched on**

*The case $T_1 \approx T_2$ – Longitudinal relaxation approximately equal to transverse relaxation*

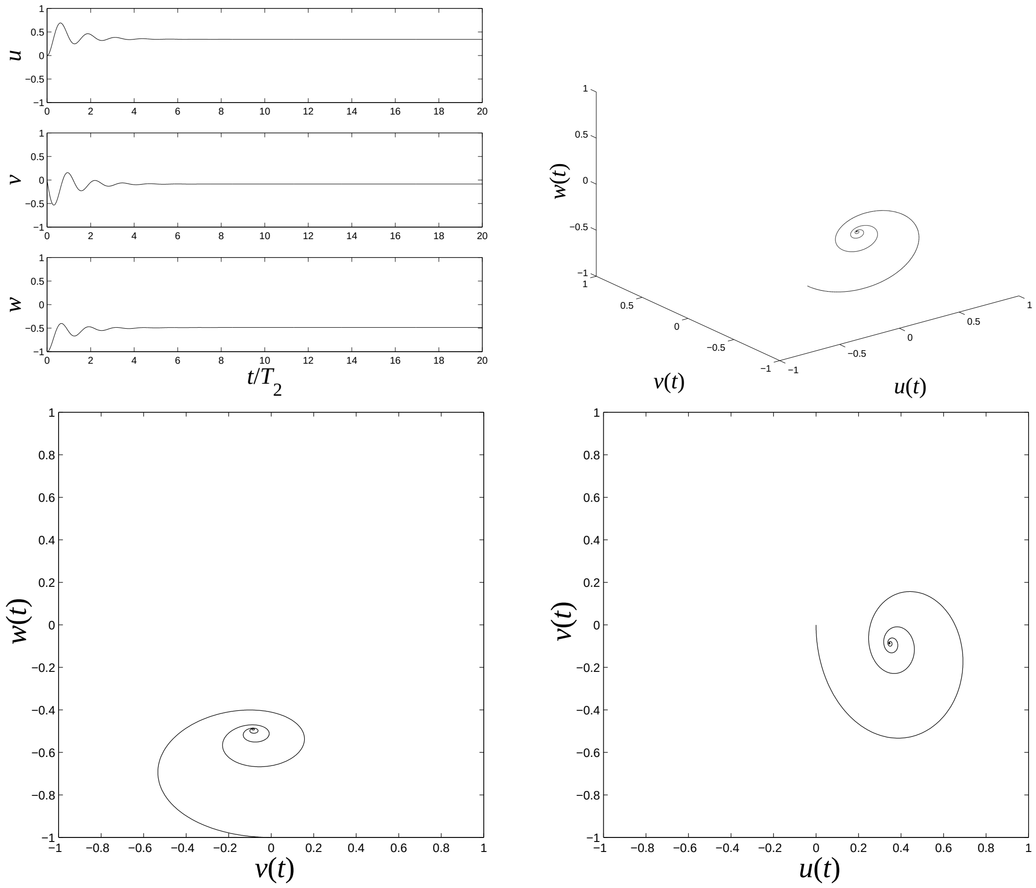

Figure 4a. Evolution of the Bloch vector $(u(t), v(t), w(t))$ as the optical field is switched on, for the off-resonant case ($\delta \neq 0$), and with the longitudinal relaxation time being approximately equal to the transverse relaxation time ($T_1 \approx T_2$). The parameters used in the simulation are $\eta \equiv T_1/T_2 = 2$, $\delta \equiv \Delta T_2 = 4$, $w_0 = -1$, and $\gamma(t) \equiv \beta(t)T_2 = 3$, $t > 0$. The medium was initially at thermal equilibrium, $(u(0), v(0), w(0)) = (0, 0, w_0) = -(0, 0, 1)$.

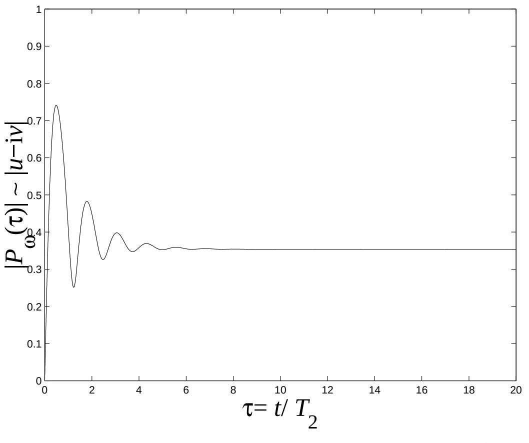

Figure 4b. Evolution of the magnitude of the polarization density $|P_\omega(t)| \sim |u(t) - iv(t)|$ as the optical field is switched on, corresponding to the simulation shown in Fig. 4a.

### Transient decay for a process tuned to exact resonance

*The case $T_1 \gg T_2$ – Longitudinal relaxation slower than transverse relaxation*

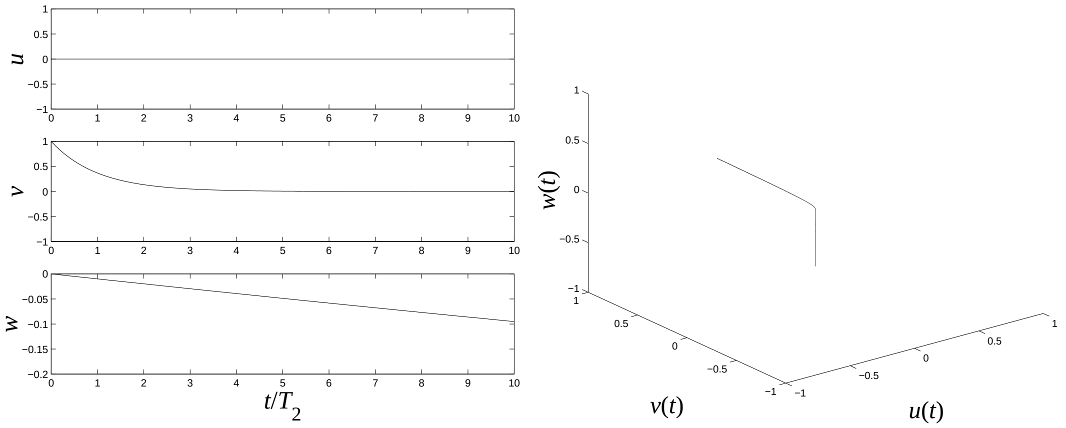

Figure 5. Evolution of the Bloch vector $(u(t), v(t), w(t))$ after the optical field is switched off, for the case of tuning to exact resonance ($\delta = 0$), and with the longitudinal relaxation time being much greater than the transverse relaxation time ($T_1 \gg T_2$). The parameters used in the simulation are $\eta \equiv T_1/T_2 = 100$, $\delta \equiv \Delta T_2 = 0$, $w_0 = -1$, and $\gamma(t) \equiv \beta(t)T_2 = 0$.

*The case $T_1 \approx T_2$ – Longitudinal relaxation approximately equal to transverse relaxation*

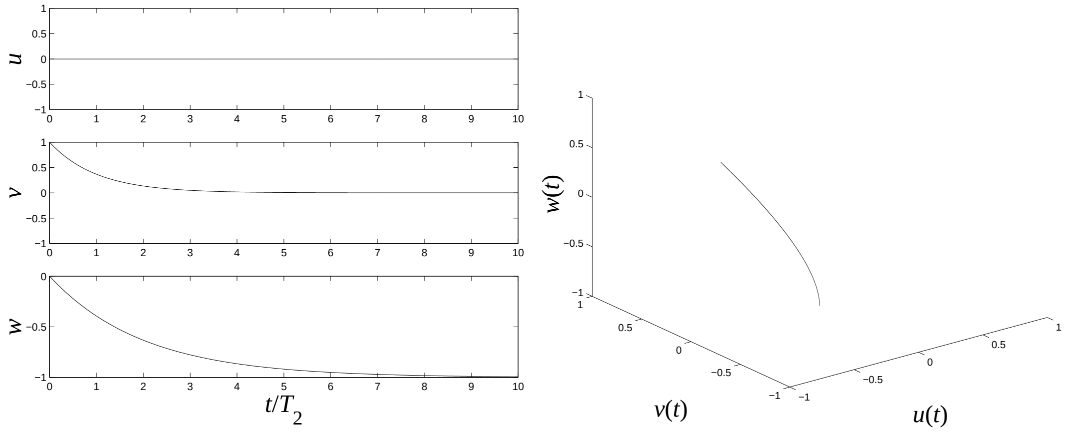

Figure 6. Evolution of the Bloch vector $(u(t), v(t), w(t))$ after the optical field is switched off, for the case of tuning to exact resonance ($\delta = 0$), and with the longitudinal relaxation time being approximately equal to the transverse relaxation time ($T_1 \approx T_2$). The parameters used in the simulation are $\eta \equiv T_1/T_2 = 2$, $\delta \equiv \Delta T_2 = 0$, $w_0 = -1$, and $\gamma(t) \equiv \beta(t)T_2 = 0$.

### Transient decay for a slightly off-resonant process

*The case $T_1 \gg T_2$ – Longitudinal relaxation slower than transverse relaxation*

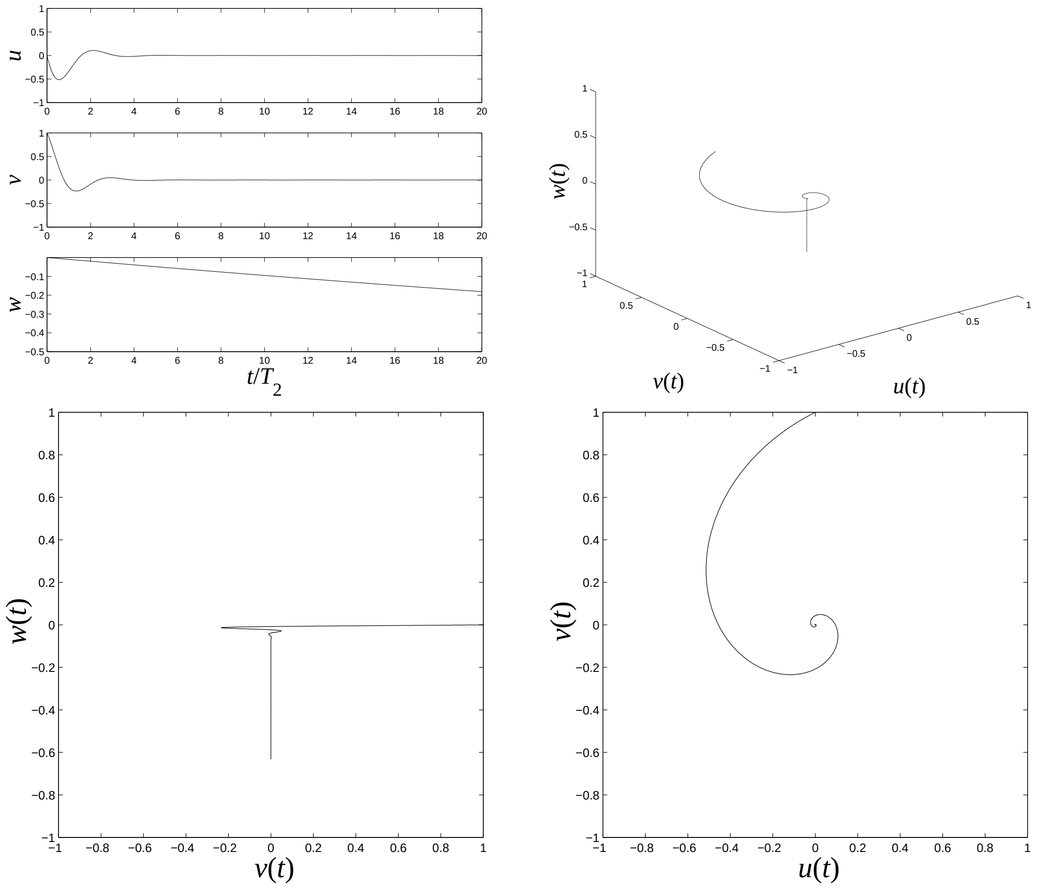

Figure 7a. Evolution of the Bloch vector $(u(t), v(t), w(t))$ after the optical field is switched off, for the off-resonant case ($\delta \neq 0$), and with the longitudinal relaxation time being much greater than the transverse relaxation time ($T_1 \gg T_2$). The parameters used in the simulation are $\eta \equiv T_1/T_2 = 100$, $\delta \equiv \Delta T_2 = 2$, $w_0 = -1$, and $\gamma(t) \equiv \beta(t)T_2 = 0$. (Compare with Fig. 5 for the exactly resonant case.)

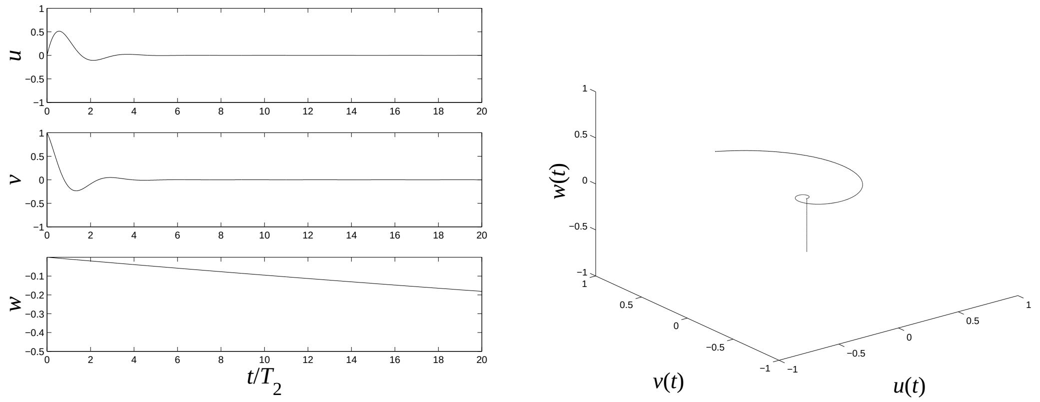

Figure 7b. Same as Fig. 7a, but with $\delta = -2$ as negative.

### *The case $T_1 \approx T_2$ – Longitudinal relaxation approximately equal to transverse relaxation*

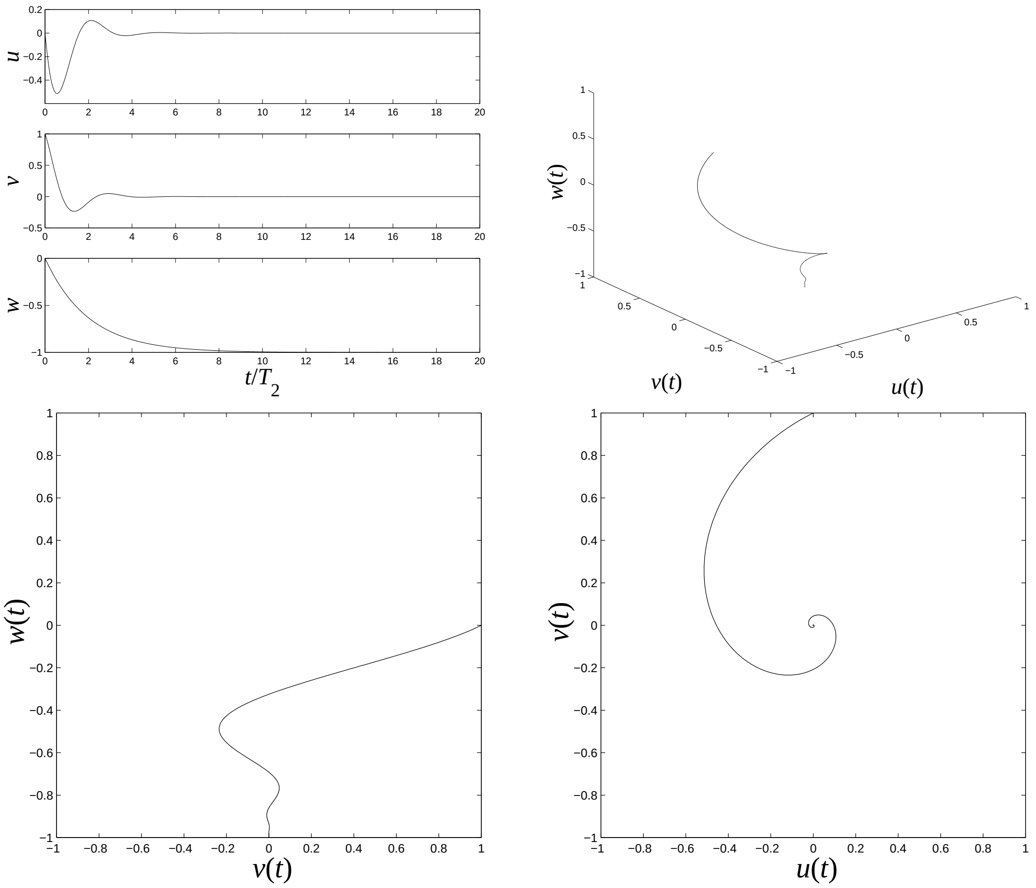

Figure 8a. Evolution of the Bloch vector $(u(t), v(t), w(t))$ after the optical field is switched off, for the off-resonant case ($\delta \neq 0$), and with the longitudinal relaxation time being approximately equal to the transverse relaxation time ($T_1 \approx T_2$). The parameters used in the simulation are $\eta \equiv T_1/T_2 = 2$, $\delta \equiv \Delta T_2 = 2$, $w_0 = -1$, and $\gamma(t) \equiv \beta(t)T_2 = 0$. (Compare with Fig. 6 for the exactly resonant case.)

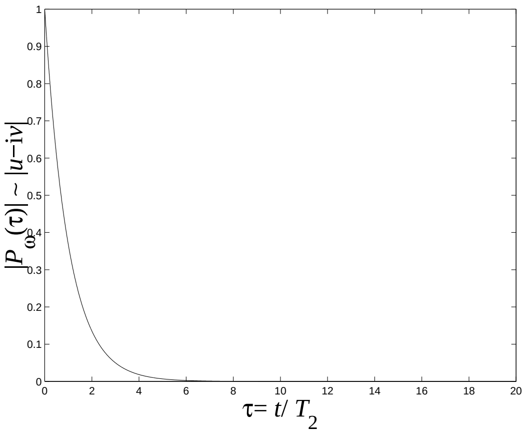

Figure 8b. Evolution of the magnitude of the polarization density $|P_\omega(t)| \sim |u(t) - iv(t)|$ as the optical field is switched on, corresponding to the simulation shown in Fig. 8a.

### Transient decay for a far off-resonant process

*The case $T_1 \gg T_2$ – Longitudinal relaxation slower than transverse relaxation*

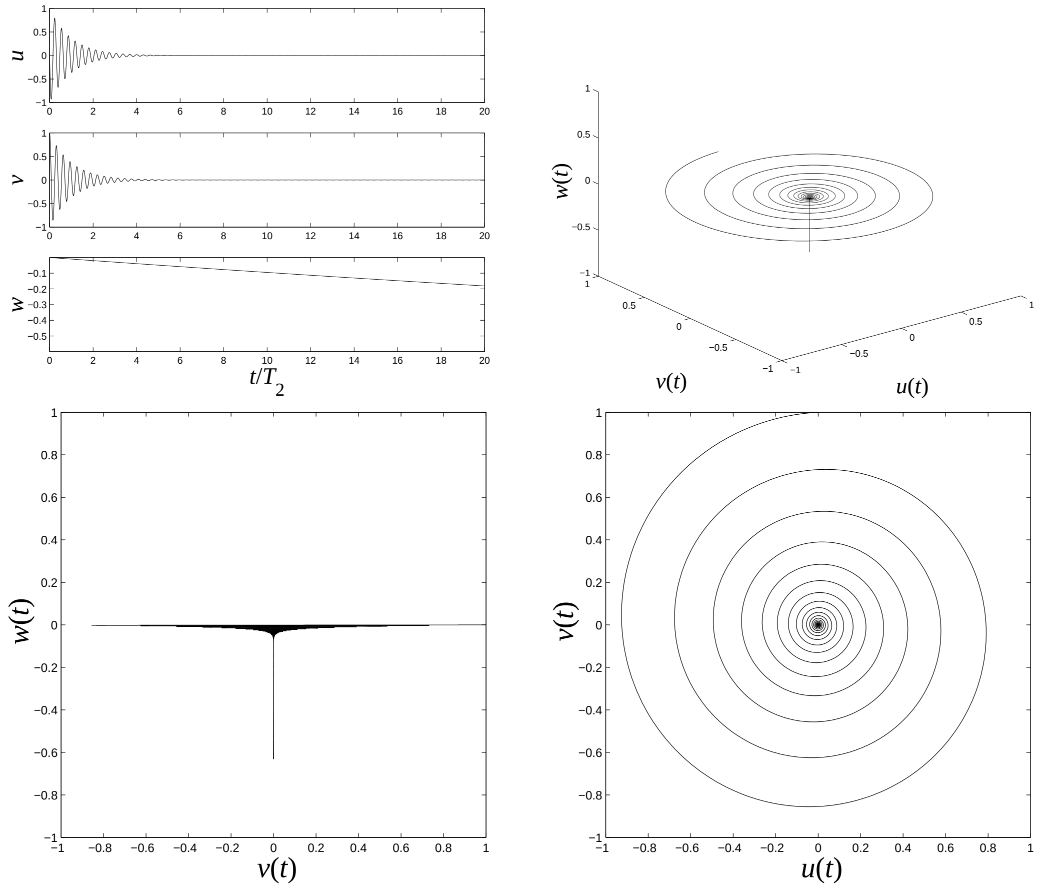

Figure 9a. Evolution of the Bloch vector $(u(t), v(t), w(t))$ after the optical field is switched off, for the far off-resonant case ($\delta \neq 0$), and with the longitudinal relaxation time being much greater than the transverse relaxation time ($T_1 \gg T_2$). The parameters used in the simulation are $\eta \equiv T_1/T_2 = 100$, $\delta \equiv \Delta T_2 = 20$, $w_0 = -1$, and $\gamma(t) \equiv \beta(t)T_2 = 0$. (Compare with Fig. 5 for the exactly resonant case, and with Fig. 7a for the slightly off-resonant case.)

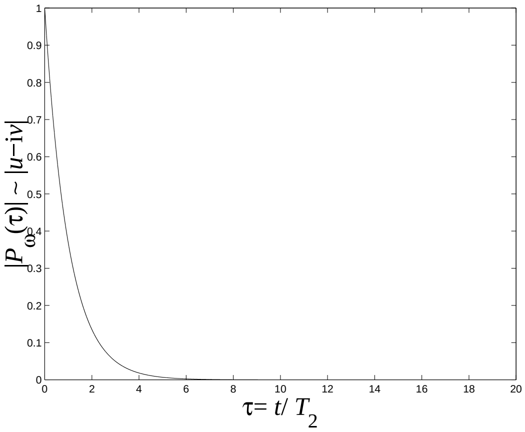

Figure 9b. Evolution of the magnitude of the polarization density $|P_\omega(t)| \sim |u(t) - iv(t)|$ as the optical field is switched on, corresponding to the simulation shown in Fig. 9a.

*The case $T_1 \approx T_2$ – Longitudinal relaxation approximately equal to transverse relaxation*

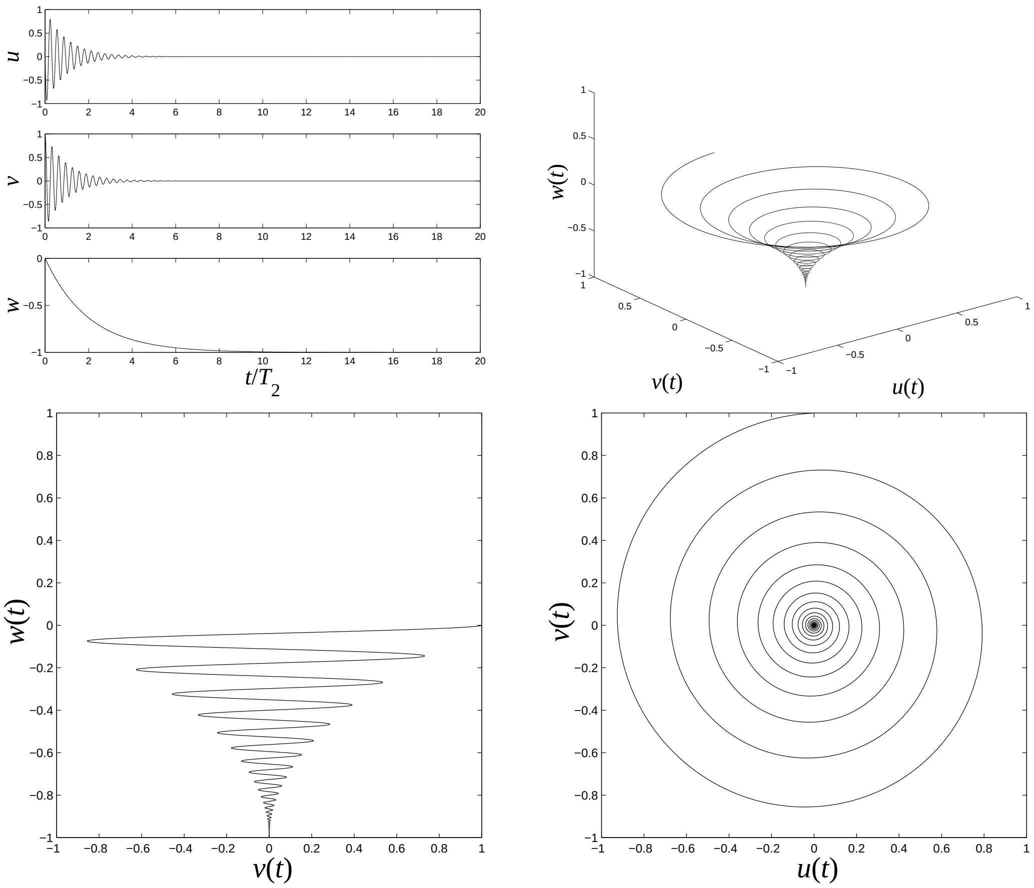

Figure 10a. Evolution of the Bloch vector $(u(t), v(t), w(t))$ after the optical field is switched off, for the far off-resonant case ($\delta \neq 0$), and with the longitudinal relaxation time being approximately equal to the transverse relaxation time ($T_1 \approx T_2$). The parameters used in the simulation are $\eta \equiv T_1/T_2 = 2$, $\delta \equiv \Delta T_2 = 20$, $w_0 = -1$, and $\gamma(t) \equiv \beta(t)T_2 = 0$. (Compare with Fig. 6 for the exactly resonant case, and with Fig. 8a for the slightly off-resonant case.)

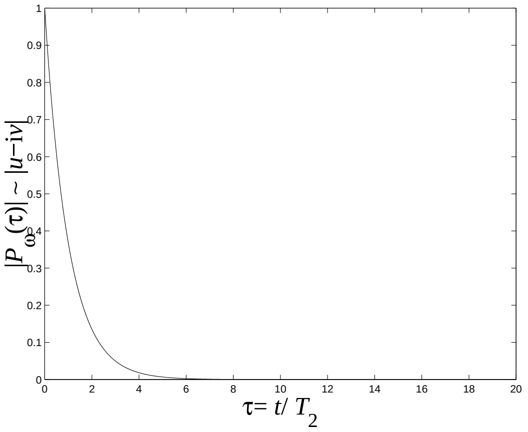

Figure 10b. Evolution of the magnitude of the polarization density $|P_\omega(t)| \sim |u(t) - iv(t)|$ as the optical field is switched on, corresponding to the simulation shown in Fig. 10a.

*The case $T_1 \ll T_2$ – Longitudinal relaxation faster than transverse relaxation*

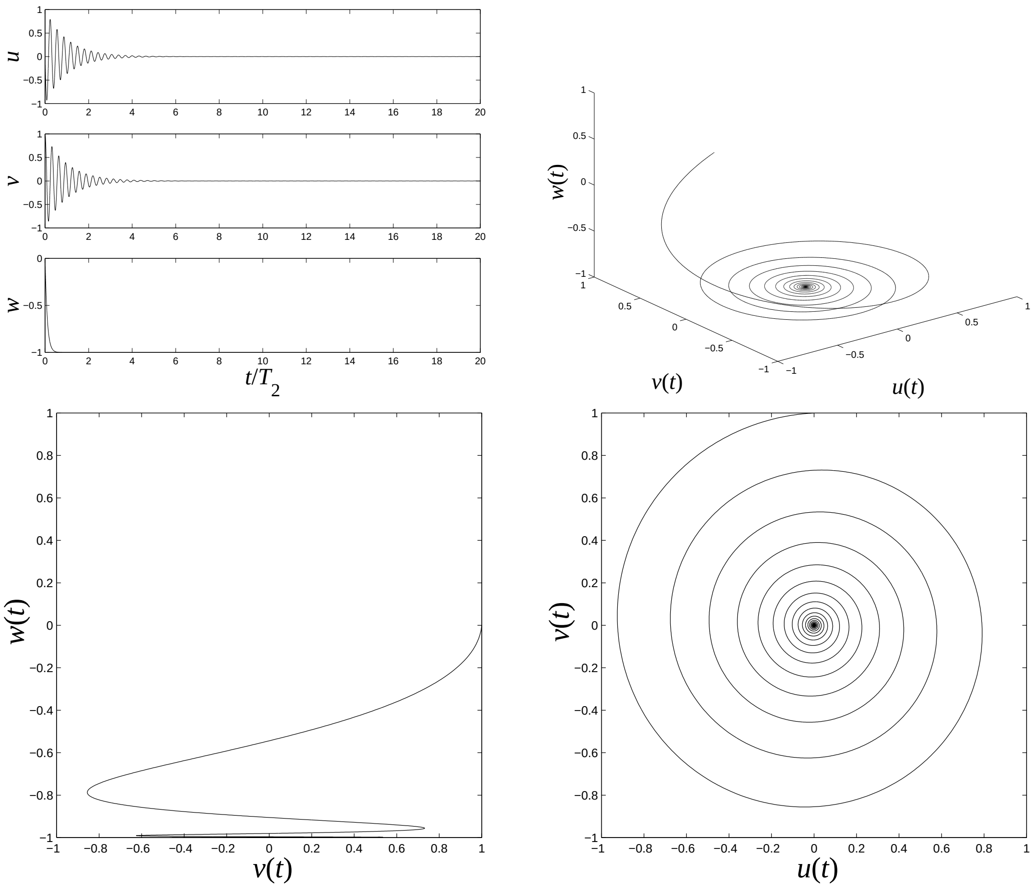

Figure 11a. Same parameter values as in Fig. 6, but with the longitudinal relaxation time being much smaller than the transverse relaxation time ($T_1 \ll T_2$), $\eta \equiv T_1/T_2 = 0.1$. (Compare with Figs. 9a and 10a for the cases $T_1 \gg T_2$ and $T_1 \approx T_2$, respectively.)

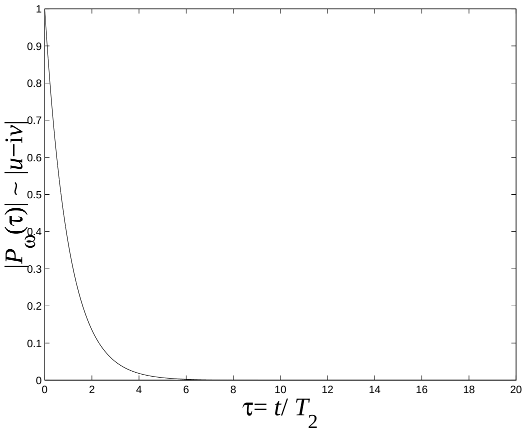

Figure 11b. Evolution of the magnitude of the polarization density $|P_\omega(t)| \sim |u(t) - iv(t)|$ as the optical field is switched on, corresponding to the simulation shown in Fig. 11a.

### The connection between the Bloch equations and the susceptibility

As an example of the connection between the polarization density obtained from the Bloch equations and the one obtained from the susceptibility formalism, we will now – once again – consider the intensity-dependent refractive of the medium.

*The intensity-dependent refractive index in the susceptibility formalism*

Previously in this course, the intensity-dependent refractive index has been obtained from the optical Kerr-effect in isotropic media, in the form

$$n = n_0 + n_2|\mathbf{E}_\omega|^2,$$

where $n_0 = [1 + \chi^{(1)}_{xx}(-\omega;\omega)]^{1/2}$ is the linear refractive index, and

$$n_2 = \frac{3}{8n_0}\chi^{(3)}_{xxxx}(-\omega;\omega,\omega,-\omega)$$

is the parameter of the intensity dependent contribution. However, since we by now are fully aware that the polarization density in the description of the susceptibility formalism originally is given as an infinity series expansion, we may expect that the general form of the intensity dependent refractive index rather would be as a power series in the intensity,

$$n = n_0 + n_2|\mathbf{E}_\omega|^2 + n_4|\mathbf{E}_\omega|^4 + n_6|\mathbf{E}_\omega|^6 + \ldots$$

For linearly polarized light, say along the $x$-axis of a Cartesian coordinate system, we know that such a series is readily possible to derive in terms of the susceptibility formalism, with the different order terms of the refractive index expansion given by the elements

$$\begin{aligned}
n_2 &\sim \chi^{(3)}_{xxxx}(-\omega;\omega,\omega,-\omega),\\
n_4 &\sim \chi^{(5)}_{xxxxxx}(-\omega;\omega,\omega,-\omega,\omega,-\omega),\\
n_6 &\sim \chi^{(7)}_{xxxxxxxx}(-\omega;\omega,\omega,-\omega,\omega,-\omega,\omega,-\omega),\\
&\vdots
\end{aligned}$$

Such an analysis would, however, be extremely cumbersome when it comes to the analysis of higher-order effects, and the obtained sum of various order terms would also be almost impossible to obtain a closed expression for. For future reference, to be used in the interpretation of the polarization density given by the Bloch equations, the intensity dependent polarization density is though shown in its explicit form below, including up to the seventh order interaction term in the Butcher and Cotter convention,

$$\begin{aligned}
P^x_\omega = {}& \varepsilon_0\chi^{(1)}_{xx}(-\omega;\omega)E^x_\omega && \text{(order } n=1)\\
&+ \varepsilon_0(3/4)\chi^{(3)}_{xxxx}(-\omega;\omega,\omega,-\omega)|E^x_\omega|^2E^x_\omega && \text{(order } n=3)\\
&+ \varepsilon_0(5/8)\chi^{(5)}_{xxxxxx}(-\omega;\omega,\omega,-\omega,\omega,-\omega)|E^x_\omega|^4E^x_\omega && \text{(order } n=5)\\
&+ \varepsilon_0(35/64)\chi^{(7)}_{xxxxxxxx}(-\omega;\omega,\omega,-\omega,\omega,-\omega,\omega,-\omega)|E^x_\omega|^6E^x_\omega && \text{(order } n=7)\\
&+ \ldots
\end{aligned}$$

The other approach to calculation of the polarization density, as we next will outline, is to use the steady-state solutions to the Bloch equations.

*The intensity-dependent refractive index in the Bloch-vector formalism*

For steady-state interaction between light and matter, the solutions to the Bloch equations yield

$$u - iv = \frac{-\beta w}{\Delta - i/T_2}, \qquad \text{[B.\,\&\,C. (6.53a)]},$$

$$w = \frac{w_0[1 + (\Delta T_2)^2]}{1 + (\Delta T_2)^2 + \beta^2 T_1 T_2}, \qquad \text{[B.\,\&\,C. (6.53b)]},$$

where, as previously, $\beta = er^{\alpha}_{ab}E^{\alpha}_{\omega}(t)/\hbar$ is the Rabi frequency, though now considered to be a slowly varying (adiabatically following) quantity, due to the assumption of steady-state behaviour. From the steady-state solutions, the $\mu$-component ($\mu = x, y, z$) of the electric polarization density $\mathbf{P}(\mathbf{r}, t) = \mathrm{Re}[\mathbf{P}_{\omega} \exp(-i\omega t)]$ of the medium hence is given as

$$\begin{aligned} P^{\mu}_{\omega} &= Ner^{\mu}_{ab}(u - iv) \\ &= -Ner^{\mu}_{ab}\frac{\beta w}{\Delta - i/T_2} \\ &= -Ner^{\mu}_{ab}\frac{\beta}{(\Delta - i/T_2)}\frac{w_0[1 + (\Delta T_2)^2]}{[1 + (\Delta T_2)^2 + \beta^2 T_1 T_2]} \\ &= -New_0\frac{r^{\mu}_{ab}}{(\Delta - i/T_2)}\frac{\beta}{\left[1 + \frac{T_1T_2}{(1+(\Delta T_2)^2)}\beta^2\right]}. \end{aligned} \tag{3}$$

In this expression for the polarization density, it might at a first glance seem as it is negative for a positive Rabi frequency $\beta$, henc giving a polarization density that is directed anti-parallel to the electric field. However, the quantity $w_0 = \rho_0(b) - \rho_0(a)$, the population inversion at thermal equilibrium, is always negative (since we for sure do not have any population inversion at thermal equilibrium, for which we rather expect the molecules to occupy the lower state), hence ensuring that the off-resonant, real-valued polarization density always is directed along the direction of the electric field of the light.

Next observation is that the polarization density no longer is expressed as a power series in terms of the electric field, but rather as a rational function,

$$P^{\mu}_{\omega} \sim X/(1 + X^2), \tag{4}$$

where

$$\begin{aligned} X &= \sqrt{T_1T_2/(1 + (\Delta T_2)^2)}\beta \\ &= \sqrt{T_1T_2/(1 + (\Delta T_2)^2)}er^{\alpha}_{ab}E^{\alpha}_{\omega}(t)/\hbar \end{aligned}$$

is a parameter linear in the electric field. The principal shape of the rational function in Eq. (4) is shown in Fig. 12.

From Eq. (4), the polarization density is found to increase with increasing $X$ up to $X = 1$, as we expect for an increasing power of an optical beam. However, for $X > 1$, we find the somewhat surprising fact that the polarization density instead *decrease* with an increasing intensity; this peculiar suggested behaviour should hence be explained before continuing.

The first observation we may do is that the linear polarizability (i. e. what we usually associate with linear optics) follows the first order approximation $p(X) = X$. In the region where the peculiar decrease of the polarization density appear, the difference between the suggested nonlinear polarization density and the one given by the linear approximation is *huge*, and since we *a priori* expect nonlinear contributions to be small compared to the alsways present linear ones, this is already an indication of that we in all practical situations do not have to consider the descrease of polarization density as shown in Fig. 12.

For optical fields of the strength that would give rise to nonlinearities exceeding the linear terms, the underlying physics will rather belong to the field of plasma and high-energy physics, rather than a bound-charge description of gases and solids. This implies that the validity of the models here applied (bound charges, Hamiltonians being linear in the optical field, etc.) are limited to a range well within $X \leq 1$.

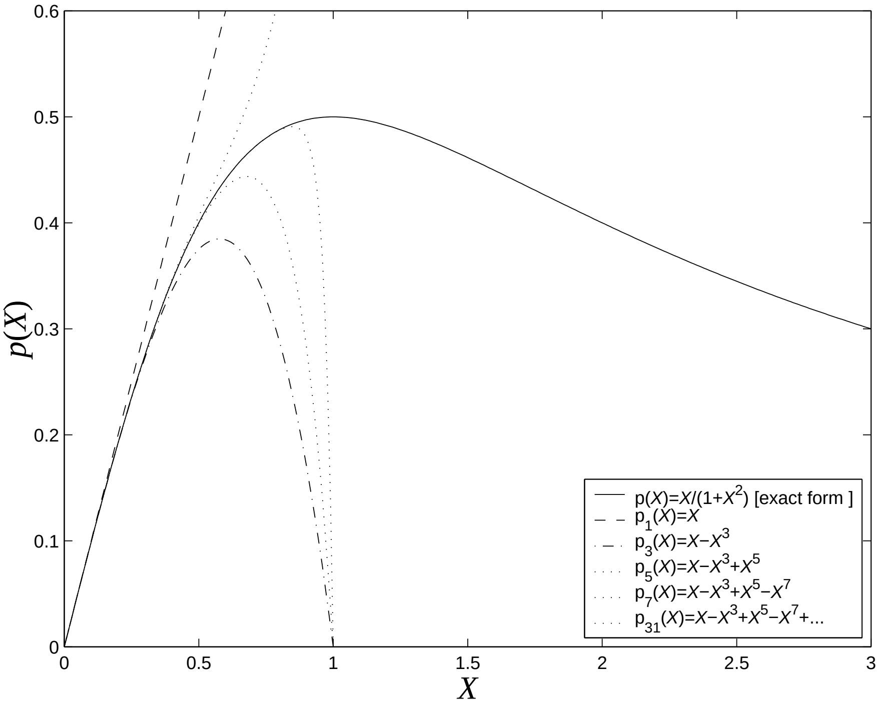

Figure 12. The principal shape of the electric polarization density of the medium, as function of the applied electric field of the light. In this figure, $X = \sqrt{T_1T_2/(1+(\Delta T_2)^2)}\beta$ is a normalized parameter describing the field strength of the electric field of the light.

Another interesting point we may observe is a more mathematically related one. In Fig. 12, we see that even for very high order terms (such as the approximating power series of degree 31, as shown in the figure), all power series expansions fail before reaching $X = 1$. The reason for this is that the power series that approximate the rational function $X/(1+X^2)$,

$$X/(1+X^2) = X - X^3 + X^5 - X^7 + \ldots,$$

is convergent only for $|X| < 1$; for all other values, the series is divergent. This means that no matter how many terms we include in the power series in $X$, it will nevertheless fail when it comes to the evaluation for $|X| > 1$. Since this power series expansion is equivalent to the expansion of the nonlinear polarization density in terms of the electrical field of the light (keeping in mind that $X$ here actually is linear in the electric field and hence strictly can be considered as the field variable), this also is an indication that at this point the whole susceptibility formalism fail to give a proper description at this working point.

This is an excellent illustration of the downturn of the susceptibility description of interaction between light and matter; no matter how many terms we may include in the power series of the electrcal field, *it will at some point nevertheless fail to give the total picture of the interaction*, and we must then instead seek other tools.

Returning to the polarization density given by Eq. (3), we may now express this in an explicit form by inserting $\Delta \equiv \Omega_{ba} - \omega$ for the angular frequency detuning, the Rabi frequency $\beta = er^{\alpha}_{ab}E^{\alpha}_{\omega}(t)/\hbar$, and the thermal equilibrium inversion $w_0 = \rho_0(b) - \rho_0(a)$. This gives the polarization density of the medium as

$$P^{\mu}_{\omega} = \varepsilon_0 \underbrace{\underbrace{\frac{Ne^2}{\varepsilon_0\hbar}(\rho_0(a)-\rho_0(b))\frac{r^{\mu}_{ab}r^{\alpha}_{ab}}{(\Omega_{ba}-\omega-i/T_2)}}_{=\chi^{(1)}_{\mu\alpha}(-\omega;\omega)\text{ for a two level medium}}\;\underbrace{\frac{1}{\left[1+\frac{T_1T_2}{(1+(\Omega_{ba}-\omega)^2T_2^2)}(er^{\gamma}_{ab}E^{\gamma}_{\omega}/\hbar)^2\right]}}_{\text{nonlinear correction factor to }\chi^{(1)}_{\mu\alpha}(-\omega;\omega)}}_{\text{the field corrected susceptibility, }\bar{\chi}(\omega;\mathbf{E}_{\omega})\text{ [see Butcher and Cotter, section 6.3.1]}} E^{\alpha}_{\omega}.$$

In this form, the polarization density is given as the product with a term which is identical to the linear susceptibility[4] (as obtained in the perturbation analysis in the frame of the susceptibility formalism), and a correction factor which is a nonlinear function of the electric field.

The nonlinear correction factor, of the form $1/(1+X^2)$, with $X = \sqrt{T_1 T_2/(1+(\Delta T_2)^2)}\beta$ as previously, can now be expanded in a power series around the small-signal limit $X = 0$, using

$$1/(1+X^2) = 1 - X^2 + X^4 - X^6 + \ldots,$$

from which we obtain the polarization density as a power series in the electric field (which for the sake of simplicitly now is taken as linearly polarized along the $x$-axis) as

$$\begin{aligned} P^x_\omega \approx\ & \varepsilon_0 \frac{Ne^2}{\varepsilon_0 \hbar}(\rho_0(a)-\rho_0(b))\frac{r^x_{ab}r^x_{ab}}{(\Omega_{ba}-\omega-i/T_2)}E^x_\omega \\ & - \varepsilon_0 \frac{Ne^4}{\varepsilon_0 \hbar^3}(\rho_0(a)-\rho_0(b))\frac{r^x_{ab}r^x_{ab}}{(\Omega_{ba}-\omega-i/T_2)}\frac{(r^x_{ab})^2}{[1/T_2^2+(\Omega_{ba}-\omega)^2](T_2/T_1)}|E^x_\omega|^2 E^x_\omega \\ & + \varepsilon_0 \frac{Ne^6}{\varepsilon_0 \hbar^5}(\rho_0(a)-\rho_0(b))\frac{r^x_{ab}r^x_{ab}}{(\Omega_{ba}-\omega-i/T_2)}\frac{(r^x_{ab})^4}{[1/T_2^2+(\Omega_{ba}-\omega)^2]^2(T_2/T_1)^2}|E^x_\omega|^4 E^x_\omega \\ & + \ldots \end{aligned} \tag{5}$$

This form is identical to one as obtained in the susceptibility formalism; however, the steps that led us to this expression for the polarization density *do not rely on the perturbation theory of the density operator*, but rather on the explicit form of the steady-state solutions to the Bloch equations.

**Summary of the Bloch and susceptibility polarization densities**

To summarize this last lecture on the Bloch equations, expressing the involved parameters in the same style as previously used in the description of the susceptibility formalism, the polarization density obtained from the steady-state solutions to the Bloch equations is

$$P^\mu_\omega = \varepsilon_0 \frac{Ne^2}{\varepsilon_0 \hbar}(\rho_0(a)-\rho_0(b))\frac{r^\mu_{ab}r^\alpha_{ab}}{(\Omega_{ba}-\omega-i/T_2)}\frac{1}{\left[1+\frac{T_1T_2}{(1+(\Omega_{ba}-\omega)^2T_2^2)}(er^\alpha_{ab}E^\alpha_\omega/\hbar)^2\right]}E^\alpha_\omega.$$

By expanding this in a power series in the electrical field, one obtains the form (5), in which we from the same description of the polarization density in the susceptibility formalism can identify

$$\begin{aligned} \chi^{(1)}_{xx}(-\omega;\omega) &= \frac{Ne^2}{\varepsilon_0\hbar}(\rho_0(a)-\rho_0(b))\frac{r^x_{ab}r^x_{ab}}{(\Omega_{ba}-\omega-i/T_2)}, \\ \chi^{(3)}_{xxxx}(-\omega;\omega,\omega,-\omega) &= -\frac{4Ne^4}{3\varepsilon_0\hbar^3}(\rho_0(a)-\rho_0(b))\frac{r^x_{ab}r^x_{ab}}{(\Omega_{ba}-\omega-i/T_2)}\frac{(r^x_{ab})^2}{[1/T_2^2+(\Omega_{ba}-\omega)^2](T_2/T_1)}, \\ \chi^{(5)}_{xxxxxx}(-\omega;\omega,\omega,-\omega,&\omega,-\omega) \\ &= \frac{8Ne^6}{5\varepsilon_0\hbar^5}(\rho_0(a)-\rho_0(b))\frac{r^x_{ab}r^x_{ab}}{(\Omega_{ba}-\omega-i/T_2)}\frac{(r^x_{ab})^4}{[1/T_2^2+(\Omega_{ba}-\omega)^2]^2(T_2/T_1)^2}, \end{aligned}$$

as being the first contributions to the two-level polarization density, including up to fifth order interactions. For a summary of the non-resonant forms of the susceptibilities of two-level systems, se Butcher and Cotter, Eqs. (6.71)–(6.73).

---

[4] In the explicit expressions for the linear susceptibility, for example Butcher and Cotter's Eqs. (4.58) and (4.111) for the non-resonant and resonant cases, respectively, there are two terms, one with $\Omega_{ba} - \omega$ in the denominator and the other one with $\Omega_{ba} + \omega$. The reason why the second form does not appear in the expression for the field corrected susceptibility, as derived from the Bloch equations, is that *we have used the rotating wave approximation in the derivation of the final expression.* (Recapitulate that in the rotating wave approximation, terms with oscillatory dependence of $\exp[i(\Omega_{ba}+\omega)t]$ were neglected.) As a result, all temporally phase-mismatched terms are neglected, and in particular only terms with $\Omega_{ba} - \omega$ in the denominator will remain. This, however, is a most acceptable approximation, especially when it comes to resonant interactions, where terms with $\Omega_{ba} - \omega$ in the denominator by far will dominate over non-resonant terms.

**Appendix: Notes on the numerical solution to the Bloch equations**

In their original form, the Bloch equations for a two-level system are given by Eqs. (1) as

$$\begin{aligned}
\frac{du}{dt} &= -\Delta v - u/T_2,\\
\frac{dv}{dt} &= \Delta u + \beta(t)w - v/T_2,\\
\frac{dw}{dt} &= -\beta(t)v - (w - w_0)/T_1.
\end{aligned}$$

By taking the time in units of the transverse relaxation time $T_2$, as

$$\tau = t/T_2,$$

the Bloch equations in this normalized time scale become

$$\begin{aligned}
\frac{du}{d\tau} &= -\Delta T_2 v - u,\\
\frac{dv}{d\tau} &= \Delta T_2 u + \beta(t)T_2 w - v,\\
\frac{dw}{d\tau} &= -\beta(t)T_2 v - (w - w_0)T_2/T_1.
\end{aligned}$$

In this system of equations, all coefficients are now normalized and physically dimensionless, expressed as relevant quotes between relaxation times and products of the Rabi frequency or detuning frequency with the transverse relaxation time. Hence, by taking the normalized parameters

$$\begin{aligned}
\delta &= \Delta T_2,\\
\gamma(t) &= \beta(t)T_2,\\
\eta &= T_1/T_2,
\end{aligned}$$

where $\delta$ can be considered as the normalized detuning from molecular resonance of the medium, $\gamma(t)$ as the normalized Rabi frequency, and $\eta$ as a parameter which describes the relative impact of the longitudinal vs transverse relaxation times, the Bloch equations take the normalized final form

$$\begin{aligned}
\frac{du}{d\tau} &= -\delta v - u,\\
\frac{dv}{d\tau} &= \delta u + \gamma(t)w - v,\\
\frac{dw}{d\tau} &= -\gamma(t)v - (w - w_0)/\eta.
\end{aligned}$$

This normalized form of the Bloch equations has been used throughout the generation of graphs in Figs. 2–11 of this lecture, describing the qualitative impact of different regimes of resonance and relaxation. The normalized Bloch equations were in the simulations shown in Figs. 2–11 integrated by using the standard routine `ODE45()` in MATLAB.

# Home Assignments

**Lecture Notes on Nonlinear Optics**
Nonlinear Optics (5A5513, 5p for advanced undergraduate and doctoral students)
Course given at the Royal Institute of Technology,
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# HOME ASSIGNMENTS

The assignments are of various algebra-intense degrees, and they are not listed in any order of level of difficulty. Solutions to the assignments by means of MapleV, Mathematica, or any other programs for symbolic analysis is encouraged. In the assignments, any reference to "Butcher and Cotter's book" should be read as "P. N. Butcher and D. Cotter, *The Elements of Nonlinear Optics* (Cambridge University Press, Cambridge, 1991), ISBN 0-521-42424-0".

The assignments give a total maximum of 44.0 marks, and successful examination is guaranteed at fifty percent of this level, or 22.0 marks.

Solutions to the assignments should be handed in no later than April 22, 2003, to Fredrik Laurell or Jens Tellefsen, at the department of Laser Physics and Quantum Optics, KTH, Roslagstullsbacken 21, Albanova Universitetscentrum (Fysikcentrum), SE-106 91, Stockholm.

Questions regarding the assignments can be directed to the lecturer and examiner Fredrik Jonsson, preferrably via email to `fredrik.jonsson@proximion.com`.

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**Assignment 1.** (*Spring model for the anharmonic oscillator*) As briefly discussed by Butcher and Cotter in the first chapter of their book, the mechanical spring model can be used as a basic, qualitative model of nonlinear interaction between light and matter, as well as providing a basis for symmetry considerations of particular medium. The spring model can also be used for illustrating the technique of perturbation analysis, frequently used in nonlinear optics as well as in a wide range of different disciplines in physics.

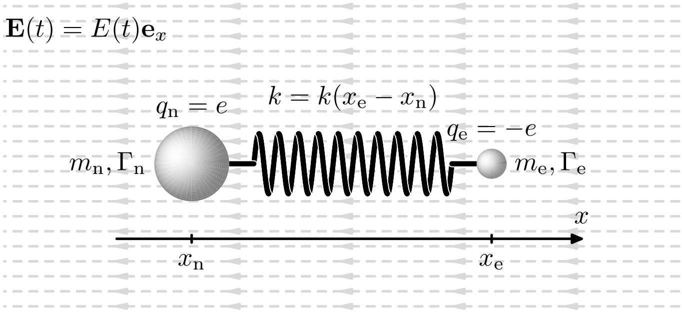


Figure 1. Setup of the one-dimensional spring model.

We here consider a spring model for the anharmonic oscillator, with the forces acting upon the electron and nucleus (with masses $m_{\rm e}$ and $m_{\rm n}$, respectively) given as

$$F_{\rm e} = -eE(t) - \Gamma_{\rm e}\frac{\partial x_{\rm e}}{\partial t} - k_0(x_{\rm e} - x_{\rm n}) - k_1(x_{\rm e} - x_{\rm n})^2,$$

$$F_{\rm n} = eE(t) - \Gamma_{\rm n}\frac{\partial x_{\rm n}}{\partial t} + k_0(x_{\rm e} - x_{\rm n}) + k_1(x_{\rm e} - x_{\rm n})^2,$$

where the first terms in the right hand sides are the electrostatic Coulomb actions on the electron and nucleus from the externally applied electric field, the second terms phenomenologically introduced damping terms, with damping constants $\Gamma_{\rm e}$ and $\Gamma_{\rm n}$ for the electron and nucleus, and where the rest of the terms are the spring actions, modeling the mutual Coulomb interaction between the electron and nucleus.

This assignment not only serves as to demonstrate the anharmonic oscillator, but also as to give an exercise in perturbation analysis, a tool that later on will be used in the quantum mechanical description of interaction between light and matter.

**1a.** [*0.2 p*] Formulate the Newtonian equations of motion for the real-valued electric dipole moment $p(t) = -e(x_{\rm e}(t) - x_{\rm n}(t))$. In order to algebraically simplify the obtained expressions, it might be helpful to introduce the centre of mass $R(t) = (m_{\rm e}x_{\rm e}(t) + m_{\rm n}x_{\rm n}(t))/(m_{\rm e} + m_{\rm n})$ and the reduced mass $m_{\rm r} = m_{\rm e}m_{\rm n}/(m_{\rm e} + m_{\rm n})$ of the system. In particular, show how the equation of motion for the dipole moment decouple from the equation of motion for the centre of mass whenever $\Gamma_{\rm n}/m_{\rm n} = \Gamma_{\rm e}/m_{\rm e}$.

**1b.** [*0.2 p*] Express the real-valued electric dipole moment of the anharmonic oscillator as a perturbation series

$$p(t) = p^{(0)}(t) + p^{(1)}(t) + p^{(2)}(t) + \ldots,$$

where each term in the series is proportional to the applied electrical field strength to the power as indicated in the superscript of repective term, and formulate the system of $n + 1$ equations for $p^{(k)}(t)$, for $k = 0, 1, 2, \ldots, n$, that define the time evolution of the electric dipole.

**1c.** [*0.2 p*] Introduce a complex notation for the electric dipole moment and the applied monochromatic electric field (the latter at angular frequency $\omega$), and reformulate the equations of motion for the complex-valued terms of the perturbation series for the electric dipole moment.

**1d.** [*0.4 p*] Solve the obtained system of differential equations for the general, non-steady state solution for the first order perturbation term of the electric dipole moment, assuming a known initial condition at $t = 0$, and $p^{(0)}(0) = 0$. Hint: What will the solution for $p^{(0)}(t)$ be?

**1e.** [*0.4 p*] In steady state, for an ensemble of $N$ identical and mutually noninteracting electric dipole oscillators per unit volume, give an expression for the complex-valued linear electric susceptibility $\chi^{(1)}_{xx}(-\omega;\omega)$. Hint: In steady state, what will the homogeneous solution for the first order perturbation term be?

**1f.** [*0.2 p*] In steady state, give an expression for the dispersion $\partial n(\omega)/\partial\omega$.

**1g.** [*0.4 p*] In steady state, still for an ensemble of $N$ identical and mutually noninteracting electric dipole oscillators per unit volume, give expressions for the complex-valued second-order electric susceptibilities $\chi^{(2)}_{xxx}(-2\omega;\omega,\omega)$ and $\chi^{(2)}_{xxx}(0;\omega,-\omega)$.

**1h.** [*0.4 p*] Can the quadratic term in the expression for the spring action cause the (third-order) optical Kerr-effect, that is to say, can it give rise to a nonzero electric dipolar susceptibility $\chi^{(3)}_{xxxx}(-\omega;\omega,\omega,-\omega)$?

**1i.** [*0.3 p*] Can the quadratic term in the expression for the spring action cause the (third-order) effect of optical third harmonic generation, that is to say, can it give rise to a nonzero electric dipolar susceptibility $\chi^{(3)}_{xxxx}(-3\omega;\omega,\omega,\omega)$?

**1j.** [*0.4 p*] If cubic terms $\pm k_2(x_{\rm e}-x_{\rm n})^3$ were to be included in the expression for the spring action on the electron and nucleus, respectively, would these terms be able to induce an additional component at angular frequency $2\omega$?

**1k.** [*0.3 p*] If cubic terms $\pm k_2(x_{\rm e}-x_{\rm n})^3$ were to be included in the expression for the spring action on the electron and nucleus, respectively, would these terms be able to cause optical Kerr-effect?

**Assignment 2.** (*Review of quantum mechanics – The density matrix*) While the mechanical spring model provides a qualitative picture that for many cases is sufficcient, it nevertheless fails to give a proper basis for a description of interaction between light and matter down to the molecular level. The aim with this assignment is to provide a summary of the maybe somewhat abstract (but necessary) quantum mechanical tools that are needed for the description of interaction between light and matter.

This assignment virtually covers all the quantum mechanics needed for understanding the basis of the nonlinear optics described in this course, and it also demonstrates the point in nonlinear optics at which we choose either to proceed with perturbation analysis of the matrix elements of the density operator (introducing the nonlinear susceptibility formalism), or to solve for the matrix elements directly by instead using the Bloch equation.

**2a.** [*2.5 p*] Starting from the time dependent Schrödinger equation for the molecular wave function, derive the equation of motion for the density matrix in the Schrödinger picture. In Butcher and Cotters book, this corresponds to filling in the details in the discussion from Eq. (3.1) to Eq. (3.50). (This may sound like a formidable task of deriving fifty quantum mechanical equations; however, most of these equations are simple definitions and corollaries, and by following the outline as described in Butcher and Cotters book, this is quite a straightforward task which hopefully will give a deeper understanding of the origin of optical nonlinearities.)

**2b.** [*1.0 p*] Starting from the equation of motion for the density matrix in the Schrödinger picture, apply perturbation analysis and derive the general solution for the $n$th order term of the density operator. In Butcher and Cotters book, this corresponds to filling in the details in the discussion from Eq. (3.50) to Eq. (3.82).

**2c.** [*0.4 p*] In your own words, explain the difference between the “Schrödinger picture” and “interaction picture” in quantum mechanics.

**Assignment 3.** (*Nonlinear optics of the hydrogen atom*) Among the simplest systems that are possible to analyse analytically, the hydrogen and helium atoms serve as to give models for nonlinear optical effects. In this assignment, the task is to essentially to perform the calculations necessary for predicting the refractive index and optical Kerr-effect of hydrogen gas. However, in order to simplify the task, we will instead perform the calculations for an *ensemble of hydrogen atoms*, rather than for the in nature commonly appearing diatomic $H_2$ molecules. This assumption makes this assignment a somewhat artificial construction, which though nevertheless serve well as to illustrate the steps leading from the atomic wavefunction (or molecular wavefunction, if we were to consider a “real” gas of $H_2$ molecules) to the concept of linear refractive index (given by the first order electric susceptibility) and optical Kerr-effect.

The wave function of the hydrogen atom is in spherical coordinates $(r, \varphi, \vartheta)$ given as [H. Haken and H. C. Wolf, *The physics of atoms and quanta*, 4th ed. (Springer-Verlag, Berlin, 1993)]

$$\psi_{nlm}(r, \vartheta, \varphi) = R_{nl}(r) P_l^m(\cos\vartheta) \exp(im\varphi),$$

where $n$ is the principal quantum number, $l$ the angular momentum quantum number, and $m$ the magnetic quantum number or directional quantum number. These numbers assume the values

$$n = 1, 2, \dots, \qquad 0 \le l \le n-1, \qquad -l \le m \le l.$$

In the angular part of the expression for the wave function,

$$P_l^0(\xi) = P_l(\xi) = \frac{1}{l!2^l}\frac{d^l}{d\xi^l}(\xi^2 - 1)^l$$

are the Legendre polynomials, and

$$P_l^m(\xi) = (1 - \xi^2)^{m/2}\frac{d^m}{d\xi^m}P_l(\xi)$$

are the associated Legendre functions, while for the radial part

$$R_{nl}(r) = N_{nl}\exp(-\kappa_n r) r^l L_{n+1}^{2l+1}(2\kappa_n r),$$

where $N_{nl}$ is a normalization constant obtained from the condition

$$\int_0^\infty R_{nl}^2(r) r^2\, dr = 1,$$

and

$$\kappa_n = \left(\frac{m_0 Z e^2}{\hbar^2 4\pi\varepsilon_0}\right)\frac{1}{n}.$$

Above $L_{n+1}^{2l+1}$ are the Laguerre function of the second kind, obtained from the Laguerre polynomials through $2l+1$-fold differentiation

$$L_{n+1}^{2l+1}(\xi) = \frac{d^{2l+1}}{d\xi^{2l+1}}L_{n+1}(\xi).$$

The temperature of the (artificial) hydrogen gas here condidered is 25 degrees Celcius, measured at 1077 mBar pressure, and it is reasonable to assume that the hydrogen atoms in the ensemble are mutually noninteracting.

**3a.** [*1.0 p*] Calculate the first order electric dipolar susceptibility $\chi^{(1)}_{\mu\alpha}(-\omega; \omega)$ and the linear refractive index $n(\omega)$, for a vacuum wavelength of 580 nm. Hint: Consider hydrogen being in the ground state and identify the atomic trasition frequencies closest to the frequency range of interest. Is it likely that resonant interaction should be taken into account?

**3b.** [*1.0 p*] Calculate the second order electric dipolar susceptibility $\chi^{(2)}_{\mu\alpha\beta}(-2\omega; \omega, \omega)$, for the same vacuum wavelength as in assignment 3a.

**3c.** [*1.0 p*] Calculate the third order electric dipolar susceptibility $\chi^{(3)}_{\mu\alpha\beta\gamma}(-\omega; \omega, \omega, -\omega)$, for the same vacuum wavelength as in assignment 3a.

**3d.** [*0.2 p*] Would you *a priori* (i. e. without actually performing the calculations) expect the elements of the linear susceptibility tensor $\chi^{(1)}_{\mu\alpha}(-\omega; \omega)$ to be lower or higher for an ensemble of helium atoms than for the ensemble of hydrogen atoms?

**3e.** [*0.2 p*] Would you *a priori* expect the elements of the quadratic susceptibility tensor $\chi^{(2)}_{\mu\alpha\beta}(-2\omega; \omega, \omega)$ to be lower or higher for an ensemble of helium atoms than for the ensemble of hydrogen atoms?

**Assignment 4.** (*Neumann's principle in linear optics*) Being a powerful tool in a variety of disciplines in physics, Neumann's principle simply states that any type of symmetry which is exhibited by the point symmetry group of the medium also is possessed by every physical property of the medium.

In many textbooks in linear optics and electromagnetic wave propagation, it is taken more or less as an axiom that the linear susceptibility tensors of cubic media are diagonal with all elements equal, i. e. in form identical to the linear susceptibility tensor of isotropic media. However, one rarely finds a strict derivation of this fact, and since Neumann's principle is a powerful tool in nonlinear optics, this assignment serves as an introduction to analysis of point symmetries of higher order, nonlinear interactions.

**4a.** [*0.2 p*] Is an isotropic medium always centrosymmetric, that is to say, does it always possess a center of inversion? (This question is of a general character, and applies to linear as well as nonlinear optical regimes.)

**4b.** [*0.2 p*] Is a centrosymmetric medium always isotropic?

**4c.** [*0.8 p*] Show that the electric dipolar susceptibility tensor $\chi^{(1)}_{\mu\alpha}(-\omega;\omega)$ takes the form $\chi^{(1)}_{xx}(-\omega;\omega)\delta_{\mu\alpha}$ for a medium belonging to cubic point-symmetry class.

**4d.** [*0.8 p*] In addition, show that the susceptibility tensor $\chi^{(1)}_{xx}(-\omega;\omega)\delta_{\mu\alpha}$ also is isotropic, i. e. that it in fact is left invariant under *any* rotation of the cordinate axes of the medium.

**4e.** [*0.8 p*] Show that the electric dipolar susceptibility tensor $\chi^{(1)}_{\mu\alpha} = \chi^{(1)}_{\mu\alpha}(-\omega;\omega)$ takes the form

$$\chi^{(1)}(-\omega;\omega) = \begin{pmatrix} \chi^{(1)}_{xx} & 0 & 0 \\ 0 & \chi^{(1)}_{xx} & 0 \\ 0 & 0 & \chi^{(1)}_{zz} \end{pmatrix}$$

for trigonal media.

**Assignment 5.** (*Neumann's principle in nonlinear optics*) As shown in the course, the principles for reduction of the elements of first-order susceptibility tensors by means of direct inspection can easily be extended and applied to susceptibilities describing nonlinear interactions as well.

While, for example, media belonging to the cubic crystallographic point symmetry group all behave as isotropic in linear optics, they will in the nonlinear optical regime generally be of non-isotropic character. In addition, calculations of symmetry properties of higher-order susceptibilities tend to be less intuitive than in linear optics. However, Neumann's principle, together with the transformation properties of polar tensors (such as electric dipolar susceptibilities), provides a powerful tool that is well suited for implementation in computer languages for symbolic analysis, significantly reducing the effort needed for solving the problems.

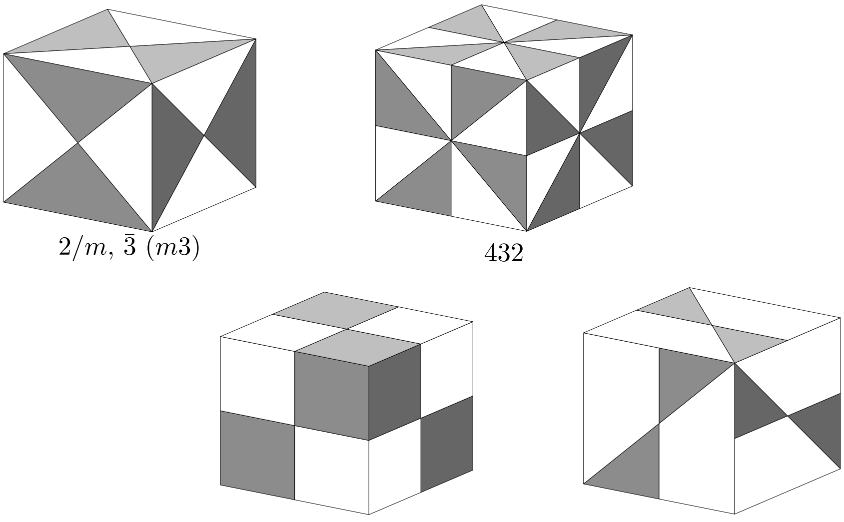
**5a.** [*0.4 p*] Within the electric dipole approximation, the linear susceptibility tensor of isotropic media is always diagonal with all elements equal (as, hopefully, also shown in assignment 5), hence preventing any coupling between polarization states. Is it true that no such polarization state coupling appears in isotropic media in the nonlinear optical regime as well? Prove or give a counterexample!

**5b.** [*0.3 p*] Prove that the elements of $\chi^{(2n)}_{\mu\alpha_1\cdots\alpha_{2n}}(-\omega_\sigma;\omega_1,\ldots,\omega_{2n})$ are all identically zero for media possessing a center of inversion, for $n$ being a positive integer.

**5c.** [*0.6 p*] In Fig. 2, objects possessing the point symmetry properties of crystallographic cubic point symmetry classes 432, $m3$, $\bar{4}3m$, and 23 are shown [N. W. Ashcroft and N. D. Mermin, *Solid state physics* (Saunders College Publishing, Orlando, 1976)].

The description of the objects is closed by the observation that the objects of these point symmetry classes all should be left invariant under any ±120 degrees rotation around any space diagonal axis, going through diametrically opposite corners.

Without entering any calculations, is it possible to immediately tell which of those point symmetry classes that will lead to an identically zero second order electric dipolar susceptibility? Motivate with a few figures!



$2/m$, $\bar{3}$ ($m3$)

432

$\bar{4}3m$

23

Figure 2. Objects possessing the point symmetry properties of 432, $m3$, $\bar{4}3m$, and 23.

**5d.** [*1.0 p*] For any of the point symmetry groups 432, $m3$, $\bar{4}3m$, or 23 that possesses a nonzero second order susceptibility, derive the set of nonzero and independent tensor elements of $\chi^{(2)}_{\mu\alpha\beta}(-\omega_\sigma;\omega_1,\omega_2)$.

**5e.** [*0.6 p*] For a light beam propagating in the $z$-direction, will any of the point symmetry groups 432, $m3$, $\bar{4}3m$, or 23 give a second order polarization density that is left invariant under rotation of the medium around the $z$-axis? (Of course under the assumption that the optical wave properties, such as the polarization state of the wave, are kept fixed in space under the rotation.)

**5f.** [*0.6 p*] Prove that in isotropic media

$$\chi^{(3)}_{xxxx} = \chi^{(3)}_{xxyy} + \chi^{(3)}_{xyxy} + \chi^{(3)}_{xyyx},$$

for arbitrary frequency arguments.

**5g.** [*0.6 p*] As listed in Appendix 3 of Butcher and Cotter's book (and as also is straightforward to verify by applying Neumann's principle and the technique of direct inspection, as in assignment 5f above), the general third-order electric dipolar susceptibility of isotropic media, for arbitrary frequency arguments, possesses the property

$$\chi^{(3)}_{xxxx} = \chi^{(3)}_{xxyy} + \chi^{(3)}_{xyxy} + \chi^{(3)}_{xyyx},$$

that is to say, the $xxxx$-element equals to the sum of the (three) distinct permutations of the tensor elements with $x$ in the first index, and with equal total number of $x$ and $y$ appearing in the indices. As a generalization of this relation, one may hence expect the fifth-order susceptibility of isotropic media, again for arbitrary frequency arguments, to possess the property

$$\begin{aligned}\chi^{(5)}_{xxxxxx} = \chi^{(5)}_{xxxyyy} &+ \chi^{(5)}_{xxyxyy} + \chi^{(5)}_{xxyyxy}\\ &+ \chi^{(5)}_{xxyyyx} + \chi^{(5)}_{xyxxyy} + \chi^{(5)}_{xyxyxy}\\ &+ \chi^{(5)}_{xyxyyx} + \chi^{(5)}_{xyyxxy} + \chi^{(5)}_{xyyxyx} + \chi^{(5)}_{xyyyxx},\end{aligned}$$

that is to say, suggesting that the $xxxxxx$-element equals to the sum of the (ten) distinct permutations of the tensor elements with $x$ in the first index, and with equal total number of $x$ and $y$ appearing in the indices. The question is: Is the above statement for the fifth order susceptibility really true?

**Assignment 6.** (*Review of quantum mechanics – The resonant two-level system*) In assignment 2, the analysis of the density operator is performed essentially under the assumption of nonresonant interaction between light and matter, even though the principles of perturbation analysis apply to weakly resonant systems as well. For strongly resonant interactions, the higher order perturbation terms can no longer be considered as small compared to lower order ones, and eventually a description in terms of the susceptibility formalism fails to give a proper picture. For such cases, the density operator is instead described by Bloch-type equations, which when solved provide the observable, macroscopic properties of the medium by evaluation of quantum-mechanical traces.

The outline for this technique, as in this assignment described for the two-level system, can equally well be applied to $n$-level systems as well, though with an increasing algebraic complexity.

**6a.** [*1.6 p*] Starting from the time dependent Schrödinger equation for the two-level molecular wave function, derive the Bloch equations in the rotating-wave approximation. In Butcher and Cotters book, this corresponds to filling in the details in the discussion from Eq. (6.22) to Eq. (6.52).

**6b.** [*0.4 p*] Can the rotating wave approximation be considered as a kind of phase matching, similar to the phase matching conditions that frequently appear elsewhere in nonlinear optics? Motivate.

**Assignment 7.** (*Nonlinear optics of silica*) In the wavelength range from ultraviolet to mid-infrared, the Sellmeier expression for the linear, field-independent refractive index $n(\lambda)$ of pure silica ($SiO_2$, quartz) is given as

$$n^2(\lambda) = 1 + \sum_{k=1}^{3} \frac{A_k \lambda^2}{\lambda^2 - \lambda_k^2},$$

where $\lambda$ is the vacuum wavelength of the light, and where the physically dimensionless constants $A_k$ are experimentally found to be

$$A_1 = 0.6961663, \qquad A_2 = 0.4079426, \qquad A_3 = 0.8974794.$$

The resonance vacuum wavelengths in the Sellmeier expression are for silica given as

$$\lambda_1 = 0.0684043\,\mu\text{m}, \qquad \lambda_2 = 0.1162414\ \mu\text{m}, \qquad \lambda_3 = 9.896161\ \mu\text{m},$$

where $\lambda_1$ and $\lambda_2$ are electronic resonances, and $\lambda_3$ a lattice vibrational resonance.

**7a.** [*0.4 p*] Using the Sellmeier expression, give an expression for the $\chi^{(1)}_{\alpha\beta}(-\omega;\omega)$ in terms of the parameters $A_k$ and $\lambda_k$. You may here assume the silica to be essentially isotropic. (This assumption holds for amorphous $SiO_2$, but it should be emphasized that this approximation in general does not hold for quartz in a pure crystalline (trigonal) state, which belongs to the crystallographic point symmetry class 32.)

**7b.** [*0.2 p*] In the visible range, say at vacuum wavelength 700 nm, calculate the relative contribution from the vibrational term to the index of refraction.

**7c.** [*0.6 p*] Using the obtained expression for $\chi^{(1)}_{\alpha\beta}(-\omega;\omega)$, identify and express the magnitudes $|er^{\alpha}_{ab}|$ of the corresponding matrix elements of the electric dipole moments, in terms of the $\lambda_k$ and $A_k$ parameters of the Sellmeier expression. Calculate the numerical values of $|er^{\alpha}_{ab}|$ in regular SI units. You may assume that (I) the $SiO_2$ molecules are in the ground state, and that (II) the lattice vibrational contribution safely can be neglected in comparison with the electronic contributions.

**7d.** [*0.4 p*] Having obtained the magnitudes of the matrix elements of the involved electric dipole moments of quartz, apply the Unsöld approximation in order to obtain a simplified, effective expression for the refractive index of the form

$$n^2_{\text{eff}}(\lambda) = 1 + \frac{A_{\text{eff}}\lambda^2}{\lambda^2 - \lambda^2_{\text{eff}}},$$

where the task now is to find expressions for $A_{\text{eff}}$ and $\lambda_{\text{eff}}$. Also find the maximum deviation of the obtained refractive index from the one given by the original Sellmeier expression, taken over the vacuum wavelength interval from 400 nm to 870 nm.

**7e.** [*0.8 p*] Make use of the obtained expressions of the matrix elements of the involved electric dipole moments of quartz to calculate an approximative value of the third-order susceptibility $\chi^{(3)}_{xxxx}(-\omega;\omega,\omega,-\omega)$, taken over the same vacuum wavelength interval as in assignment 7d.

**Assignment 8.** (*The Bloch equation*) Suppose a $\mathrm{Mg}^{26}$ atom is trapped and excited with a laser beam tuned close to the $3s^2\,{}^1S_0$–$3s3p\,{}^1P_1$ transition (with the angular frequency of the laser light taken as $\omega = \omega_{3s3p\,{}^1P_1} - \omega_{3s^2\,{}^1S_0} + \Delta\omega$). Suppose the atom can be considered as a two-level system, and that the excitation hence can be described by the Bloch equations for a two-level system. The energy level for the $3s3p\,{}^1P_1$ state of $\mathrm{Mg}^{26}$ is 35051 $\mathrm{cm}^{-1}$. (See, for example, *Atomic Energy Levels*, National Bureau of Standards, Circular 467.)

**8a.** [*1.0 p*] For a circular laser beam of 1 mm radius, with top-hat profile, calculate at which optical power the Rabi angular frequency $\Omega$ equals to $\Omega = 1/(3\tau)$, where $\tau = 2$ ns is the life time of the $3s3p\,{}^1P_1$ state of $\mathrm{Mg}^{26}$.

**8b.** [*0.8 p*] Calculate the probability that the atom is in the upper state for $\Delta\omega = 0$ and $\Delta\omega = 1/\tau$, for steady state between the optical field and relaxation processes.

Hint: Do not start the calculations until you have made it clear in which way the assumption of steady state simplifies the problem.

**Assignment 9.** (*Vector model for the Bloch equation*) The time dependent solution to the Bloch equations for an optical field tuned to exact resonance, when the electromagnetic field and the natural life time are the only broadening mechanisms, and with the initial values $u(0) = v(0) = 0$ and $w(0) = -1$, are

$$\begin{aligned} u(t) &= 0, \\ v(t) &= \frac{2\Omega T_2}{2 + (\Omega T_2)^2}\left\{\left[\cos(\beta t) + \frac{1 - (\Omega T_2)^2}{2\beta T_2}\sin(\beta t)\right]\exp(-3t/2T_2) - 1\right\}, \\ w(t) &= \frac{(\Omega T_2)^2}{2 + (\Omega T_2)^2}\left\{1 - \left[\cos(\beta t) + \frac{3}{2\beta T_2}\sin(\beta t)\right]\exp(-3t/2T_2)\right\} - 1, \end{aligned}$$

with $\beta = [\Omega^2 - 1/(4T_2^2)]^{1/2}$. Now suppose that $\mathrm{Mg}^{26}$ is contained in a cell at 200 degrees Celsius. Choose $\Delta\omega = 0$ and an optical intensity such that $\Omega T_2 = 5$.

**9a.** [*0.7 p*] As the optical field is switched on, the atoms of the $\mathrm{Mg}^{26}$ sample are excited to the upper level. As the Bloch vector rotates, the population in the upper state will at some time $T_0$ start to decrease again. Calculate the time $T_0$ and the population density $\rho_{22}(T_0)$ at this time.

**9b.** [*1.0 p*] Calculate the length of the cell in order for the incindent optical intensity to decrease by 10 percent at $t = T_0$. The energy emitted by spontaneous emission is here neglected for simplicity, as in 9c.

**9c.** [*0.7 p*] Calculate $\rho_{22}(2T_0)$ and the relative amount of the incident light energy that has been absorbed at $t = 2T_0$. If the system had been undamped, how much of the light would then have been absorbed after the Bloch vector had rotated one turn?

**9d.** [*1.0 p*] Draw approximately how the Bloch vector evolve, from the time at which the light is switched on until steady state is reached, and how it then evolves as the light is switched off again.

**Assignment 10.** (*Optical bistability*) As a medium possessing optical Kerr-effect is placed inside a cavity, either in a ring cavity (in which only unidirectional wave propagation is considered) or in a Fabry-Pérot cavity (in which counter-propagating waves may couple as well), one introduces an optical feedback to the nonlinear interaction between light and matter. For certain configurations this feedback leads to phenomena where the system is latched into stable states, in particular with the transmitted intensity taking two possible stable values for a given input, so-called optical bistability, where the two states of the system can be pictured as "low" (switched off) and "high" (switched on).

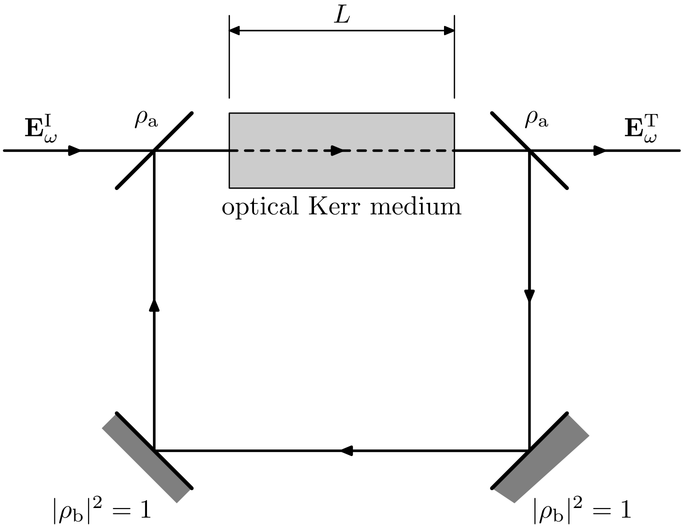


Figure 3. Setup of the ring cavity for optical bistability.

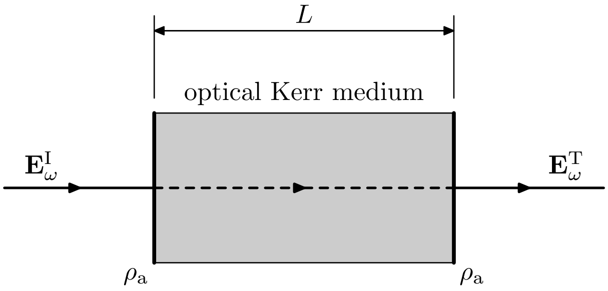


Figure 4. Setup of the Fabry-Pérot cavity for optical bistability.

**10a.** [*1.0 p*] For the ring cavity configuration, using an all-dispersive isotropic optical Kerr-medium, formulate the equation that relates the transmitted intensity $I_{\rm T} = (\varepsilon_0 c/2)|\mathbf{E}^{\rm T}_{\omega}|^2$ to the incident intensity $I_{\rm I} = (\varepsilon_0 c/2)|\mathbf{E}^{\rm I}_{\omega}|^2$. In the ring cavity configuration, the two semitransparent mirrors are assumed to have a complex amplitude reflectance of $\rho_{\rm a}$ each, while the rest of the mirrors have a reflectance of unity. For simplicity, you may neglect the reflectance of the optical beam at the end surfaces of the nonlinear medium, hence neglecting any back-reflected wave in the ring cavity. You may also assume the light to be linearly polarized (TE or TM with respect to the plane of incidence on the mirrors), enabling a scalar theory of wave propagation to be applicable to the problem.

**10b.** [*1.0 p*] Same as 10a, but now for the Fabry-Pérot cavity configuration, where the nonlinear medium is enclosed by two mirrors of complex amplitude reflectance $\rho_{\rm a}$.

**10c.** [*0.4 p*] For the ring cavity or the Fabry-Pérot cavity configuration (your choice), explain how the obtained relations between transmitted and incident intensities lead to optical bistability.

**10d.** [*0.4 p*] In purely dispersive optical bistability, for which there is no absorption, what happens to the light energy contained in the cavity as the system switches from the upper to the lower state of transmission?

*Instead of solving the above problems, you may solve one of the following two additional problems on optical bistability:*

**10′.** [*2.8 p*] Formulate the equation for absorptive optical bistability with an inhomogeneously broadened isotropic two-level medium. Prove that this equation can (or cannot) exhibit optical bistability.

**10″.** [*2.8 p*] Compare and contrast absorptive and dispersive optical bistability for isotropic media. How are the incident and transmitted field phases related for each case?

**Assignment 11.** (*Optical solitons*) As shown in the course, optical solitons are solutions to the wave equation in optical Kerr-media, where the intensity-dependent refractive index of the medium balance the dispersion.

**11a.** [*1.0 p*] Prove that for an initial pulse shape given by $u(0, s) = N \operatorname{sech}(s)$, with $N = 1$ for the fundamental soliton, an exact soliton solution $u(\zeta, s)$ to the nonlinear Schrödinger equation is given by

$$u(\zeta, s) = \operatorname{sech}(s) \exp(i\zeta/2).$$

**11b.** [*0.4 p*] For quartz, the numerical value of the $xyyx$ element of the nonlinear susceptibility is given as

$$\chi^{(3)}_{xyyx}(-\omega; \omega, \omega, -\omega) = 8.0 \times 10^{-23}\ \mathrm{m^2/V^2},$$

at a vacuum wavelength of 1 694 nm. Calculate the numerical values of $\chi^{(3)}_{xxxx}(-\omega; \omega, \omega, -\omega)$ and the nonlinear refractive index $n_2$ at this vacuum wavelength. Hint: Consider Kleinman symmetry to hold.

**11c.** [*0.6 p*] Suggest pulse duration time and peak intensity of an optical pulse at 1 694 nm vacuum wavelength in order to form a fundamental soliton in quartz. Hint: The linear optical properties of quartz at this wavelength may, for example, be obtained from the Sellmeier expression given in assignment 7.

**Assignment 12.** (*Third harmonic generation in isotropic media*) In this assignment, the task is to formulate the theory of third harmonic generation, in similar to the second harmonic generation analyzed in the lectures. While second harmonic generation requires the medium to be non-centrosymmetric (if the medium is centrosymmetric, the second order susceptibility is zero, as shown in the lectures), the third harmonic generation does not have this constraint. In particular, we will in this assignment consider the third harmonic generation in isotropic media.

Note that even though this problem concerns isotropic media, the third harmonic signal is highly dependent on the polarization state of the pump wave. As an example, circularly polarized light cannot be used for third harmonic generation in isotropic media. (This is, for example, shown in section 5.3.4, page 144, of Butcher and Cotter's *The Elements of Nonlinear Optics*.)

This assignment serves as an illustration of a general scheme for solving wave propagation problems in nonlinear optics, as well as an example of the application of the Butcher and Cotter convention of degeneracy factors in nonlinear optics. When solving this assignment, you may benefit from applying the scheme for solving problems in nonlinear optics, as outlined in lecture nine of this course.

**12a.** [*1.0 p*] In the infinite plane wave limit, assuming all fields to be polarized in the $xy$-plane and propagating in the positive $z$-direction, formulate the polarization density of the isotropic medium, at the angular frequency $\omega$ of the fundamental (pump) wave.

**12b.** [*1.0 p*] Using the same assumptions as in 12a, formulate the polarization density at the angular frequency $3\omega$ of the third harmonic wave.

**12c.** [*1.0 p*] For a continuous-wave configuration, with the pump wave being linearly polarized in the $x$-direction, formulate the coupled system of wave equations for the pump and third harmonic wave. What will the polarization state of the third harmonic wave be?

**12d.** [*1.0 p*] Assuming the conversion efficiency from the pump to the third harmonic wave to be small, we may assume the pump wave to be essentially non-depleted, i. e. $\mathbf{A}_\omega(z) \approx \text{const.}$ (non-depleted pump approximation). For this case, derive an expression for the intensity of the third harmonic wave after a geometrical propagation distance $L$. (As a hint of the form of this expression, you may consult Butcher and Cotter, Eq. (7.30).)

**Assignment 13.** (*The degeneracy factor*) The system of including degenerate terms of the polarization density by means of the degeneracy factor $K(-\omega_\sigma;\omega_1,\ldots,\omega_n)$ is very convenient, since it, for example, allow us to use only one of the possible combinations of $(\omega_1,\ldots,\omega_n)$ for the description of some particular nonlinear interaction, instead of considering all possible permutations. However, the convention of taking the real-valued electrical field as $\mathbf{E}(\mathbf{r},t)=\mathrm{Re}[\mathbf{E}_\omega\exp(-i\omega t)]$ is not universal, and in the 1960's, the convention was rather to take it as $\mathbf{E}(\mathbf{r},t)=\mathbf{E}_\omega\exp(-i\omega t)+\text{c. c.}$, i. e. as twice of the convention of Butcher and Cotter. Sometimes it is though useful to apply the "old" convention, especially if it comes to the interpretation of pioneering publications in the field of nonlinear optics.

**13a.** [*3.0 p*] Reformulate the degeneracy factor given by Butcher and Cotter, Eq. (2.56), for the case where the real-valued field instead is taken as $\mathbf{E}(\mathbf{r},t)=\mathbf{E}_\omega\exp(-i\omega t)+\text{c. c.}$.

**13b.** [*1.0 p*] Why would one suspect that a misinterpretation of the convention of the real-valued field has a greater impact in nonlinear optics rather than in linear optics?

**Assignment 14.** (*Electro-optic phase modulation – The Pockels effect*) In this assignment, we consider an electro-optic light modulator, consisting of a nonlinear optical crystal of litium niobate, $\mathrm{LiNbO_3}$, with electrodes plated on the top and bottom surfaces, as shown in Fig. 5. As a DC voltage is applied to the electrodes, the refractive index experienced by the light is changed through the linear electro-optical Pockels effect, described by the second-order susceptibility

$$\chi^{(2)}_{\mu\alpha\beta}(-\omega;\omega,0),$$

and the resulting phase shift of the light over the crystal can be used in an interferometric setup, for modulation of light intensity.

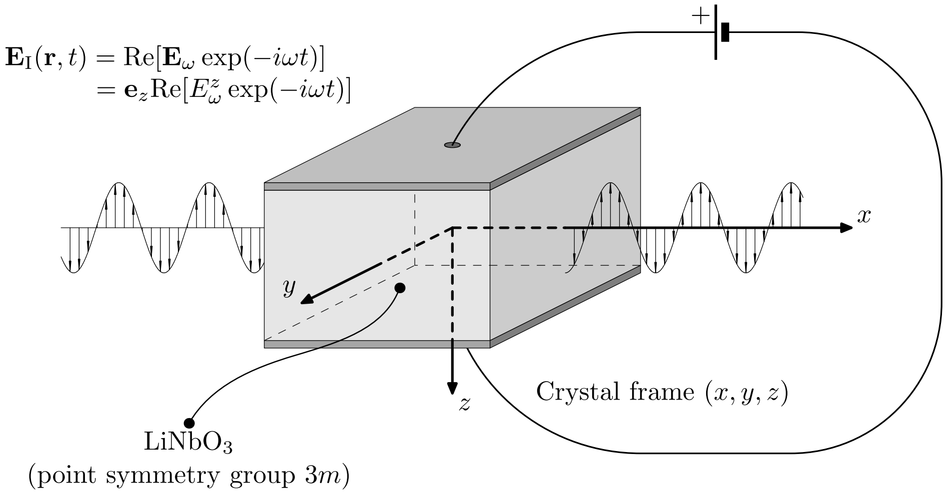


Figure 5. The phase shifting element of the litium niobate electro-optic modulator.

As one example of an interferometric setup in which the phase shift of the light may be detected as a modulation of intensity is the Mach-Zender configuration, as shown in Fig. 6, where the input light waveguide is split into two, with each waveguide now carrying half of the input intensity. A phase shifting electro-optical element is placed in one of the arms of the interferometer (the lower one as shown in Fig. 6), and by modulating the the applied voltage over it, a relative phase shift between the light in the two arms is obtained. As the two guided light waves are combined again, the resulting output will experience either constructive interference (if the relative phase shift between the beams is a multiple of $2\pi$), or destructive interference (if the phase shift instead is $\pi$ plus a multiple of $2\pi$).

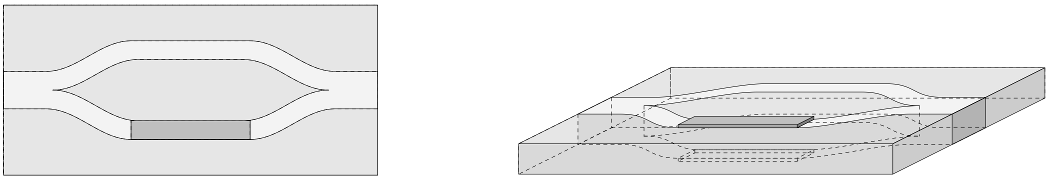


Figure 6. The Mach-Zender configuration of an integrated electro-optic modulator.

The task is now to formulate the theory of the basic electro-optic element shown in Fig. 5.

**14a.** [*1.0 p*] Assuming the applied voltage over the crystal to generate a homogeneous DC electrical field $E^z_{\mathrm{DC}}$ in the $z$-direction, and also assuming the optical field to be linearly polarized along the $z$-direction, propagating in the positive $x$-direction, formulate the polarization density at angular frequency $\omega$ of the medium.

**14b.** [*1.0 p*] For a continuous-wave optical field, formulate the wave equation for the wave propagation through the medium.

**14c.** [*1.0 p*] Solve the wave equation for the envelope of the electrical field. To solve the wave equation, it might be helpful to apply the slowly-varying envelope approximation.

**14d.** [*1.0 p*] Formulate the refractive index change of the medium due to the Pockels effect, as function of the applied DC electrical field $E^z_{\mathrm{DC}}$.