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Polarization state dependence of optical parametric processes in artificially gyrotropic media

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Abstract. The theory of parametric generation and amplification in artificially gyrotropic (magneto-optic) media is presented. Complete solutions for signal and idler are given in terms of Stokes' parameters, and polarization state dependences of applied static magnetic field strength are discussed in terms of trajectories of the reduced Stokes vector on the Poincaré sphere.

Keywords: Magneto-optical devices, parametric amplification, nonlinear magneto-optics

1. Introduction

Three-wave coherent interactions of the type $\omega_1 + \omega_2 = \omega_3$ underly some of the most efficient nonlinear optical processes with important applications such as optical parametric generation (OPG) and amplification, which in a confined geometry lead to the onset of optical parametric oscillation. Until now these processes have been almost exclusively studied in linearly birefringent (anisotropic) media where phase matching, a necessary prerequisite for the efficiency of these processes, is achieved by exploiting the linear birefringence. For continuous tuning of the phase matching, however, this approach introduces certain complications, related to wave propagation in anisotropic media, the most evident being the walk-off effect [1]. This and other related problems can be bypassed by instead exploiting the circular birefringence induced by an externally applied static magnetic field, which occurs in isotropic media as well. In such media the left- and right-circular polarization state degeneracy is lifted and the corresponding phase velocities differ by an amount that depends on the magnetic field strength, and can hence be used to achieve phase matching still preserving optical isotropy and avoiding walk-off in the so-called Faraday configuration; this is a consequence of the specific impact the magnetic field has on the optical properties of the medium, and is in the linear regime also reflected in the relation [2]

$$\boldsymbol{D}_\omega = \varepsilon_0(\varepsilon \boldsymbol{E}_\omega + \mathrm{i}\boldsymbol{E}_\omega \times \boldsymbol{g}),$$

between the electric field $\boldsymbol{E}(\boldsymbol{r}, t) = \mathrm{Re}\,[\boldsymbol{E}_\omega \mathrm{e}^{-\mathrm{i}\omega t}]$ and induction $\boldsymbol{D}(\boldsymbol{r}, t) = \mathrm{Re}\,[\boldsymbol{D}_\omega \mathrm{e}^{-\mathrm{i}\omega t}]$, with ε being the dielectric constant (electrical permittivity), a scalar for isotropic media, and $\boldsymbol{g}$ the gyration vector which conveys the influence of the magnetic field. In addition, the magnetic field introduces new features related to the breakdown of the time-reversal symmetry, which in the case of confined geometry, such as in a cavity, can lead to strikingly novel behaviour because of the concomitant onset of nonreciprocity. In particular, this leads to the development of nonreciprocal nonlinear optical devices with applications for unidirectional control or shielding, such as in the case of polarization state controlled switching [3]. Furthermore, in this case the conservation of angular momentum of the light beams, together with the crystalline lattice, electronic spins and orbitals, must be inserted along with the photon energy and momentum conservation laws, as described by the well known Manley–Rowe relations for three-wave interactions.

2. The polarization density

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

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

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

$$\begin{aligned}\boldsymbol{P}_{\omega_k} = \varepsilon_0[&\boldsymbol{\chi}^{(ee)} : \boldsymbol{E}_{\omega_k} + \boldsymbol{\chi}^{(eem)} : \boldsymbol{E}_{\omega_k}\boldsymbol{H}_0 \\ &+ \boldsymbol{\chi}^{(eee)} : (\boldsymbol{E}\boldsymbol{E})_{\omega_k} + \boldsymbol{\chi}^{(eeem)} : (\boldsymbol{E}\boldsymbol{E})_{\omega_k}\boldsymbol{H}_0],\end{aligned}$$

where $\boldsymbol{\chi}^{(ee)}$ and $\boldsymbol{\chi}^{(eee)}$ are the first- and second-order optical susceptibility tensors; $\boldsymbol{\chi}^{(eem)}$ and $\boldsymbol{\chi}^{(eeem)}$ are the second- and third-order magneto-optical susceptibility tensors, governing the Faraday effect [2] and magneto-optical parametric generation (magnetic field induced OPG) [4,5], respectively.

Above, the notation $(\boldsymbol{EE})_{\omega_k}$ refers to the combination of two electric fields that give rise to a field at angular frequency ω_k, and all susceptibility tensors are to be taken with respect to the particular frequency combination following them.

In order to clarify the main features imposed by the gyrotropy, we have chosen to analyse 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 coordinate frame (x, y, z). For this point-symmetry class, the tensors $\boldsymbol{\chi}^{(ee)}$ and $\boldsymbol{\chi}^{(eem)}$ are isotropic (invariant under rotation of the crystal frame relative to the laboratory frame), and the nonzero elements of $\boldsymbol{\chi}^{(eee)}$ and $\boldsymbol{\chi}^{(eeem)}$, taken in the laboratory frame, are listed in the appendix, tables A.1 and A.2. In the Faraday configuration, with $\boldsymbol{H}_0 = H_0^z \boldsymbol{e}_z$, we then have rotational symmetry of the polarization density around the z-axis, and the symmetry of the combined system, crystal plus light, must be preserved around the direction of propagation.

By projecting the polarization density onto the circularly polarized basis $\boldsymbol{e}_\pm = (\boldsymbol{e}_x \pm \mathrm{i}\boldsymbol{e}_y)/\sqrt{2}$, where a plus denotes left-circular polarization (LCP) and a minus right-circular polarization (RCP), one obtains the LCP and RCP components $P^\pm_{\omega_k} = \boldsymbol{e}^*_\pm \cdot \boldsymbol{P}_{\omega_k}$ of the polarization density expressed in terms of circularly polarized electric field components $E^\pm_{\omega_k} = \boldsymbol{e}^*_\pm \cdot \boldsymbol{E}_{\omega_k}$ as [5]

$$
\begin{aligned}
P^\pm_{\omega_1} &= \varepsilon_0[(n_1^2 - 1 \pm \gamma_1)E^\pm_{\omega_1} \\
&\quad + \sqrt{2}(1 \pm \mathrm{i})(p_1 \pm q_1)E^\mp_{\omega_3}E^{\pm *}_{\omega_2}], \\
P^\pm_{\omega_2} &= \varepsilon_0[(n_2^2 - 1 \pm \gamma_2)E^\pm_{\omega_2} \\
&\quad + \sqrt{2}(1 \pm \mathrm{i})(p_2 \pm q_2)E^\mp_{\omega_3}E^{\pm *}_{\omega_1}], \\
P^\pm_{\omega_3} &= \varepsilon_0[(n_3^2 - 1 \pm \gamma_3)E^\pm_{\omega_3} \\
&\quad + \sqrt{2}(1 \pm \mathrm{i})(p_3 \pm q_3)E^\mp_{\omega_1}E^\mp_{\omega_2}],
\end{aligned}
$$

where we defined

$$
\begin{aligned}
n_k^2 &= 1 + \chi^{(ee)}_{xx}(-\omega_k; \omega_k), \\
\gamma_k &= \mathrm{i}\chi^{(eem)}_{xyz}(-\omega_k; \omega_k, 0)H_0^z, \\
p_{1,2} &= \chi^{(eee)}_{xxx}(-\omega_{1,2}; \omega_3, -\omega_{2,1}), \\
p_3 &= \chi^{(eee)}_{xxx}(-\omega_3; \omega_1, \omega_2), \\
q_{1,2} &= -\mathrm{i}\chi^{(eeem)}_{xxxz}(-\omega_{1,2}; \omega_3, -\omega_{2,1}, 0)H_0^z, \\
q_3 &= -\mathrm{i}\chi^{(eeem)}_{xxxz}(-\omega_3; \omega_1, \omega_2, 0)H_0^z.
\end{aligned}
$$

3. Wave propagation

The solution for the propagating light is conveniently expressed in terms of Stokes' parameters [6], which for the idler are taken as

$$
S_0^{(i)} = |E^+_{\omega_1}|^2 + |E^-_{\omega_1}|^2, \qquad S_1^{(i)} = 2\mathrm{Re}\,[E^{+*}_{\omega_1}E^-_{\omega_1}],
$$
$$
S_3^{(i)} = |E^+_{\omega_1}|^2 - |E^-_{\omega_1}|^2, \qquad S_2^{(i)} = 2\mathrm{Im}\,[E^{+*}_{\omega_1}E^-_{\omega_1}].
$$

The signal and pump are similarly described by pairwise replacement of $\{(i), \omega_1\}$ with $\{(s), \omega_2\}$ and $\{(p), \omega_3\}$, respectively. We also define the parameters $\beta_k = \omega_k n_k/c$, $\alpha_k = \omega_k\gamma_k/(2n_kc)$, $\delta_k = q_k/p_k$, $\nu = \eta_1/\eta_2$, and the gain $g = [\eta_1\eta_2 S_0^{(p)}/2]^{1/2}$, where $\eta_k = \omega_k p_k/(n_kc)$, and the dimensionless propagation coordinate $\zeta = gz$. Here β_k, $k = 1, 2, 3$, are the respective regular propagation constants of the idler, signal, and pump waves with the static magnetic field switched off, while α_k are their magnetic field induced additional contributions, governing the circular birefringence (Faraday rotation). The quotes δ_k describe the strengths of the static magnetic field induced parametric interactions relative to the all-optically induced ones.

For simplicity, we make a restriction to the case of optical parametric amplification (OPA), with a zero idler present at $\zeta = 0$. By employing the slowly varying envelope and nondepleted pump approximation, one then obtains solutions to the wave equation (1) in terms of hyperbolic functions [5]. In terms of Stokes' parameters, the solutions for the idler and signal can be written as

$$
\begin{aligned}
S_0^{(i)}(\zeta)/S_0^{(s)}(0) &= (\nu/2)[f_1^+(\zeta) + f_1^-(\zeta)] \\
&\quad + (\nu/2)[f_1^+(\zeta) - f_1^-(\zeta)]\epsilon_s(0), &(2a)
\end{aligned}
$$

$$
S_1^{(i)}(\zeta)/S_0^{(i)}(\zeta) = [1 - \epsilon_i^2(\zeta)]^{1/2}\cos[\vartheta_i(\zeta)], \qquad (2b)
$$
$$
S_2^{(i)}(\zeta)/S_0^{(i)}(\zeta) = [1 - \epsilon_i^2(\zeta)]^{1/2}\sin[\vartheta_i(\zeta)], \qquad (2c)
$$
$$
\epsilon_i(\zeta) = \frac{[f_1^+(\zeta) - f_1^-(\zeta)] + [f_1^+(\zeta) + f_1^-(\zeta)]\epsilon_s(0)}{[f_1^+(\zeta) + f_1^-(\zeta)] + [f_1^+(\zeta) - f_1^-(\zeta)]\epsilon_s(0)}, \qquad (2d)
$$

and

$$
\begin{aligned}
S_0^{(s)}(\zeta)/S_0^{(s)}(0) &= [f_2^+(\zeta) + f_2^-(\zeta)]/2 \\
&\quad + [f_2^+(\zeta) - f_2^-(\zeta)]\epsilon_s(0)/2, &(3a)
\end{aligned}
$$

$$
S_1^{(s)}(\zeta)/S_0^{(s)}(\zeta) = [1 - \epsilon_s^2(\zeta)]^{1/2}\cos[\vartheta_s(\zeta)], \qquad (3b)
$$
$$
S_2^{(s)}(\zeta)/S_0^{(s)}(\zeta) = [1 - \epsilon_s^2(\zeta)]^{1/2}\sin[\vartheta_s(\zeta)], \qquad (3c)
$$
$$
\epsilon_s(\zeta) = \frac{[f_2^+(\zeta) - f_2^-(\zeta)] + [f_2^+(\zeta) + f_2^-(\zeta)]\epsilon_s(0)}{[f_2^+(\zeta) + f_2^-(\zeta)] + [f_2^+(\zeta) - f_2^-(\zeta)]\epsilon_s(0)}, \qquad (3d)
$$

respectively, 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 the idler, signal and pump, respectively, and where we defined

$$
f_1^\pm(\zeta) = (1 \pm \delta_1)^2[1 \mp \epsilon_p(0)]\frac{\sinh^2[(\xi_\pm^2 - \phi_\pm^2)^{1/2}\zeta]}{(\xi_\pm^2 - \phi_\pm^2)},
$$
$$
f_2^\pm(\zeta) = \cosh^2[(\xi_\pm^2 - \phi_\pm^2)^{1/2}\zeta] + \phi_\pm^2\frac{\sinh^2[(\xi_\pm^2 - \phi_\pm^2)^{1/2}\zeta]}{(\xi_\pm^2 - \phi_\pm^2)},
$$
$$
\xi_\pm^2 = (1 \pm \delta_1)(1 \pm \delta_2)[1 \mp \epsilon_p(0)],
$$
$$
\phi_\pm = (1 \mp \Delta\alpha/\Delta\beta)\phi,
$$
$$
\vartheta_i = g^{-1}(\alpha_3 + \alpha_2 - \alpha_1)\zeta - \vartheta_s(0) - \pi/2,
$$

$$
\begin{aligned}
\vartheta_s &= \vartheta_s(0) + g^{-1}(\alpha_3 - \alpha_2 + \alpha_1)\zeta \\
&\quad + \arctan\left(\frac{\phi_-}{(\xi_-^2 - \phi_-^2)^{1/2}}\tanh[(\xi_-^2 - \phi_-^2)^{1/2}\zeta]\right) \\
&\quad - \arctan\left(\frac{\phi_+}{(\xi_+^2 - \phi_+^2)^{1/2}}\tanh[(\xi_+^2 - \phi_+^2)^{1/2}\zeta]\right),
\end{aligned}
$$

with $\Delta\beta = \beta_3 - \beta_2 - \beta_1$, $\Delta\alpha = \alpha_1 + \alpha_2 + \alpha_3$; $\phi = \Delta\beta/(2g)$ is the normalized phase-mismatch in the limit of zero-applied static magnetic field, and the quote $\Delta\alpha/\Delta\beta$ has the role of a differential phase mismatch between LCP and RCP, originating from the Faraday effect. In deriving equations (2) and (3), without loss of generality, we assumed $\delta_k^2 < 1$, $k = 1, 2, 3$. The orientation of the polarization ellipses of the fields are determined by ϑ_i and ϑ_s, being twice the angles between the main axes of the polarization ellipses of idler and signal relative to the x-axis of the laboratory frame, where $\vartheta_s(0) = \arg[E^+_{\omega_2}(0)] - \arg[E^-_{\omega_2}(0)]$ is determined by the orientation of the polarization ellipse of the input signal.

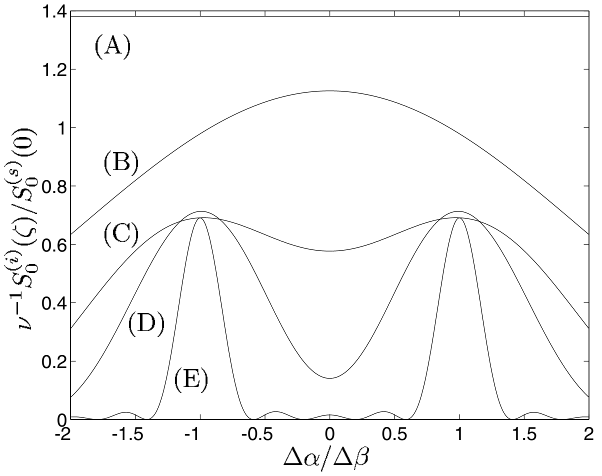

Figure 1. Conversion efficiency $S_0^{(i)}(\zeta)/(\nu S_0^{(s)}(0))$ versus differential phase mismatch $\Delta\alpha/\Delta\beta$, for a linearly polarized input pump and signal, $\epsilon_s(0) = \epsilon_p(0) = 0$, in the case with negligible magneto-optically induced parametric generation. Employed parameter values are $\zeta = 1$, $\delta_1 = \delta_2 = 0$, and (A) $\phi = 0$, (B) $\phi = 0.8$, (C) $\phi = 1.6$, (D) $\phi = 2.4$, (E) $\phi = 8.0$.

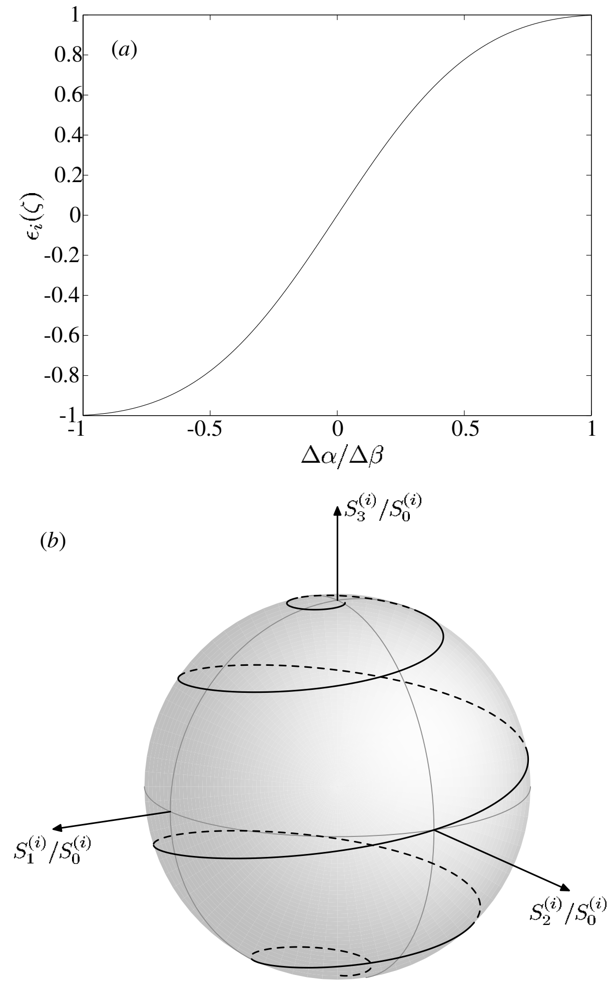

Figure 2. (*a*) Ellipticity $\epsilon_i(\zeta) = S_3^{(i)}(\zeta)/S_0^{(i)}(\zeta)$ of idler versus applied magnetic field normalized in terms of differential phase mismatch $\Delta\alpha/\Delta\beta$, and (*b*) Poincaré map of transmitted idler polarization state $(s_1^{(i)}, s_2^{(i)}, s_3^{(i)})$, $s_k^{(i)} = S_k^{(i)}(\zeta)/S_0^{(i)}(\zeta)$, as function of applied magnetic field in interval corresponding to $-1 \leqslant \Delta\alpha/\Delta\beta \leqslant 1$, for a linearly polarized input pump (RCP) and signal (LCP), $S_3^{(p)}(0) = S_3^{(s)}(0) = 0$, in the case with negligible magneto-optically induced parametric generation, $\delta_1 = \delta_2 = 0$. Employed parameter values are $\zeta = 1$ and $\phi = 1.6$.

4. Discussion

From equation (2*a*), one obtains the selection rule that with a RCP (LCP) pump, the intensity of the idler will be nonzero only for a signal having a nonzero LCP (RCP) component. The generated idler will then be in a pure LCP (RCP) state, and phase matching is obtained for $\Delta\alpha = \Delta\beta$ ($\Delta\alpha = -\Delta\beta$). This selection rule is a direct consequence of the fact that for each photon carrying angular momentum $+\hbar$ (LCP) added to the signal wave (ω_2), one photon with angular momentum $+\hbar$ (LCP) is added to the idler wave (ω_1), and one photon with angular momentum $-\hbar$ (RCP) is removed from the pump wave (ω_3). The change of field angular momentum of $3\hbar$ is compensated by a change of angular momentum of the crystal lattice of the medium, i.e. as a steady state torque on the crystal†. A similar scheme applies to the case of second-harmonic generation with circularly polarized fields [8], where two circularly polarized fundamental quanta combine to a second-harmonic quantum with opposite sense of circular polarization.

For a linearly polarized pump and signal, the solutions for $S_0^{(i)}$ and $S_0^{(s)}$ become symmetric with respect to the magnetic field, while $S_3^{(i)}$ and $S_3^{(s)}$ become antisymmetric. The nonlinear source terms related to magnetic field induced OPG, described by the quotes δ_k, are generally small compared with the all-optical ones in optically nonresonant frequency regimes. For simplicity, these terms can therefore be neglected for most practical purposes without any greater loss of generality.

In figure 1 the signal-to-idler conversion efficiency for a linearly polarized pump and signal is shown as a function of applied static magnetic field, expressed in terms of differential phase mismatch $\Delta\alpha/\Delta\beta$, for certain values of ϕ, the phase mismatch in the limit of zero static magnetic field. For small values of ϕ, the maximum signal-to-idler conversion efficiency is obtained for zero magnetic field. This corresponds to cases where the classical photon momentum conservation requirement $\beta_1 + \beta_2 = \beta_3$ holds to a good degree. However, as ϕ is increased, the point for maximum conversion efficiency bifurcates into two, corresponding to phase matching for each of the circularly polarized eigenmodes. For high values of ϕ, one may always obtain phase matching in the vicinity of $\Delta\alpha/\Delta\beta = \pm 1$ for either set $(E^{\pm}_{\omega_1}, E^{\pm}_{\omega_2}, E^{\mp}_{\omega_3})$ of the circularly polarized fields, though for a linearly polarized pump essentially one half of the pump energy is lost in the mismatched mode since phase matching cannot be simultaneously achieved for both LCP and RCP.

Typical dependences of polarization states on magnetic field, corresponding to the curves in figure 1, are shown in figure 2. In figure 2(*a*), the ellipticity of the idler wave is

† For discussions on conservation of photon angular momentum in dielectrica, see [7].

shown as a function of applied magnetic field in an interval corresponding to $-1 \leqslant \Delta\alpha/\Delta\beta \leqslant 1$.

The overall behaviour of the polarization state of the idler is conveniently displayed by mapping the trajectory described by the normalized reduced Stokes vector $(S_1^{(i)}, S_2^{(i)}, S_3^{(i)})/S_0^{(i)}$ which, since $S_0^2 = S_1^2 + S_2^2 + S_3^2$ for monochromatic light, is confined to the unitary Poincaré sphere. In figure 2(b), the Poincaré map corresponding to figure 2(a) is shown, for input signal being linearly polarized along the y-axis of the laboratory frame, $\vartheta_s(0) = \pi$. Figure 2(b) shows that for a zero magnetic field the generated idler will be linearly polarized as well. As the magnetic field is varied, the trajectory of the Stokes vector will describe helix-like paths towards either the LCP or RCP poles of the Poincaré sphere as $\Delta\alpha/\Delta\beta$ approach the phase matched special cases of $\Delta\alpha/\Delta\beta = \pm 1$. The pitch of the described helix will increase with decreasing $\partial\vartheta_i/\partial\zeta$.

Allowing for a depleted pump, the Manley–Rowe relations, obtained from $\partial W/\partial t = (\frac{1}{2})\sum_k \omega_k \mathrm{Im}\,[\boldsymbol{E}^*_{\omega_k} \cdot \boldsymbol{P}_{\omega_k}]$, reflecting the energy exchange rate between the beams, become

$$\frac{(\partial|E^{\pm}_{\omega_1}|^2/\partial\zeta)}{\eta_1(1 \pm \delta_1)} = \frac{(\partial|E^{\pm}_{\omega_2}|^2/\partial\zeta)}{\eta_2(1 \pm \delta_2)} = -\frac{(\partial|E^{\mp}_{\omega_3}|^2/\partial\zeta)}{\eta_3(1 \mp \delta_3)},$$

being satisfied separately for each set of circularly polarized field components.

5. Summary

In conclusion, we have presented the nonzero elements of the second-order optical and third-order magneto-optical susceptibility tensors, governing optical and magneto-optical parametric generation. The evolution of the signal and generated idler waves were presented in terms of Stokes' parameters, conveniently used for interpretating conversion efficiencies as well as polarization state evolutions. We have discussed the selection rules for the parametric process in terms of transfer of photon angular momentum to the medium. Finally, the general Manley–Rowe relations for this parametric process were presented, reflecting photon number conservation.

The main advantage with the proposed phase-matching technique is that continuous tuning of the phase-matching condition can be obtained in a fixed geometry, in the Faraday configuration, eliminating any walk-off effects. In addition, the magnetic field can be used as a controlling parameter for the polarization states of the idler and signal. For linearly polarized input pump and signal, the sign reversal of the applied magnetic field is shown to cause the energy transfer from the pump to signal and idler to switch from one circularly polarized mode to the orthogonally polarized one.

Appendix

Tables of nonzero elements of the second-order optical and third-order magneto-optical susceptibilities for point-symmetry class $\bar{4}3m$, taken in the laboratory coordinate frame (x, y, z), with the z-axis in the direction of the crystal (111)-direction. In the tables lower-case (capital) lettering in subscripts denotes tensor components taken in the laboratory (crystal) coordinate system.

Table A.1. Nonzero elements of the second-order optical susceptibility $\chi^{(eee)}$ for point-symmetry class $\bar{4}3m$, in the laboratory coordinate frame (x, y, z), with the z-axis in the direction of the crystal (111)-direction.

Nonzero elements[a] of $\chi^{(eee)}_{ijk}$
$\chi^{(eee)}_{xxx} = \chi^{(eee)}_{yyy} = \chi^{(eee)}_{zzz}/2 = a$
$\chi^{(eee)}_{xxy} = \chi^{(eee)}_{yyx} = \chi^{(eee)}_{xyx} = \chi^{(eee)}_{yxy} = \chi^{(eee)}_{xyy} = \chi^{(eee)}_{yxx} = -a$
$\chi^{(eee)}_{xxz} = \chi^{(eee)}_{zxx} = \chi^{(eee)}_{yyz} = \chi^{(eee)}_{zyy} = \chi^{(eee)}_{xzx} = \chi^{(eee)}_{yzy} = -a$
$a = \chi^{(eee)}_{XYZ}/\sqrt{3}$

[a] Calculated using [1, table A3.2].

Table A.2. Nonzero elements of the third-order magneto-optical susceptibility $\chi^{(eeem)}$ for point-symmetry class $\bar{4}3m$, in the laboratory coordinate frame (x, y, z), with the z-axis in the direction of the crystal (111)-direction.

Nonzero elements[a] of $\chi^{(eeem)}_{ijkl}$
$\chi^{(eeem)}_{yxxx} = \chi^{(eeem)}_{zyyy} = -\chi^{(eeem)}_{xyyy} = -\chi^{(eeem)}_{zxxx}$
$= \chi^{(eeem)}_{zyyx} = \chi^{(eeem)}_{zyxy} = -\chi^{(eeem)}_{zxxy} = -\chi^{(eeem)}_{zxyx}$
$= \chi^{(eeem)}_{zxyy} = -\chi^{(eeem)}_{zyxx} = a + b + c,$
$\chi^{(eeem)}_{yyyx} = \chi^{(eeem)}_{xxxz} = -\chi^{(eeem)}_{xxxy} = -\chi^{(eeem)}_{yyyz}$
$= \chi^{(eeem)}_{xxyz} = \chi^{(eeem)}_{xyxz} = -\chi^{(eeem)}_{yyxz} = -\chi^{(eeem)}_{yxyz}$
$= \chi^{(eeem)}_{yxxz} = -\chi^{(eeem)}_{xyyz} = a + b - c,$
$\chi^{(eeem)}_{yyxy} = \chi^{(eeem)}_{xxzx} = -\chi^{(eeem)}_{xxyx} = -\chi^{(eeem)}_{yyzy}$
$= \chi^{(eeem)}_{xxzy} = \chi^{(eeem)}_{xyzx} = -\chi^{(eeem)}_{yyzx} = -\chi^{(eeem)}_{yxzy}$
$= \chi^{(eeem)}_{yxzx} = -\chi^{(eeem)}_{xyzy} = a - b + c,$
$\chi^{(eeem)}_{xyxx} = \chi^{(eeem)}_{yzyy} = -\chi^{(eeem)}_{yxyy} = -\chi^{(eeem)}_{xzxx}$
$= \chi^{(eeem)}_{yzyx} = \chi^{(eeem)}_{yzxy} = -\chi^{(eeem)}_{xzxy} = -\chi^{(eeem)}_{xzyx}$
$= \chi^{(eeem)}_{xzyy} = -\chi^{(eeem)}_{yzxx} = a - b - c,$
$\chi^{(eeem)}_{xzzy} = -\chi^{(eeem)}_{yzzx} = 2(a + b)$, $\chi^{(eeem)}_{zxyz} = -\chi^{(eeem)}_{zyxz} = 2(a - b)$,
$\chi^{(eeem)}_{xzyz} = -\chi^{(eeem)}_{yzxz} = 2(a + c)$, $\chi^{(eeem)}_{zxzy} = -\chi^{(eeem)}_{zyzx} = 2(a - c)$,
$\chi^{(eeem)}_{xyzz} = -\chi^{(eeem)}_{yxzz} = 2(b + c)$, $\chi^{(eeem)}_{zzxy} = -\chi^{(eeem)}_{zzyx} = 2(b - c)$,
$a = \chi^{(eeem)}_{XXYY}/(2\sqrt{3})$, $b = \chi^{(eeem)}_{XYXY}/(2\sqrt{3})$, $c = \chi^{(eeem)}_{XYYX}/(2\sqrt{3})$

[a] Calculated using [4, table 2].

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