Eigenmodes of $\chi^{(2)}$ wave mixings: cross-induced second-order nonlinear refraction

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Received March 12, 1993

Eigenmodes of three-wave interactions in second-order [$\chi^{(2)}$] nonlinear materials are explored that have a no-energy-exchange property between all the waves. They are shown to exhibit cross-induced nonlinear refraction and to be ideal candidates for amplitude-dependent phase control in $\chi^{(2)}$ materials that emulate nonlinear refractive-index effects in third-order [$\chi^{(3)}$] materials. It is shown that all these eigenmodes are spatially stable.

The second-order nonlinear refraction occurring in second-harmonic (SH) generation and emulating the third-order nonlinear refractive index at the fundamental harmonic (FH) has attracted much interest recently.[1-5] The cascading process, whereby the transformation FH–SH–FH results in amplitude-dependent phase velocity for the FH, was demonstrated experimentally in CDA and KTP crystals[2,3]; its particular manifestation is self-focusing and self-defocusing.[3,5] In this Letter we show that (i) an alternative no-energy-exchange regime (eigenmode) can be used as a possible new candidate for $\chi^{(2)}$ cross-induced nonlinear refraction, offering interesting new opportunities, and (ii) similar eigenmodes may be expected in the general case of three-wave mixing (TWM) with three frequencies engaged instead of only two, as in SH generation.

Second-order nonlinear interactions,[6] in particular, parametric amplifiers and oscillators,[7] are some of the best known and widely used processes in nonlinear optics. In general they are viewed as a TWM process, whereby waves at three interacting frequencies ($\omega_1 + \omega_2 = \omega_3$) are phase matched [i.e., $\mathbf{k}_1 + \mathbf{k}_2 \simeq \mathbf{k}_3$, where $k_j = \omega_j n_j/c$ and $\mathbf{k}_j = \mathbf{k}(\omega_j)$ and n_j are wave vectors and linear refractive indices, respectively]. Depending on the intensities $I_j = I(\omega_j)$ of the waves at the incidence boundary, these interactions are regarded as either sum-frequency or difference-frequency generation and, in the degenerate case, $\omega_1 = \omega_2 = \omega_3/2 \equiv \omega$, as either a SH or second-subharmonic generation. All these interactions presume energy exchange between all the waves. In this Letter we explore special TWM regimes in which there is no energy exchange between any of the waves; the interaction is manifested only by linear (with respect to the distance of propagation) phase change in each of the waves as they propagate. They may be regarded as nonlinear eigenmodes of $\chi^{(2)}$ nonlinear interactions.[8] Since Eqs. (1) (see below) in the spatial domain are isomorphic to the equations for electronic parametric oscillators in the time domain, these eigenmodes are counterparts of stationary parametric oscillations in lumped systems.[9]

For the $\chi^{(2)}$ eigenmodes to exist, certain boundary conditions must be arranged among all three intensities (as well as phases) of the waves. The phase property of these eigenmodes amounts to the amplitude-dependent and phase-sensitive change of phase velocity of each of the waves (with their intensities unchanged) as they propagate. This makes the $\chi^{(2)}$ eigenmodes interesting candidates for $\chi^{(2)}$ cross-induced nonlinear refraction emulating the third-order nonlinear refractive index. In contrast to any other TWM (whereby both the phase and the amplitude have a complicated spatial dynamics pattern, including jumplike behavior near exact phase matching), the eigenmodes constitute the only second-order nonlinear process with the unchanging amplitude, and with the phase changing linearly with the distance of propagation, and thus may be the only true example of $\chi^{(2)}$ nonlinear refraction. Furthermore, whereas other modes provide a phase change greater than 2π for only nonideal phase matching,[3,4] the eigenmodes work for arbitrary phase mismatch, in particular, for ideal phase matching. Compared with SH generation cascading,[1-5] the eigenmodes offer broader opportunities, generally engaging more waves with more independently controllable parameters.

If all three cw waves are plane (or confined in a fiber-optic waveguide) and propagate (e.g., along the z axis) in a medium without losses, the spatial dynamics of their electric fields $E_j = E(\omega_j)$ is governed by the equations[6]

$$E_j' = -iq_j E_{3-j}^* E_3 \exp[i(\Delta k)z] \qquad (j = 1, 2),$$

$$E_3' = -iq_3 E_1 E_2 \exp[-i(\Delta k)z], \tag{1}$$

where a prime denotes d/dz, $\Delta k = (k_3 - k_1 - k_2)_z$, and $q_j = \chi^{(2)}\omega_j/cn_j$. The first integral of Eqs. (1) is the familiar Manley–Rowe relation $\omega_1^{-1}I_1' = \omega_2^{-1}I_2' = -\omega_3^{-1}I_3'$, where $I_j = |E_j|^2 n_j c$ are respective intensities. No energy exchange means that $|E_j| =$ constant. By writing the fields in the form $E_j = |E_j|\exp[i\zeta_j(z)]$, where $|E_j|$ and ζ_z are real functions, and by introducing a parameter $\alpha = \chi^{(2)}(cn_1n_2n_3)^{-1/2}$ and vacuum wave vectors $k_{0j} = \omega_j/c$, one transforms Eqs. (1) (with $|E_j| =$ constant) into

$$\zeta_j' = -\alpha k_{0j}\sqrt{I_{3-j}I_3/I_j}\exp(i\Phi) \qquad (j = 1, 2),$$

$$\zeta_3' = -\alpha k_{03}\sqrt{I_1 I_2/I_3}\exp(-i\Phi), \tag{2}$$

0146-9592/93/151223-03$6.00/0

where $\Phi(z) \equiv (\Delta k)z + \zeta_3 - \zeta_1 - \zeta_2$. Since ζ_j's are real, one has $\Phi = 0$ or π; i.e., $S \equiv \exp(i\Phi) = \pm 1$. Equations (2) imply that for eigenmodes, $d\zeta_1/d\zeta_2 =$ constant, $d\zeta_1/d\zeta_3 =$ constant. Thus

$$\zeta_j = \beta_j z + \phi_j \qquad (j = 1, 2, 3), \tag{3}$$

where β_j and ϕ_j are constants; the rates β_j are

$$\beta_j = -\alpha S k_{0j} \sqrt{I_1 I_2 I_3}/I_j;$$

$$\beta_3 - \beta_1 - \beta_2 + \Delta k = 0, \tag{4}$$

and the condition on individual phases is $\phi_3 - \phi_1 - \phi_2 = \Phi = \pi(1 - S)/2$. The phase dynamics equation [Eq. (3)] reflects changes in the respective wave vectors; i.e., $[(k_j)_z]_{\text{eff}} = (k_j)_z + \beta_j$. This amplitude-dependent phase-velocity control is similar to the well-known nonlinear refractive-index effects in $\chi^{(3)}$ materials. The significant difference, though, is that the sign of the change (i.e., self-focusing or self-defocusing) in the case of $\chi^{(2)}$ nonlinear refractive index depends not on the sign of nonlinearity but on the mutual phase arrangement Φ at the incident boundary (see below). [Note also that in the $\chi^{(2)}$ case, the nonlinear change of refractive index is proportional to the amplitude of one of the waves, provided that two others are fixed, instead of the intensity, as in the case of $\chi^{(3)}$ nonlinear refractive index.] The nonlinear refraction in $\chi^{(2)}$ eigenmodes is explained by the fact that with the proper choice of amplitudes (see below) and phases, the nonlinear feedback at each wave [right-hand parts of Eqs. (1)] is purely imaginary, thus resulting only in phase changes. Using Eq. (4), one arrives at the condition on the intensities of all three waves I_j for an $\chi^{(2)}$ eigenmode

$$\alpha S \sqrt{I_1 I_2 I_3}(\omega_1/I_1 + \omega_2/I_2 - \omega_3/I_3) + c\Delta k = 0. \tag{5}$$

In the degenerate case ($\omega_1 = \omega_2 = \omega_3/2$), it becomes

$$\alpha\omega_3(I_3 - I_1) + S \cdot c\sqrt{I_3}\Delta k = 0. \tag{6}$$

In the case of exact phase matching ($\Delta k = 0$), Eq. (5) reduces to

$$\omega_3/I_3 = \omega_1/I_1 + \omega_2/I_2; \tag{7}$$

in the degenerate case ($\omega_1 = \omega_2 = \omega_3/2$) it reduces to

$$I(2\omega) = I(\omega). \tag{8}$$

In the general case ($\omega_1 \neq \omega_2, \Delta k \neq 0$), Eq. (5) is readily solved for one of the intensities (e.g., I_3), provided that two other intensities are given:

$$I_3 = J[F \pm (F^2 - 1)^{1/2}], \tag{9}$$

where $J \equiv \omega_3/(\omega_1/I_1 + \omega_2/I_2)$ and $F \equiv 1 + (\Delta k/\alpha k_{03})^2 J/2I_1 I_2$. In Eq. (9), both the solutions can exist; this situation may be regarded as some kind of amplitude bistability. The way must be found, though, to use this bistability for possible applications. (Note that eigenmodes are not the only modes possible in the system for the same total energy; they can be excited only with certain boundary conditions.) It is worth noting that in the degenerate case, when $I_1 = I_2 = I(\omega)$ and thus $J = I(\omega)$, if the problem is reversed, i.e., if the intensity J is sought as a function of $I_3 = I(2\omega)$, one has $I(\omega) = I(2\omega) \pm \sqrt{I(2\omega)}(\Delta k/\alpha k_{03})$. Thus we have bistable dependence of $J = I(\omega)$ as a function of $I(2\omega)$ only for sufficiently low $I(2\omega)$ [i.e., $I(2\omega) < I_{\max} = (\alpha k_{03}/\Delta k)$]; with $I(2\omega) > I_{\max}$ there is only one possible solution for $I(\omega)$. In the case of exact phase matching, Eq. (9) reduces to Eq. (7) [or to Eq. (8) if $\omega_1 = \omega_2$].

The eigenmodes with $I_3 = 0$ and either $I_1 = 0$ or $I_2 = 0$ are relatively trivial and spatially stable (see below). However, as follows from Eq. (5), the eigenmodes (all of them stable) with $I_1 = I_2 = 0$ and $I_3 \neq 0$ have a limit as to the maximum possible intensity I_3, $0 \leq I_3 \leq (I_3)_{\max} = (c\Delta k/2\alpha)^2/\omega_1\omega_2$, which is exactly a threshold pumping for parametric instability with nonideal phase matching (i.e., when $\Delta k \neq 0$). All the modes (not eigenmodes) with $I_1 = I_2 = 0$ and $I_3 \geq (I_3)_{\max}$ are unstable. Of special interest are photon-balanced eigenmodes, i.e., those having the same number of photons N_j at each of three frequencies ω_j; since $N \propto I_j/\omega_j$, this implies that $I_1/\omega_1 = I_2/\omega_2 = I_3/\omega_3$. In this case we have an exact equality, $I_3 = (I_3)_{\max}$, which suggests again some sort of amplitude bistability mentioned above, since now $I_1 \neq 0$, $I_2 \neq 0$.

From the nonlinear refraction standpoint, the $\chi^{(2)}$ eigenmodes have a peculiar property distinguishing them from $\chi^{(3)}$ nonlinear refractivity; specifically, the nonlinear changes of phase velocity, Eqs. (2) and (4), can be either positive or negative for the same intensities of all of the three waves, depending on the combined phase Φ at the boundary of incidence. Just by switching Φ from 0 to π (which can be achieved, for example, by switching an individual phase ϕ_j at any of three frequencies ω_j), one can reverse the signs of nonlinear refractivities β_j [see Eq. (4)]. [The eigenmodes with both phases are stable (see below).] This property suggests the interesting device applications, phase–phase switching and phase amplification, whereby the change of input phase ϕ_i at any frequency ω_i would result in the change of output phases ζ_i at the distance z at each of the frequencies ω_j by the amount $2\beta_j z$, some of which could be much greater than π. This kind of phase switching and amplifying can be realized even by manipulating the phase of a very weak wave. Considering, for example, ideal phase matching, $\Delta k = 0$, and assuming that $I_2, I_3 \ll I_1$ and $I_3/I_2 = \omega_3/\omega_2$, we find that when ϕ_1 or ϕ_3 is switched by π, the changes of the output phases at frequencies ω_1 and ω_3 are $\Delta\zeta_j = 2|\beta_j z|$ $(j = 1, 3)$ with $|\beta_2| = |\beta_3| = |\alpha/c|\sqrt{\omega_2\omega_3}\sqrt{I_i}$, which can be large because of the strong signal at the frequency ω_1 (in which case $|\beta_1| \ll |\beta_{2,3}|$).

To explore small-perturbation stability of the $\chi^{(2)}$ eigenmodes along the propagation axis (which is sufficient for truly one-dimensional problems, such as nonlinear waveguiding), let us represent a field at each frequency ω_j in the form $\mathcal{E}_j = E_j(1 + \delta_j)$, where E_j is an eigenwave and $|\delta_j| \ll 1$ is a small perturbation, and look for the solution in the form $\delta_j = a_j \exp(\gamma z) + b_j^* \exp(\gamma^* z)$. The system is unstable if any solution for γ has a positive real part. Six linear equations for a_j and b_j ($j = 1, 2, 3$) resulting from Eqs. (1) yield the equation for γ as follows: $\gamma^4(\gamma^2 + \gamma_0{}^2) = 0$, where

$$\gamma_0{}^2 = 2(\alpha/c)^2 I_1 I_2 I_3[(\omega_1/I_2)^2 + (\omega_2/I_2)^2 + (\omega_3/I_3)^2] - \Delta k^2. \tag{10}$$

The solution $\gamma^2 = 0$ is double degenerate; it corresponds to the perturbations that result in the new mode's again being an eigenmode with slightly different amplitudes and/or phases; thus the perturbations are stable. Using Eq. (5), one can prove that, in Eq. (10), $\gamma_0{}^2 \geq 0$ for any eigenmode. Therefore the only nontrivial solution, $\gamma = \pm i\gamma_0$, is imaginary and also corresponds to a stable mode, regardless of whether $\Phi = 0$ or $\Phi = \pi$. Thus, when it is perturbed, an eigenmode will either remain at a new state or slightly oscillate around it with a spatial period $L = 2\pi/\gamma_0$. In the simplest degenerate case with $\Delta k = 0$, $\gamma_0 = 2\alpha k_0(3I)^{1/2}$.

For the pulse applications, in the case of relatively long (e.g., millisecond) pulses, the eigenmodes can be preserved by use of rectangular pulses. Recent advances in the development of high-repetition-rate femtosecond parametric oscillators (see, for example, Ref. 10) suggest ways of overcoming a walk-off effect related to the group-velocity mismatch, even in the picosecond and femtosecond domains. The fact that the wings of a short pulse will slip out of eigenmatching may result in a useful pulse shortening.

In conclusion, we showed that $\chi^{(2)}$ eigenmodes provide an interesting opportunity in nonlinear optics and in applications related to cross-induced nonlinear refraction. In particular, they can be used for amplitude–phase and phase–phase nonlinear control in $\chi^{(2)}$ materials, combining advantages of relatively large $\chi^{(2)}$ nonlinearity with nonlinear refractivity usually found in $\chi^{(3)}$ materials.

This study is supported by the U.S. Air Force Office of Scientific Research. The author is grateful to G. I. Stegeman, E. Van Stryland, and their associates at the Center for Research in Electro-Optics and Lasers (CREOL), University of Central Florida, for most interesting and stimulating discussions during his visit, which was supported by CREOL.

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