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Modulational instability and four-wave mixing in anisotropic $\chi^{(3)}$ magneto-optic media

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Abstract

Nearly-degenerate four-wave mixing (FWM) in anisotropic magneto-optic media is theoretically investigated. It is shown that the anisotropic nature of the nonlinear $\chi^{(3)}$ tensor is responsible for the appearance of a novel FWM instability branch for which phase matching can be conveniently controlled by the application of an axial static magnetic field.

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Four-wave-mixing (FWM) is a fundamental form of nonlinear wave interaction in nonlinear $\chi^{(3)}$ media which represents a well-known technique for the generation of new frequencies in nonlinear optics [1]. Despite FWM is a rather old topic in nonlinear optics, the continuous interest, both theoretical and experimental, toward the study of parametric wave mixing phenomena is motivated by their enormous and widespread applications in different fields of nonlinear optics, including frequency conversion, phase conjugation and parametric amplification, and by their importance in the technologically-important field of optical fiber communications [2], where FWM may be useful for, e.g., all-optical switching, demultiplexing, frequency conversion and spectral inversion [3]. In case of a partially degenerate interaction, FWM manifests as the conversion of two pump photons at frequency ω_p into signal and idler sideband photons at frequencies ω_s and ω_i satisfying the energy-conservation relation $2\omega_p = \omega_s + \omega_i$. In the small-signal interaction limit, efficient conversion of pump photons occurs when the phase matching condition $2k_p = k_s + k_i$ is realized, where the wave numbers entering in this equation account for both material dispersion and nonlinear pump-induced cross- and self-phase modulation terms. In absence of any signal seeding, FWM manifests as an instability of the propagating pump wave with the growth from noise of sideband frequencies (modulational instability), with a maximum growth rate for frequencies satisfying the phase matching condition. When FWM is used for frequency conversion or for parametric amplification, a signal wave is seeded into the nonlinear medium, and the conversion efficiency and gain-bandwidth are determined again by phase-matching arguments [2,4]. In both cases, phase matching techniques are of major importance for the achievement of tunability and broad-band conditions in the frequency conversion process. Circular birefringence due to the Faraday effect has been proposed as a means for continuous control of phase-matching in nonlinear wave interactions occurring in $\chi^{(2)}$ media [5,6]. However, in case of second-order nonlinear interactions, the phase mismatch due to material dispersion is usually relatively large and the magneto-optic effects achievable with externally-applied magnetic fields are, comparatively, weak and not enough to compensate for dispersion mismatch [5]. In case of nearly degenerate FWM in third-order nonlinear media, the phase mismatch among nearly degenerate pump, signal and idler fields is relatively weak, and it is thus expected that magneto-optic effects may become of some experimental relevance in the control of phase matching [7]. Nevertheless, since in *isotropic* third-order nonlinear media the spin angular momentum of interacting waves associated to their polarization state is conserved in the nonlinear process, it is envisaged that a significant magneto-optic contribution to the phase matching should occur solely in a medium with an *anisotropic* $\chi^{(3)}$ nonlinear tensor.

In this Letter we present the theory of nearly-degenerate FWM interaction in anisotropic magneto-optic $\chi^{(3)}$ media, and we reveal the existence of a novel FWM instability branch, closely related to the anisotropic nature of the tensorial $\chi^{(3)}$ nonlinearity, for which phase-matching can be conveniently and continuously controlled by the application of a longitudinal static magnetic field exploiting the magneto-optic properties of the medium. We illustrate this by considering in details the case of cubic crystals of classes 432, $\overline{4}3m$ and $m3m$ [1], and assume plane-wave fields copropagating along the z axis of the crystal. The present analysis covers, as a particular case, that corresponding to FWM in an isotropic $\chi^{(3)}$ medium; furthermore, it could be extended to other anisotropic crystal classes, such as hexagonal and trigonal classes. We assume a cartesian reference frame (x, y, z) coincident with the crystal axes x, y and z, and consider a plane-wave quasi-monocromatic field $\mathcal{E}(z,t)$ propagating along the z-axis with a static magnetic field H_z applied along the same direction, i.e. we set $\mathcal{E}(z,t) = \sum_{\omega} \mathrm{Re}[\mathbf{E}(\omega)\exp(-i\omega t)]$, where the sum is extended over a band of frequencies close to that of the pump field ω_p. The positive-frequency parts $\mathbf{E}(\omega)$ of the field satisfy the coupled wave equations [1]:

$$\frac{\partial^2 \mathbf{E}(\omega)}{\partial z^2} + \left(\frac{\omega}{c_0}\right)^2 \mathbf{E}(\omega) = -\mu_0 \omega^2 \mathbf{P}(\omega) \tag{1}$$

where c_0 is the speed of light in vacuum and $\mathbf{P}(\omega)$ is the positive-frequency part of polarization, which accounts for both linear polarization $P_k^L(\omega) = \epsilon_0 \chi_{kl}^{(ee)} E_l(\omega) - i\epsilon_0 \chi_{klz}^{(eem)}(\omega;\omega,0) E_l(\omega) H_z$ due to linear material dispersion and magneto-optic effect, and nonlinear one $P_k^{NL}(\omega) = \epsilon_0 \sum_{\omega_\alpha+\omega_\beta+\omega_\gamma=\omega} g_{\alpha,\beta,\gamma} \chi_{klmn}^{(eeee)}(\omega;\omega_\alpha,\omega_\beta,\omega_\gamma) E_l(\omega_\alpha) E_m(\omega_\beta) E_n(\omega_\gamma)$. In these equations, $\chi_{kl}^{(ee)}$ is the linear electric susceptibility tensor, $\chi_{klz}^{(eem)}$ the magneto-optic susceptibility ($\chi_{klz}^{(eem)}$= - $\chi_{lkz}^{(eem)}$), $\chi_{klmn}^{(eeee)}$ the tensorial third-order electric susceptibility, $g_{\alpha,\beta,\gamma}$ the degeneracy factor due to intrinsic permutation symmetry of $\chi_{klmn}^{(eeee)}$, and the indices k, l, m, n vary over x, y, z. In case of a cubic medium, the linear tensorial electric susceptibility $\chi_{kl}^{(ee)}(\omega)$ reduces to a scalar $\chi^{(ee)}(\omega)$, whereas

$\chi^{(eeee)}_{klmn}$ has 21 nonzero elements, of which only 4 are independent [1]. It is worth introducing a circular polarization basis for the electric field and polarization vectors by setting $E_\pm = (E_x \pm iE_y)/\sqrt{2}$, $P_\pm = (P_x \pm iP_y)/\sqrt{2}$, so that Eqs.(1) assume the form:

$$\frac{\partial^2 E_\pm(\omega)}{\partial z^2} + k_\pm^2(\omega)E_\pm(\omega) = -\mu_0\omega^2 P_\pm^{NL}(\omega) \tag{2}$$

In Eqs.(2), the wave-numbers $k_\pm$ for right- and left-circularly polarized field components, which account for linear dispersion and magneto-optic effect, are given by $k_\pm^2(\omega) = (\omega/c_0)^2\left[n^2(\omega) \mp \chi^{(eem)}_{xyz}(\omega;\omega,0)H_z\right]$, where $n(\omega) \equiv \sqrt{1+\chi^{(ee)}(\omega)}$ is the refractive index of the crystal. Notice that, in absence of nonlinear terms introduced by $P_\pm^{NL}$, the left- and right-circularly polarized field components are decoupled and propagate with different phase velocities in presence of the applied magnetic field H_z. Let us now consider the case where an intense *circularly-polarized* pump field at frequency ω_p is incident upon the nonlinear crystal. For the sake of definiteness, let us assume that the pump field has a left circular polarization. In this case, the evolution equation for the pump field $E_-(\omega_p)$ is given by Eq.(2) with $\omega = \omega_p$, the nonlinear driving polarization term at leading order being given by $P_-^{NL}(\omega_p) = (3/8)\epsilon_0 a|E_-(\omega_p)|^2E_-(\omega_p)$, where $a \equiv \chi^{(eeee)}_{xxxx}(\omega_p;\omega_p,\omega_p,-\omega_p) + 2\chi^{(eeee)}_{xxyy}(\omega_p;\omega_p,\omega_p,-\omega_p) - \chi^{(eeee)}_{xyyx}(\omega_p;\omega_p,\omega_p,-\omega_p)$. The solution to the pump wave equation, assuming a forward wave, is thus $E_-(\omega_p,z) = \sqrt{I_p}\exp(ik_{p-}z)$, where I_p is proportional to the constant pump field intensity and k_{p-} is the nonlinear pump wave number, given by $k_{p-}^2 = k_-^2(\omega_p) + (3/8)a(\omega_p/c_0)^2 I_p$. A linear stability analysis of this solution can be carried out by considering the evolution of small field perturbations $E_\pm(\omega) \neq E_-(\omega_p)$, as ruled by Eqs.(2), with the polarization driving terms given by:

$$P_+^{NL}(\omega) = (3/8)\epsilon_0\left[bE_-^2(\omega_p)E_+^*(2\omega_p-\omega) + 2c|E_-(\omega_p)|^2E_+(\omega)\right] \tag{3}$$

$$P_-^{NL}(\omega) = (3/8)\epsilon_0\left[aE_-^2(\omega_p)E_-^*(2\omega_p-\omega) + 2a|E_-(\omega_p)|^2E_-(\omega)\right] \tag{4}$$

In Eqs.(3,4) we have set $b \sim \chi^{(eeee)}_{xxxx}(\omega_p;\omega_p,\omega_p,-\omega_p) - 2\chi^{(eeee)}_{xxyy}(\omega_p;\omega_p,\omega_p,-\omega_p) - \chi^{(eeee)}_{xyyx}(\omega_p;\omega_p,\omega_p,-\omega_p)$ and $c \sim \chi^{(eeee)}_{xxxx}(\omega_p;\omega_p,\omega_p,-\omega_p) + \chi^{(eeee)}_{xyyx}(\omega_p;\omega_p,\omega_p,-\omega_p)$. Furthermore, in deriving the above equations, we used the intrinsic permutation symmetry and the Kleinmann symmetry of the $\chi^{(3)}$ tensor, valid for a purely reactive medium and neglecting the dispersion of the $\chi^{(3)}$ tensor by assuming ω close to the pump frequency ω_p. An analysis of Eqs.(2-4) reveals that there exist, in general, three distinct sources of instabilities. The first one is a parametric instability for the right circular polarization of pump field $E_+(\omega_p)$, whereas the second and the third one correspond to a modulational (FWM) instability for sideband signal ($\omega_s = \omega \neq \omega_p$) and idler ($\omega_i = 2\omega_p - \omega_s$) frequency generation of left (type-I FWM) and right (type-II FWM) circular polarizations, respectively. Two of these instability branches, namely the polarization instability for the pump field and type-II FWM instability, vanish whenever $b = 0$, a situation that occurs in an isotropic medium, where $\chi^{(eeee)}_{xxxx} = \chi^{(eeee)}_{xxyy} + \chi^{(eeee)}_{xyxy} + \chi^{(eeee)}_{xyyx}$ [1]. In the following, we will be mainly concerned with the FWM instabilities, which correspond to frequency conversion, and will not consider further the polarization instability of the pump field. After ntroduction of the slowly-varying field amplitudes by the ansatz $E_\pm(\omega_{s,i}) = A^{s,i}_\pm(z)\exp(ik^{s,i}_\pm z)$, with the nonlinear wave numbers $k^{s,i}_\pm$ defined by $(k^{s,i}_+)^2 \equiv k^2_+(\omega_{s,i}) + (3/4)(\omega_{s,i}/c_0)^2 cI_p$, $(k^{s,i}_-)^2 \equiv k^2_-(\omega_{s,i}) + (3/4)(\omega_{s,i}/c_0)^2 aI_p$, the equations that govern type-I and type-II FWM instabilities are readily obtained feom Eqs.(2-4) and read explicitly:

$$\frac{\partial A^{s,i}_\pm}{\partial z} = id_\pm I_p (A^{i,s}_\pm)^* \exp(i\Delta k_\pm z) \tag{5}$$

In Eqs.(5), we have set $d_+ \sim 3b\omega_p/(16c_0 n)$, $d_- \sim 3a\omega_p/(16c_0 n)$, and the phase mismatch terms $\Delta k_\pm$ can be cast in the form:

$$\Delta k_+ = \Delta k_{disp} - \gamma_+ I_p + 4VH_z \tag{6}$$

$$\Delta k_- = \Delta k_{disp} - \gamma_- I_p \tag{7}$$

where $\gamma_+ \equiv (3/8)\omega_p(2c - a)/(c_0 n)$, $\gamma_- \equiv (3/8)\omega_p a/(c_0 n)$, $V \equiv (\omega_p/2c_0 n)\chi^{(eem)}$ is the Verdet constant at frequency $\sim \omega_p$, and $\Delta k_{disp} \equiv 2(\omega_p/c_0)n(\omega_p) - (\omega_s/c_0)n(\omega_s) - (\omega_i/c_0)n(\omega_i)$ is the phase mismatch due to material dispersion [8]. Notice that, for both type-I and type-II FWM, Eqs.(5) have the standard form of parametric interaction encountered in scalar wave field models [2]; however, these two processes differ basically for the appearance, in FWM of type-II, of the magneto-optic effect in the phase mismatch (see Eq.(6)). This is ascribable to the circumstance that type-II FWM corresponds to the annihilation of two left circularly-polarized pump photons and to the creation of signal and idler photons with right-circular polarization, i.e. it does not conserve the angular photon momentum (see Eq.(3)). Notice also that, in case of an isotropic $\chi^{(3)}$ medium, type-II FWM vanishes and solely the instability branch associated to type-I FWM remains, for which the phase matching is largely independent (at least close to degeneracy) of the applied magnetic field. The parametric gain for the four-wave interaction is given by $g_{\pm} = \left[d_{\pm}^2 I_p^2 - (\Delta k_{\pm}/2)^2\right]^{1/2}$, and a parametric instability takes place for $\mathrm{Re}(g_{\pm}) > 0$, with a maximum gain at phase matching $\Delta k_{\pm} = 0$. Notice that, close to degeneracy, we may write $\Delta k_{disp} \sim -\beta_2(\omega_{s,i} - \omega_p)^2$, where β_2 is the second-order material dispersion [8], so that the peak of parametric gain, in case of type-II FWM, is attained at the sideband frequencies $\Omega_{MAX}^2 \equiv (\omega_p - \omega_{s,i})^2 \sim (-\gamma_+ I_p + 4VH_z)/\beta_2$. If we neglect the cross-phase modulation term $\gamma_+ I_p$, we thus have $\Omega_{MAX} \sim (4VH_z/\beta_2)^{1/2}$, and the parametric gain peak can be conveniently tuned by the application of the axial magnetic field H_z. Notice that, since H_z can be either positive or negative, phase matching tuning by magnetic field application is effective regardless of the sign of material dispersion. In Fig.1 the behavior of the parametric gain $\mathrm{Re}(g_+)$ versus Δk_{disp} for type-II FWM is plotted for a few values of VH_z, showing magnetic-field-induced shift of the peak gain. As a practical example of tunability, let us consider Terbium Gallium Garnet (TGG) as a anisotropic cubic $\chi^{(3)}$ medium, which exhibits a high Verdet constant and is transparent in the visible and infrared spectral regions. Owing to these qualities, TGG is generally used for constructing Farady effect devices, such as rotators and optical isolators, and it is also an appropriate material for optical waveguides and substrates [9]. At the pump wavelength $\lambda_p = 1.5$ μm the values of dispersion and Verdet constants are, respectively, $\beta_2 \sim 92$ ps^2/km [10] and $V \sim 25$ rad/Tm [11]. Notice that, since the material dispersion is positive, no phase matching may occur for TGG in absence of an applied magnetic field. Assuming $\gamma_+ I_p \sim 1$ m^{-1}, one obtains $\nu_{MAX} \sim 0$ for an applied magnetic field $H_z \sim 10$ mT, and $\nu_{MAX} \sim 3.67$ THz for $H_z \sim 500$ mT, corresponding to a shift of the parametric peak gain of more than 3 THz.

In conclusion, nearly-degenerate FWM in magneto-optic media with anisotropic $\chi^{(3)}$ nonlinearity has been investigated, and phase matching control of parametric gain exploiting the magneto-optic effect has been proposed and associated to the existence of a FWM instability branch which does not conserve the spin angular momentum of interacting waves. In this paper we have considered an example referring to rare earth garnets (including TGG). However this research may be important for any anisotropic medium which combines high values of third-order nonlinearity and Verdet constant, e.g. glass ceramics optical fibers and waveguides [12] where a suitable dispersed nanocrystalline structure in glass could provide the functionality of an anisotropic $\chi^{(3)}$ magneto-optic medium over considerable lengths and under tight light confinement.

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Figure Captions.

Fig.1. Behavior of parametric gain $\mathrm{Re}(g_+)$ versus dispersion mismatch Δk_{disp} in case of type-II FWM instability for a few values of VH_z: (1) VH_z=0; (2) VH_z=5 m^{-1}; (3) VH_z =-5 m^{-1}. Parameter values are: $d_+ I_p = 3$ m^{-1} and $\gamma_+/d_+ = 3$.

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Fig. 1