2027

Optical Parametric Processes in isotropic third-order nonlinear media with magneto-optic properties

Valerio Pruneri

Optoelectronics Research Centre, Southampton University, Southampton S017 1BJ, U.K.

Fax: 0044 (0)2380 593149, Tel: 0044 (0)2380 593083 , Email: vp3@orc.soton.ac.uk

Stefano Longhi

Istituto Nazionale di Fisica per la Materia, Dipartimento di Fisica and CEQSE-CNR, Politecnico di Milano, Piazza L. da Vinci, 32 I-20133 Milano, Italy

Abstract

Optical parametric interaction in isotropic third-order nonlinear media with magneto-optic properties is investigated. It is shown that new phase-matching conditions with magneto-optic contribution are possible. In particular we study four-wave mixing and electric-field induced three-wave parametric processes in the presence of a magnetic field applied along the direction of propagation of the interacting waves. Control of the new phase-matching branches can be achieved by tuning of the magnetic field.

Submitted to Opt. Lett.
05/2000

In order to achieve efficient nonlinear optical processes it is necessary to ensure that the relative phase mismatch between interacting fields and corresponding nonlinear driving polarizations is set to zero (i.e. the interaction is said to be phase-matched). The material (and waveguide) dispersion, which is responsible for the different phase-velocities of the interacting field and polarization waves in the medium, would prevent cumulative growth of the fields over the entire interaction length [1]. It has been shown that phase-matching in second-order nonlinear ($\chi^{(2)}$) media can be obtained by exploiting the natural [2] or induced [3] circular birefringence. In particular Ref.[3] shows that phase-matching in some $\chi^{(2)}$ magneto-optic crystals could be in principle achieved by applying a magnetic field along the direction of propagation of the interacting waves and by using appropriate right or left circular polarizations. However, due to large values of dispersion, the magnetic fields required for complete phase-matching are usually too high for practical devices. In this paper we study optical parametric interactions in third-order nonlinear ($\chi^{(3)}$) magneto-optic media where the magneto-optic contribution to phase matching is used to induce new phase-matching branches rather than to compensate for the full phase-mismatch (which is compensated intrinsically or by other means). In particular we focus on Four-Wave-Mixing (FWM) and Three-Wave-Mixing (TWM) in the presence of a periodic electrostatic field in *isotropic* $\chi^{(3)}$ magneto-optic media. FWM is the typical interaction in a $\chi^{(3)}$ medium, i.e. the conversion of two pump photons at frequency ω_3 and ω_4 into a signal (ω_1) and idler (ω_2) photons: $\omega_3 + \omega_4 = \omega_1 + \omega_2$. In the small-signal gain limit, efficient conversion occurs when the wave vectors of interacting waves are phase matched, i.e. $k_3 + k_4 = k_1 + k_2$ [4]. Degenerate FWM in isotropic nonlinear gyrotropic media has been studied, however without particular reference to phase-matching issues (given the fact that the interaction was degenerate, i.e. $\omega_1 = \omega_2 = \omega_3 = \omega_4 = \omega$) [5]. Another interesting interaction which can occur in a $\chi^{(3)}$ medium is electric-field induced TWM (EI-TWM), i.e. a FWM where one of the pump waves becomes a low frequency or electrostatic field ($\omega_4 \sim 0$) leading to an equivalent $\chi^{(2)}$ nonlinearity. In this case, quasi-phase matching (QPM) techniques have been proven to be powerful means to make efficient the TWM process $\omega_3 = \omega_1 + \omega_2$. Typical examples are second-harmonic generation (SHG) and parametric down-conversion (PDC) in optical glass waveguides where a periodic electric field is applied [6] or stored using poling techniques [7]. In this paper we will refer more to PDC rather than SHG although one can apply straightforwardly to the latter case the same concepts.

To study FWM and EI-TWM interactions in an isotropic $\chi^{(3)}$ magneto-optic medium, we define a (x, y, z) Cartesian frame and we consider, without loosing of generality, electromagnetic (e.m.) waves propagating along the z-direction which are plane and quasi-monocromatic. We also assume that a magnetic field H_z is applied along the z-axis. Each e.m. field component $E_k(z,t)$ oscillating at frequency ω_δ can then be represented as $E_k(z,t) = \mathrm{Re}[E_k(z,\omega_\delta)\exp(-i\omega_\delta t)]$ where $k = x, y$ and $\delta = 1,2,3,4$; instead of $E_k(z,\omega)$, we may equivalently write $E_k^{\omega}(z)$. The total field will excite in the material a polarization $P_k(z,t)$ made of two contributions, a linear one $P_k^L(z,t)$, due to linear material dispersion and magneto-optic effect (Faraday effect), and a nonlinear one $P_k^{NL}(z,t)$ due to $\chi^{(3)}$. If we use for the polarizations a representation similar to that used for the fields and we omit for simplicity the dependence on z, the components $P_k^L(\omega_\delta)$ and $P_k^{NL}(\omega_\delta)$ oscillating at ω_δ are given by: $P_k^L(\omega_\delta) = \epsilon_0\chi_{kl}^{(ee)}(\omega_\delta;\omega_\delta)E_l^{\omega_\delta} - i\epsilon_0\chi_{klz}^{(eem)}(\omega_\delta;\omega_\delta,0)E_l^{\omega_\delta}H_z$ and $P_k^{NL}(\omega_\delta) = \epsilon_0\sum_{\alpha,\beta,\gamma}\chi^{(eeee)}(\omega_\delta = \omega_\alpha + \omega_\beta + \omega_\gamma;\omega_\alpha,\omega_\beta,\omega_\gamma)E_l^{\omega_\alpha}E_m^{\omega_\beta}E_n^{\omega_\gamma}$, where $\chi_{kl}^{(ee)}$ is the linear electric susceptibility tensor (which reduces to a scalar, $\chi^{(ee)}$, for isotropic media), $\chi_{klz}^{(eem)}$ is the magneto-optic susceptibility, $\chi_{klmn}^{(eeee)}$ is the third-order nonlinear electric susceptibility tensor, and $g_{\alpha,\beta,\gamma}$ the degeneracy factor due to intrinsic permutation symmetry of $\chi_{klmn}^{(eeee)}$. For isotropic media there are only 21 nonzero components, three of which are indipendent ($\chi_{xxxx} = \chi_{xxyy} + \chi_{xyxy} + \chi_{xyyx}$, where we have omitted the superscript $^{(eeee)}$ for simplicity). In addition, in the case of electronic response, $\chi_{xxyy} = \chi_{xyxy} = \chi_{xyyx}$. In a general FWM process ($\omega_3 + \omega_4 = \omega_1 + \omega_2$), if we assume negligible pump depletion and weak fields at ω_1 and ω_2, the nonlinear polarization $P_x^{NL}(\omega_1)$ oscillating at ω_1 is given by:

$$P_x^{NL}(\omega_1) = \epsilon_0 g_{deg}\chi^{(3)}\left[E_x^{\omega_4}E_x^{\omega_3}E_x^{\omega_2 *} + \frac{1}{3}E_x^{\omega_4}E_y^{\omega_3}E_y^{\omega_2 *} + \frac{1}{3}E_y^{\omega_4}E_x^{\omega_3}E_y^{\omega_2 *} + \frac{1}{3}E_y^{\omega_4}E_y^{\omega_3}E_x^{\omega_2 *}\right] \qquad (1)$$

where $\chi^{(3)} = \chi_{xxxx}(\omega_1 = \omega_3 + \omega_4 - \omega_2;\omega_3,\omega_4,-\omega_2)$ and g_{deg} is the degeneracy factor of the process. For simplicity, in writing Eq.(1) we have neglected pump-induced cross-phase modulation terms, which would lead merely to a pump-dependent shift of wave numbers of interacting fields. The expression for $P_x^{NL}(\omega_2)$ is obtained by the formal substitution $\omega_1 \leftrightarrow \omega_2$, whereas the expressions for $P_y^{NL}(\omega_1)$ and $P_y^{NL}(\omega_2)$ are obtained from those of $P_x^{NL}(\omega_1)$ and $P_x^{NL}(\omega_2)$ by the substitution $x \leftrightarrow y$. It is useful to use a circular representation for the field and polarization vectors by setting: $E_\pm = (E_x \pm iE_y)/\sqrt{2}$ and $P_\pm = (P_x \pm iP_y)/\sqrt{2}$. In the circular representation the polarization $P_+^{NL}(\omega_1)$ becomes:

$$P_+^{NL}(\omega_1) = \frac{2}{3}\epsilon_0 g_{deg}\chi^{(3)}\left[E_+^{\omega_4}E_+^{\omega_3}E_+^{\omega_2 *} + E_-^{\omega_4}E_+^{\omega_3}E_-^{\omega_2 *} + E_+^{\omega_4}E_-^{\omega_3}E_-^{\omega_2 *}\right] \tag{2}$$

The expressions for $P_+^{NL}(\omega_2)$, $P_-^{NL}(\omega_1)$ and $P_-^{NL}(\omega_2)$ are obtained from Eq.(2) by the formal substitutions $\omega_1 \leftrightarrow \omega_2$ and $+ \leftrightarrow -$. The propagation equation for the fields oscillating at ω_1 and ω_2 in the circular polarization basis can be written as:

$$\partial_z^2 E_\pm^{\omega_{1,2}} + k_\pm^2(\omega_{1,2})E_\pm^{\omega_{1,2}} = -\mu_0\omega_{1,2}^2 P_\pm^{NL}(\omega_{1,2}) \tag{3}$$

where $k_\pm$ are the wave-vectors for right and left-circularly polarised components which account for both linear dispersion (in general material and waveguide) and magneto-optic effect, i.e. $k_\pm^2(\omega) = (\omega/c_0)^2[n^2(\omega) \mp \chi_{xyz}^{(eem)}(\omega;\omega,0)H_z]$ and $n^2(\omega) = 1 + \chi^{(ee)}(\omega)$ is the refractive index. For simplicity, besides the material isotropy of the medium, we have also assumed absence of any waveguide birefringence.

As example of magneto-optic effects on the phase-matching condition, we study now in more details two kinds of interactions: FWM with degenerate pumps ($\omega_3 = \omega_4$) and EI-PDC when a periodic electrostatic field is applied to the isotropic medium. To this aim, it is worth introducing the slowly-varying approximation for the signal and idler fields by the ansatz:

$$E_\pm^{\omega_{1,2}}(z) = \left[\frac{\omega_{1,2}n(\omega_{1,2})}{\omega_3}\right]^{1/2} A_\pm^{\omega_{1,2}}(z)\exp\left[ik_\pm(\omega_{1,2})z\right] \tag{4}$$

Furthermore, for the sake of definiteness we consider a right circularly polarized pump field assuming $E_-^{\omega_3} = 0$ and $E_+^{\omega_3} = I_3^{1/2}\exp\left[ik_+(\omega_3)z\right]$; similar results would be obtained in case of a left circularly polarized pump.

FWM process ($2\omega_3 = \omega_1 + \omega_2$): in this case one has $g_{deg} = 3/4$ and only the right circularly polarized fields $E_+^{\omega_{1,2}}$ will experience gain. After substituting the expression for the nonlinear polarization (Eq.(2)) in the propagation equation (Eq.(3)) and using the slowly varying envelope approximation (Eq.(4)), it is straighforward to show that the coupled equations for the signal and idler field envelopes$A_+^{\omega_{1,2}}$ takes the usual form of parametric processes [4]: $\partial_z A_+^{\omega_{1,2}} = i\Gamma I_3 A_+^{\omega_{2,1}*}\exp(i\Delta k_+ z)$, where $\Gamma = \chi^{(3)}[\omega_1\omega_2/n(\omega_1)n(\omega_2)]^{1/2}/4c_0$ (c_0 is the speed of light in vacuum), $\Delta k_+ = \Delta k_{disp} + H_z[V(\omega_1)+V(\omega_2)-2V(\omega_3)]$ with $\Delta k_{disp} = 2(\omega_3/c_0)n(\omega_3) - (\omega_2/c_0)n(\omega_2) - (\omega_1/c_0)n(\omega_1)$ being the wave-vector mismatch which in general takes account of both material and waveguide dispersion and $V(\omega) = [\omega\chi^{(eem)}(\omega)]/[2n(\omega)c_0]$ the Verdet constant. From the expression of Δk_+ it is clear that a magneto-optic contribution in isotropic $\chi^{(3)}$ media is present only when ω_1 and ω_2 are relatively different from ω_3 (i.e. off degeneracy), so that one could exploit the dispersion for the Verdet constant (which can be quite significant for certain materials [8]). Note that the same results apply for left circularly polarized pump, signal and idler fields. When the pump field has both a left- and right- circular polarization component (e.g. linearly polarized), coupling between opposite circular polarizations for signal and idler fields are possible (see Eq.(2), last two terms). However even for this case the magneto-optic contribution to phase-matching, given by $H_z[V(\omega_1) - V(\omega_2)]$, can be significant only away from degeneracy.

EI-PDC process ($\omega_3 = \omega_1+\omega_2$, $\omega_4 \sim 0$): in this case we apply a periodic static electric field E_0, oriented along the x-axis, to the isotropic medium. Let us assume a periodic +/0 square modulation of E_0 along the direction of propagation z with period $\Lambda \sim 2\pi/\Delta k_{disp}$, which induces a periodic effective $\chi^{(2)}$ proportional to $\chi^{(3)}E_0$. Equation (2) still holds apart from the formal substitutions $g_{deg} = 3$ and $E_\pm^{\omega_4} \leftrightarrow E_0/\sqrt{2}$. For a right circularly polarized pump, after substitution of Eqs.(2) and (4) into Eq.(3) and assuming first-order QPM, the following evolution equations for signal and idler fields can be derived using multiple-scale (or averaging) techniques:

$$\partial_z A_+^{\omega_1} = i\theta\left[A_+^{\omega_2*}\exp(i\Delta k_1 z) + A_-^{\omega_2*}\exp(i\Delta k_3 z)\right] \tag{5a}$$

$$\partial_z A_-^{\omega_1} = i\theta A_+^{\omega_2*}\exp(i\Delta k_2 z) \tag{5b}$$

$$\partial_z A_+^{\omega_2*} = -i\theta\left[A_+^{\omega_1}\exp(-i\Delta k_1 z) + A_-^{\omega_1}\exp(-i\Delta k_2 z)\right] \tag{5c}$$

$$\partial_z A_-^{\omega_2*} = -i\theta A_+^{\omega_1}\exp(-i\Delta k_3 z) \tag{5d}$$

where $\theta = \sqrt{2}\Gamma E_0\sqrt{I_3}/\pi$, $\Delta k_i = \Delta k_{disp} - 2\pi/\Lambda + \Delta k_i^{MO}$ ($i = 1,2,3$), and the magneto-optic contributions Δk_i^{MO} are given by $\Delta k_1^{MO} = H_z[V(\omega_1) + V(\omega_2) - V(\omega_3)]$, $\Delta k_2^{MO} = H_z[-V(\omega_1) + V(\omega_2) - V(\omega_3)]$ and $\Delta k_3^{MO} = H_z[V(\omega_1) - V(\omega_2) - V(\omega_3)]$. The general solution to Eqs.(5) is a linear combination of solutions of the form $(A_+^{\omega_1}, A_-^{\omega_1}, A_+^{\omega_2 *}, A_-^{\omega_2 *})^T = (y_1 \exp(i\Delta k_1 z), y_2 \exp(i\Delta k_2 z), y_3, y_4 \exp[i(\Delta k_1 - \Delta k_3)z])^T \exp(\eta z)$, where η and $(y_1, y_2, y_3, y_4)^T$ can be found as eigenvalues and corresponding eigenvectors of a fourth-order matrix. Efficient pump conversion thus occurs if $\mathrm{Re}(\eta) > 0$, and the parametric gain σ of the process can be defined as the maximum growth rate $\mathrm{Re}(\eta)$ among the four eigenvalues. A typical behavior of σ as a function of $(\Delta k_{disp} - 2\pi/\Lambda)$ is shown in Fig.1. In absence of a magnetic field one has $\Delta k_1 = \Delta k_2 = \Delta k_3$ and efficient PDC occurs near the QPM condition $\Delta k_{disp} = 2\pi/\Lambda$. When a magnetic field is applied, degeneracy of Δk_i $(i = 1, 2, 3)$ is left and three parametric instability branches appear corresponding to the three different QPM conditions $\Delta k_i = 0$ and to the generation of signal and idler fields with different polarization states. For example when $\Delta k_1 = 0$ efficient PDC occurs for $A_+^{\omega_1}$ and $A_+^{\omega_2}$, whereas when $\Delta k_2 = 0$ for $A_-^{\omega_1}$ and $A_+^{\omega_2}$ (see Eqs.(5)). In the case where the three branches do not overlap, each PDC process can be well described by standard two coupled equations of parametric processes, for which analytical solution for the parametric gain can be found and reads: $\sigma_i = \mathrm{Re}\left[\sqrt{\theta^2 - (\Delta k_i/2)^2}\right]$ $(i = 1, 2, 3)$ (see dashed lines in Fig.1). Note that, close to degenerate PDC (i.e. $\omega_1 \sim \omega_2$) one has $\Delta k_2 \sim \Delta k_3$ and thus only two distinct parametric instability branches occur. A continuous tuning of the instability branches can be achieved by a control of the phase-matching terms entering in Eqs.(5) exploiting the magneto-optic contribution Δk_i^{MO}. Compared to the FWM case studied above, the magneto-optic contributions are here more significant, given the fact that they are made of only three terms (as the number of optical frequencies) which can also be very different because of the dispersion of the Verdet constant and the significant difference between the optical frequencies.

As an example, let us consider a lead-oxide (SF57 Schott glass) based optical fibre. SF57 is a glass with a relatively high refractive index, hence $\chi^{(3)}$ (about 5-10 times that of silica [9]), and high V constant and dispersion of V ($V \sim 5$ rad/Tm and 15 rad/Tm at 1550 and 775 nm respectively [8]). In addition lead-oxide glasses can be drawn into single-mode optical fibres [9]. Figure 2 shows the parametric gain as a function of signal wavelength for a typical SF57 fibre in case of near degenerate PDC around 1550 nm for different values of applied magnetic field. As evident from

Fig. 2 significant tuning and/or shaping of the QPM branches for nonlinear interactions involving the lowest-order (i.e. combination of degenerate and orthogonal LP_{01}) circularly polarised mode fields can be achieved by applying reasonable magnetic fields.

In conclusion new phase-matching conditions can be generated for circularly polarized four- and three-wave interactions in isotropic $\chi^{(3)}$ magneto-optic media by applying a magnetic-field. In particular we have shown that the technique can be practically implemented for broad-band and/or tuning of parametric gain. New research is in progress, which should also lead to the first experimental results.

V. Pruneri is the Pirelli Research Fellow of Photonics at Southampton. The authors acknowledge M. Petrovich for providing data on Verdet constants.

REFERENCES

[1] J. A. Armstrong, N. Bloembergen, J. Ducuing, and S. Pershan, Phys. Rev. **127**, 1918 (1962).

[2] H. Rabin and P. P. Bey, Phys. Rev. **156**, 1010 (1967).

[3] F. Jonsson and C. Flytzanis, Opt. Lett. **24**, 1514 (1999).

[4] R. H. Stolen and J. E. Bjorkholm, IEEE J. Quant. Electron. **QE-18**, 1062 (1982); G.P. Agrawal, *Nonlinear Fibre Optics*, 2nd ed. (Academic, New York, 1995).

[5] P. Unsbo and C. Flytzanis, J. Opt. Soc. Am. B **14**, 560 (1997).

[6] R. Kashyap, J. Opt. Soc. Am. B **6**, 313 (1989).

[7] V. Pruneri, G.Bonfrate, P. G. Kazansky, D. J. Richardson, N. G. Broderick, J. P. De Sandro, C. Simonneau, P. Vidakovic, and J. A. Levenson, Opt. Lett. **24**, 208 (1999); G. Bonfrate, V. Pruneri, P. G. Kazansky, P. Tapster, and J. G. Rarity, Appl. Phys. Lett. **75**, 2356 (1999).

[8] N. F. Borrelli, The Journal of Chem. Phys. **41**, 3289 (1964).

[9] N. F. Borrelli, B. G. Aitken, and M. A. Newhouse, J. Non-Cryst. Solids **185**, 109 (1995); M. A. Newhouse, D. L. Weidman, and D. W. Hall, Opt. Lett. **15**, 1185 (1990).

[10] Optical Glass, Schott catalogue.

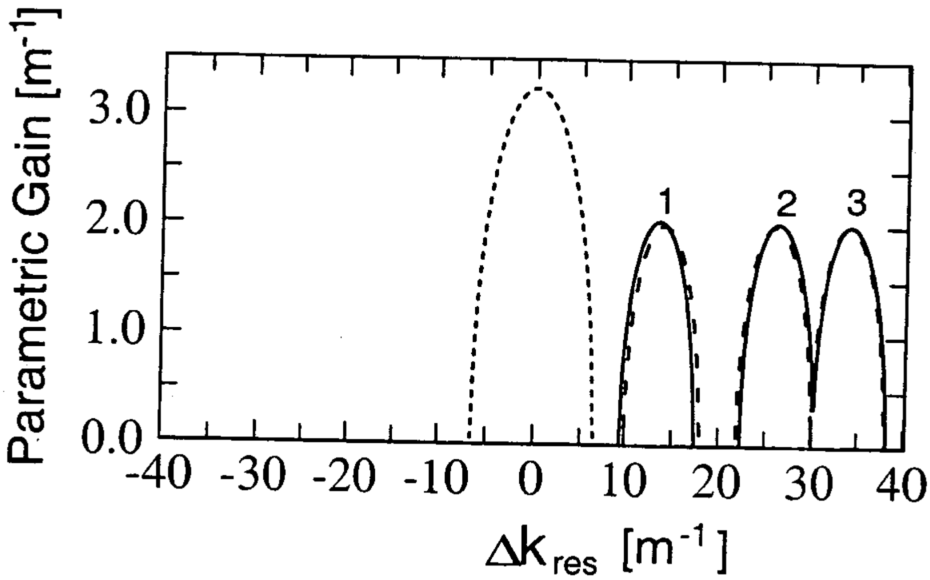
Figure Captions.

Fig.1. Parametric gain for EI-PDC as a function of residual phase-mismatch $\Delta k_{res} = \Delta k_{disp} - 2\pi/\Lambda$ in absence (dotted line) and with applied magnetic field (continuous line). The three instability branches 1,2 and 3 correspond to the QPM conditions $\Delta k_1 = 0$, $\Delta k_2 = 0$ and $\Delta k_3 = 0$ respectively. The dashed curves, nearly overlapped with the continuous lines, are the parametric instability branches as predicted by approximate uncoupled parametric processes (see text). Parameter values are: $\theta = 2\text{m}^{-1}$, $\Delta k_1^{MO} = -14\text{m}^{-1}$, $\Delta k_2^{MO} = -26\text{m}^{-1}$, $\Delta k_3^{MO} = -34\text{m}^{-1}$.

Fig.2. Parametric gain for near-degenerate EI-PDC as a function of signal (idler) wavelength in a SF57 fibre for different values of magnetic field. Dotted line: $H_z = 0$ mT; dashed line: $H_z=$ -200 mT; continuous line: $H_z=$ -400 mT. The fibre has a numerical aperture of 0.2 and a core radius of 2.5 μm. Parameter values are: pump wavelength λ_3=775 nm, Λ=26.752 μm (corresponding to $\Delta k_{disp} = 0$ at degeneracy $\lambda_1 = \lambda_2$ =1550 nm) and $\theta = 0.3$ m^{-1}. For the calculation of the wavelength dependent term Δk_{disp} the Sellmeier equation given in Ref.[10] has been used.

V. Pruneri and S. Longhi, "Optical Parametric Processes ..."

Fig. 1



V. Pruneri and S. Longhi, "Optical Parametric Processes ..."

Fig. 2