Optical parametric generation and phase matching in magneto-optic media

Fredrik Jonsson

Department of Physics, The Royal Institute of Technology, S-100 44 Stockholm, Sweden

Christos Flytzanis

Laboratoire d'Optique Quantique, Ecole Polytechnique, F-911 28 Palaiseau Cedex, France

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We derive the equations and main characteristics of optical parametric generation in magneto-optic media. The key performances are discussed in terms of Stokes parameters and conservation laws of electromagnetic linear and angular momentum.

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Three-wave coherent interactions of the type $\omega_1 + \omega_2 = \omega_3$ with all energy exchanges occurring among the three field components without loss to the material medium were analyzed previously almost exclusively with linearly polarized light fields in linearly birefringent (anisotropic) crystals in which one could achieve phase matching by exploiting the linear birefringence. An alternative approach is to exploit the circular birefringence, natural or artificially induced, to achieve phase matching. This procedure has already been discussed for second- and third-harmonic generation in the case of optically active crystals[1,2] (natural optical activity) for linearly polarized light but has had no followup.

Here we analyze optical parametric interactions, specifically, optical parametric generation and amplification $\omega_1 = \omega_3 - \omega_2$, for the case of artificial optical activity induced by a static magnetic field in the Faraday configuration[3] with all three beams propagating collinearly in the direction of the applied static magnetic field, chosen to be the z axis in the laboratory reference frame (x, y, z). The major advantage of this configuration is that the magnetic field can be used for continuous tuning of the phase matching in a fixed geometry, eliminating any walk-off effects. To illustrate the concept we have chosen propagation in an isotropic noncentrosymmetric medium, or along the (111) or c axis in $\bar{4}3m$ (zinc blende) or $6mm$ (wurtzite) crystalline structures, respectively, to evaluate the efficiency and characteristics of the parametric process; the approach can be extended and applied to more complex crystalline configurations. The simplifying features here are that the analysis can be conducted in terms of circularly polarized modes that are the natural eigenmodes for these particular orientations in the Faraday configuration and that the results can be cast into a concise analytical form in terms of Stokes parameters.

We assume quasi-monochromatic fields $\mathbf{E}(\mathbf{r}, t) = \sum_{k=1}^{3} \mathrm{Re}[\mathbf{E}_{\omega_k} \exp(-i\omega_k t)]$ and keep only lowest-order optical and magneto-optical nonlinearities; each frequency component obeys the wave equation

$$\frac{\partial^2 \mathbf{E}_{\omega_k}}{\partial z^2} + \frac{\omega_k^2}{c^2}\mathbf{E}_{\omega_k} = -\omega_k^2 \mu_0 \mathbf{P}_{\omega_k}, \qquad (1)$$

where $k = 1, 2, 3$ designate idler, signal, and pump, respectively, and where the electric polarization density for each frequency component ω_k is

$$\mathbf{P}_{\omega_k} = \mathbf{P}_{\omega_k}^{(\mathrm{ee})} + \mathbf{P}_{\omega_k}^{(\mathrm{eem})} + \mathbf{P}_{\omega_k}^{(\mathrm{eee})} + \mathbf{P}_{\omega_k}^{(\mathrm{eeem})},$$

where

$$\mathbf{P}_{\omega_k}^{(\mathrm{ee})} = \epsilon_0 \boldsymbol{\chi}^{(\mathrm{ee})}:\mathbf{E}_{\omega_k},$$

$$\mathbf{P}_{\omega_k}^{(\mathrm{eem})} = \epsilon_0 \boldsymbol{\chi}^{(\mathrm{eem})}:\mathbf{E}_{\omega_k}\mathbf{H}_0$$

are the linear polarization densities and their modifications brought about by the static magnetic field,[3,4] respectively, and

$$\mathbf{P}_{\omega_k}^{(\mathrm{eee})} = \epsilon_0 \boldsymbol{\chi}^{(\mathrm{eee})}:(\mathbf{E}\mathbf{E})_{\omega_k},$$

$$\mathbf{P}_{\omega_k}^{(\mathrm{eeem})} = \epsilon_0 \boldsymbol{\chi}^{(\mathrm{eeem})}:(\mathbf{E}\mathbf{E})_{\omega_k}\mathbf{H}_0$$

are the nonlinear source terms that govern optical parametric generation[5] and magneto-optical parametric generation,[4] respectively. $(\mathbf{E}\mathbf{E})_{\omega_k}$ refers to the particular combination of two electrical fields that give rise to a field at angular frequency ω_k. Each susceptibility tensor is to be taken with respect to the particular frequency combination that follows it.

We have chosen to analyze wave propagation in the (111) direction of a crystal of point-symmetry class $\bar{4}3m$, with this direction as the z axis of the laboratory frame (x, y, z). In the Faraday configuration, with $\mathbf{H}_0 = H_0{}^z\mathbf{e}_z$, we then have that no linear direction of polarization has preference, and the symmetry of the combined system, crystal plus light, must be preserved about the direction of propagation. We introduce the circularly polarized basis $\mathbf{e}_\pm = (\mathbf{e}_x \pm i\mathbf{e}_y)/\sqrt{2}$ and separate the electrical fields into circularly polarized components $E_{\omega_k}^{\pm} = \mathbf{e}_\pm^* \cdot \mathbf{E}_{\omega_k}$, where a plus denotes left-circular polarization (LCP) and a minus denotes right-circular polarization (RCP).

Projecting the polarization density onto the circularly polarized basis $P_{\omega_k}^{\pm} = \mathbf{e}_\pm^* \cdot \mathbf{P}_{\omega_k}$ then gives

$$P_{\omega_1}^{\pm} = \epsilon_0[(n_1^2 - 1 \pm \gamma_1)E_{\omega_1}^{\pm} + \sqrt{2}(1 \pm i)(p_1 \pm q_1)E_{\omega_3}^{\mp}E_{\omega_2}^{\pm *}],$$

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$$P^{\pm}_{\omega_2} = \epsilon_0[(n_2^2 - 1 \pm \gamma_2)E^{\pm}_{\omega_2} + \sqrt{2}(1 \pm i)(p_2 \pm q_2)E^{\mp}_{\omega_3}E^{\pm *}_{\omega_1}],$$

$$P^{\pm}_{\omega_3} = \epsilon_0[(n_3^2 - 1 \pm \gamma_3)E^{\pm}_{\omega_3} + \sqrt{2}(1 \pm i)(p_3 \pm q_3)E^{\mp}_{\omega_1}E^{\mp}_{\omega_2}],$$

with

$$n_k^2 = 1 + \chi^{(\mathrm{ee})}_{xx}(-\omega_k;\omega_k),$$

$$\gamma_k = i\chi^{(\mathrm{eem})}_{xyz}(-\omega_k;\omega_k,0)H_0^z,$$

$$p_{1,2} = \chi^{(\mathrm{eee})}_{xxx}(-\omega_{1,2};\omega_3,-\omega_{2,1}),$$

$$p_3 = \chi^{(\mathrm{eee})}_{xxx}(-\omega_3;\omega_1,\omega_2),$$

$$q_{1,2} = -i\chi^{(\mathrm{eeem})}_{xxxz}(-\omega_{1,2};\omega_3,-\omega_{2,1},0)H_0^z,$$

$$q_3 = -i\chi^{(\mathrm{eeem})}_{xxxz}(-\omega_3;\omega_1,\omega_2,0)H_0^z.$$

In terms of the susceptibility elements taken in the crystal frame, we have $\chi^{(\mathrm{eee})}_{xxx} = \chi^{(\mathrm{eee})}_{XYZ}/\sqrt{3}$ and $\chi^{(\mathrm{eeem})}_{xxxz} = [\chi^{(\mathrm{eeem})}_{XXYY} + \chi^{(\mathrm{eeem})}_{XYXY} - \chi^{(\mathrm{eeem})}_{XYYX}]/(2\sqrt{3})$, where lowercase (capital) lettering in subscripts denotes tensor components taken in the laboratory (crystal) reference frame.

By using the ansatz

$$E^{\pm}_{\omega_k} = \sqrt{C_k^{\pm}}\,A^{\pm}_{\omega_k}\exp[i(\beta_k \pm \alpha_k)z], \qquad k = 1,2,3,$$

with

$$C_k^{\pm} = \eta_k(1 \pm i)(1 \pm \delta_k)/\sqrt{2}, \qquad \eta_k = (\omega_k p_k)/(n_k c),$$

$$\beta_k = \omega_k n_k/c, \qquad \alpha_k = \omega_k\gamma_k/(2n_k c), \qquad \delta_k = q_k/p_k,$$

and employing the slowly varying envelope approximation, with the assumption of a nondepleted pump $A^{\pm}_{\omega_3}(z) = A^{\pm}_{\omega_3}(0) = \text{constant}$, one obtains Eq. (1) as

$$\frac{\partial A^{\pm}_{\omega_1}}{\partial z} = i\kappa_{\pm}A^{\mp}_{\omega_3}(0)A^{\pm *}_{\omega_2}\exp[i(\Delta\beta \mp \Delta\alpha)z], \qquad (2a)$$

$$\frac{\partial A^{\pm}_{\omega_2}}{\partial z} = i\kappa_{\pm}A^{\mp}_{\omega_3}(0)A^{\pm *}_{\omega_1}\exp[i(\Delta\beta \mp \Delta\alpha)z], \qquad (2b)$$

where

$$\Delta\beta = \beta_3 - \beta_2 - \beta_1, \qquad \Delta\alpha = \alpha_1 + \alpha_2 + \alpha_3,$$

$$\kappa_{\pm} = (C_1^{\pm}C_2^{\pm *}C_3^{\mp})^{1/2}.$$

In the following treatment, without loss of generality, we assume $\kappa_{\pm}A^{\mp}_{\omega_3}(0)$ to be real. On eliminating any explicit z dependence, by taking new variables $a^{\pm}_{\omega_k}$ such that

$$a^{\pm}_{\omega_k} = A^{\pm}_{\omega_k}\exp[-i(\Delta\beta \mp \Delta\alpha)z/2], \qquad k = 1,2,$$

from Eqs. (2) we obtain the set of decoupled equations of evolution for the envelopes of the idler and the signal as

$$\frac{\partial^2 a^{\pm}_{\omega_k}}{\partial z^2} = [\kappa_{\pm}^2 A^{\mp 2}_{\omega_3}(0) - (\Delta\beta \mp \Delta\alpha)^2/4]a^{\pm}_{\omega_k}, \qquad k = 1,2,$$

whose solutions can easily be obtained as

$$a^{\pm}_{\omega_1} = a^{\pm}_{\omega_1}(0)\left[\cosh(K_{\pm}z) - i\frac{(\Delta\beta \mp \Delta\alpha)}{2K_{\pm}}\sinh(K_{\pm}z)\right] + ia^{\pm *}_{\omega_2}(0)\frac{\kappa_{\pm}A^{\mp}_{\omega_3}(0)}{K_{\pm}}\sinh(K_{\pm}z), \qquad (3a)$$

$$a^{\pm}_{\omega_2} = a^{\pm}_{\omega_2}(0)\left[\cosh(K_{\pm}z) - i\frac{(\Delta\beta \mp \Delta\alpha)}{2K_{\pm}}\sinh(K_{\pm}z)\right] + ia^{\pm *}_{\omega_1}(0)\frac{\kappa_{\pm}A^{\mp}_{\omega_3}(0)}{K_{\pm}}\sinh(K_{\pm}z), \qquad (3b)$$

where $K_{\pm} = [\kappa_{\pm}^2 A^{\mp 2}_{\omega_3}(0) - (\Delta\beta \mp \Delta\alpha)^2/4]^{1/2}$.

We concentrate our attention on the case of optical parametric amplification and assume a zero idler at the input, or $a^{\pm}_{\omega_1}(0) = 0$. The total solution for the propagating light is conveniently expressed in terms of Stokes parameters,[6] which for the idler are taken as

$$S_0^{(i)} = |E^{+}_{\omega_1}|^2 + |E^{-}_{\omega_1}|^2, \qquad S_1^{(i)} = 2\,\mathrm{Re}(E^{+*}_{\omega_1}E^{-}_{\omega_1}),$$

$$S_3^{(i)} = |E^{+}_{\omega_1}|^2 - |E^{-}_{\omega_1}|^2, \qquad S_2^{(i)} = 2\,\mathrm{Im}(E^{+*}_{\omega_1}E^{-}_{\omega_1}).$$

We describe the signal and the pump similarly by pairwise replacing $[(i), \omega_1]$ with $[(s), \omega_2]$ and $[(p), \omega_3]$, respectively. We also define the parameter $\nu = \eta_1/\eta_2$, the gain $g = [\eta_1\eta_2 S_0^{(p)}/2]^{1/2}$, and the dimensionless propagation coordinate $\zeta = gz$, normalized to the pump intensity. With these transformations, Eqs. (3) for the idler become

$$\frac{S_0^{(i)}(\zeta)}{S_0^{(s)}(0)} = \frac{\nu}{2}[f_+(\zeta) + f_-(\zeta)] + \frac{\nu}{2}[f_+(\zeta) - f_-(\zeta)]\epsilon_s(0), \qquad (4a)$$

$$\frac{S_1^{(i)}(\zeta)}{S_0^{(i)}(\zeta)} = [1 - \epsilon_i^2(\zeta)]^{1/2}\cos[\arg(E^{-}_{\omega_1}) - \arg(E^{+}_{\omega_1})], \qquad (4b)$$

$$\frac{S_2^{(i)}(\zeta)}{S_0^{(i)}(\zeta)} = [1 - \epsilon_i^2(\zeta)]^{1/2}\sin[\arg(E^{-}_{\omega_1}) - \arg(E^{+}_{\omega_1})], \qquad (4c)$$

$$\epsilon_i(\zeta) = \frac{[f_+(\zeta) - f_-(\zeta)] + [f_+(\zeta) + f_-(\zeta)]\epsilon_s(0)}{[f_+(\zeta) + f_-(\zeta)] + [f_+(\zeta) - f_-(\zeta)]\epsilon_s(0)}, \qquad (4d)$$

where $\epsilon_k(\zeta) = S_3^{(k)}(\zeta)/S_0^{(k)}(\zeta)$, $k = i, s, p$ are the normalized ellipticities of the polarization states of idler, signal, and pump, respectively, with $\epsilon_k = 1$ for LCP and $\epsilon_k = -1$ for RCP and where we have defined

$$f_{\pm}(\zeta) = \frac{(1 \pm \delta_1)^2[1 \mp \epsilon_p(0)]}{(\xi_{\pm}^2 - \phi_{\pm}^2)}\sinh^2[(\xi_{\pm}^2 - \phi_{\pm}^2)^{1/2}\zeta],$$

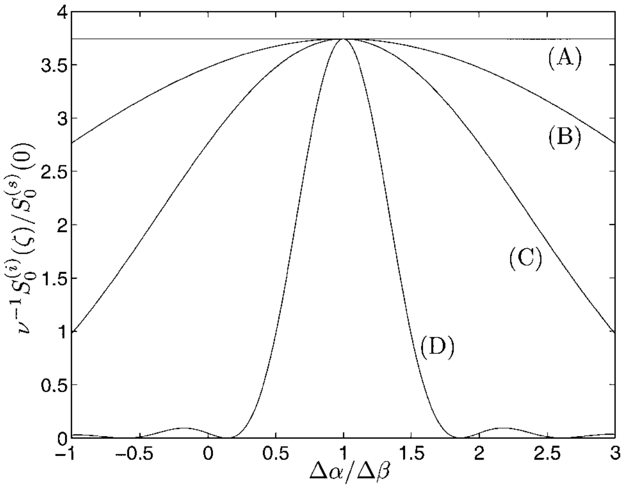

Fig. 1. Conversion efficiency $\nu^{-1}S_0^{(i)}(\zeta)/S_0^{(s)}$ versus differential phase mismatch $\Delta\alpha/\Delta\beta$ for a right-circularly polarized input pump and a left-circularly polarized signal, $\epsilon_p(0) = -1$, $\epsilon_s(0) = 1$, in the case with negligible magneto-optically induced parametric generation, $\delta_1 = \delta_2 = 0$. Parameter values are $\zeta = 1$ and (A), $\phi = 0$, (B) $\phi = 0.5$, (C) $\phi = 1$, and (D) $\phi = 4$.

$$\xi_\pm^2 = (1 \pm \delta_1)(1 \pm \delta_2)[1 \mp \epsilon_p(0)],$$

$$\phi_\pm = (1 \mp \Delta\alpha/\Delta\beta)\phi\,,$$

with $\phi = \Delta\beta/(2g)$ being the normalized phase mismatch. In the notation above, $\Delta\alpha/\Delta\beta$ has the role of a differential phase mismatch between LCP and RCP that originates from the Faraday effect.

By direct inspection, from Eqs. (3) we have

$$\arg(E_{\omega_1}^-) - \arg(E_{\omega_1}^+) = \varphi_0 + \varphi_1 + \varphi_2(\zeta)\,,$$

where $\varphi_0 = \arg[E_{\omega_2}^+(0)] - \arg[E_{\omega_2}^-(0)]$ determines the orientation of the polarization ellipse of the input signal and

$$\varphi_1 = (\pi/2)\{\mathrm{sgn}[(1 + \delta_1)(1 + \delta_2)] - \mathrm{sgn}[(1 - \delta_1)(1 - \delta_2)] - 1\},$$

$$\varphi_2(\zeta) = g^{-1}(\alpha_3 + \alpha_2 - \alpha_1)\zeta\,,$$

where $\varphi_2(\zeta)$ governs the rotation of the polarization ellipse of the idler. The above expressions completely characterize the fields in terms of magnitude, phase, and polarization state after interaction over a distance z and given initial conditions for the input beams.

Certain conclusions can immediately be drawn from Eqs. (4). The idler intensity $S_0^{(i)}(\zeta)$ varies linearly with $S_0^{(s)}(0)$ and $\epsilon_s(0)$, the input signal intensity and ellipticity, respectively. From Eq. (4a) we obtain the selection rule that, with a RCP (LCP) pump, the intensity of the idler will be nonzero only for a signal that has a nonzero LCP (RCP) component. In particular, from Eq. (4d), the generated idler will then be in a pure LCP (RCP) state, and phase matching will be obtained for $\Delta\alpha = \Delta\beta$ ($\Delta\alpha = -\Delta\beta$).

With zero magnetic field $\phi_+ = \phi_-$, and for a linearly polarized pump $\epsilon_p(0) = 0$, we have $f_+(\zeta) = f_-(\zeta)$, so the idler intensity and the ellipticity, from Eqs. (4a) and (4d), become independent of the signal ellipticity and intensity, respectively.

The energy exchange rate between the beams can be obtained from $\partial W/\partial t = (1/2)\sum_k \omega_k \,\mathrm{Im}(\mathbf{E}_{\omega_k}^* \cdot \mathbf{P}_{\omega_k})$, from which Manley–Rowe-type relations can be derived, reflecting conservation of momentum (Poynting vector) and angular momentum (symmetry of Maxwell's electromagnetic stress tensor) of the electromagnetic field.[7] For the classic phase-matched case $\Delta\beta = 0$, reflecting conservation of photon momentum, for global phase matching of the parametric process considered here, one must have in addition $\Delta\alpha = 0$, reflecting conservation of the additional angular momentum of the electromagnetic wave imposed by the gyrotropy.[8]

From solutions (4) we single out the simplified case of a setup with a RCP pump and a LCP signal, for which Fig. 1 shows the conversion efficiency versus differential phase mismatch $\Delta\alpha/\Delta\beta$ in the case of negligible magneto-optical parametric generation. For typical parameters $z \sim 10^{-2}\ m$, $n_k \sim 1$, $\omega_k/c \sim 10^7\ \mathrm{m}^{-1}$, and $p_k \sim 5 \times 10^{-12}\ \mathrm{mV}^{-1}$, a value of $\zeta = 1$, as used in Fig. 1, hence corresponds to a pump intensity of $I_p \approx 1.1 \times 10^{10}\ \mathrm{W/m}^2$. Interest in such a configuration is ultimately stimulated by the possibility of using the circular birefringence induced through the Faraday effect to compensate for the dispersion mismatch. This circular birefringence was measured for several materials and expressed in terms of the Verdet constant, defined by $\vartheta_F = VH_0^z L$, where ϑ_F is the rotation angle and L is the propagation length. From the definition of $\vartheta_F = (\omega L/2c)(n_- - n_+)$, where n_+ and n_- are the refractive indices for LCP and RCP, respectively, one may obtain values of α_k. There are, however, neither measured nor estimated values for the coefficients q_k related to $\boldsymbol{\chi}^{(\mathrm{eeem})}$. By referring to their quantum-mechanical expressions[4,9] and using dimensionality arguments to compare with $\boldsymbol{\chi}^{(\mathrm{eee})}$, which is known for several materials from second-harmonic generation data, one obtains $\delta_k = q_k/p_k \approx 10^{-4}$ for a magnetic field of 1 T and a Verdet constant $V = 20\ \mathrm{rad\ T}^{-1}\ \mathrm{m}^{-1}$, far away from any resonance.

F. Jonsson's e-mail address is fj@optics.kth.se.

References

1. H. Rabin and P. P. Bey, Phys. Rev. **156,** 1010 (1967).
2. P. P. Bey and H. Rabin, Phys. Rev. **162,** 794 (1967).
3. L. D. Landau, E. M. Lifshitz, and L. P. Pitaevskiĭ, *Electrodynamics of Continuous Media*, 2nd ed. (Butterworth & Heinemann, Oxford, 1984).
4. S. Kielich and R. Zawodny, Acta Phys. Pol. A **43,** 579 (1973).
5. P. N. Butcher and D. Cotter, *The Elements of Nonlinear Optics* (Cambridge U. Press, New York, 1990).
6. J. D. Jackson, *Classical Electrodynamics*, 2nd ed. (Wiley, New York, 1975).
7. N. Bloembergen, in *Polarisation, Matiere et Rayonnement*, C. Cohen-Tannoudji, ed. (Presses Universitaires, Paris, 1969), pp. 189–199.
8. A. N. Godlevskaya and V. N. Kapshai, Opt. Spectrosc. (USSR) **66,** 486 (1989).
9. G. Wagnière, J. Chem. Phys. **77,** 2786 (1982).