Transverse modulational instability of counterpropagating quasi-phase-matched beams in a quadratically nonlinear medium

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We find the threshold and the generation angle for transverse instability of quasi-phase-matched counterpropagating fundamental and second-harmonic beams in a bulk $\chi^{(2)}$ medium. Numerical estimates indicate that the instability should be observable with currently available materials.

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Pattern formation is an active topic in nonlinear optics. Early theoretical investigations demonstrated that cross-phase modulation between counterpropagating beams in media with cubic nonlinearity leads to cooperative, absolute instability[1–3] and to the formation of spatial patterns.[4] This instability was first observed with counterpropagating beams in a bulk medium.[5] Concurrent work revealed similar phenomena in optical cavities containing a passive nonlinear medium.[6–8] The cavity geometry is intrinsically more complex owing to the interplay of linear and nonlinear resonances, which leads to new features, such as pattern formation in the presence of nonlinear loss.[9]

In the past few years pattern formation in parametric $\chi^{(2)}$ media with a nonlinear polarization that is a quadratic function of the optical fields was investigated extensively.[10–13] Convective modulational instability was observed in a forward-propagating traveling-wave interaction.[14] To generate patterns it is necessary to provide feedback such that the system exhibits an absolute instability. One way of introducing feedback is to allow the interacting beams to counterpropagate. Although backward parametric interactions were proposed in the 1960's,[15] there is not sufficient birefringence in available materials for phase matching of a fundamental wave E_1 at frequency ω_1 with a counterpropagating second harmonic E_2 at frequency $\omega_2 = 2\omega_1$. Studies of pattern formation in quadratic media to date have therefore been based on mean-field analysis of an intracavity geometry in which the cavity provides the feedback necessary for an absolute instability.[10–13]

In this Letter we present a study of transverse instability in a bulk quadratic medium of length L without a cavity, with the geometry shown in Fig. 1. The absence of cavity effects allows the transverse instability to be studied in a more basic form. To provide the necessary coupling between counterpropagating beams we consider a backward quasi-phase-matched interaction[16,17] in a periodically poled material with a nonlinear susceptibility of the form $\chi^{(2)} = 2\epsilon_0 d_m \cos k_m z$, where ϵ_0 is the vacuum permittivity and d_m is the effective value of the quadratic susceptibility tensor. When $k_m \simeq k_2 + 2k_1$, where $k_i = \omega_i n_i/c$ (n_i is the refractive index of field E_i and c is the speed of light in vacuum), a forward-propagating beam at ω_1 couples to a backward-propagating beam at ω_2. As was suggested in Ref. 17, transverse instabilities can be excited in this geometry. When the counterpropagating pump beams have equal intensities, we find a simple dispersion relation that describes the presence of an absolute instability for nonzero phase mismatch ($\Delta k = 2k_1 + k_2 - k_m \neq 0$). Quasi-phase matching relaxes restrictions on the choice of wavelength and allows access to the largest components of the electro-optic tensor in a given material. As we show below, threshold powers for observation of the transverse instability by use of, for example, periodically poled $LiNbO_3$ are in the range of a few megawatts and are thus accessible with pulsed lasers.

Counterpropagation of a fundamental field $E_1 = (\mathcal{E}_1/2)\exp[i(k_1 z - \omega_1 t)] + \text{c.c.}$ and its second harmonic $E_2 = (\mathcal{E}_2/2)\exp[i(-k_2 z - 2\omega_1 t)] + \text{c.c.}$ is described by the set

$$\left(\frac{n_1}{c}\frac{\partial}{\partial t} + \frac{\partial}{\partial z} - \frac{i}{2k_1}\nabla_\perp{}^2\right)\mathcal{E}_1 = i\,\frac{\omega_1}{cn_1}\,\frac{d_m}{2} \times \exp(-i\Delta k z)\mathcal{E}_1{}^*\mathcal{E}_2\,, \quad (1a)$$

$$\left(\frac{n_2}{c}\frac{\partial}{\partial t} - \frac{\partial}{\partial z} - \frac{i}{2k_2}\nabla_\perp{}^2\right)\mathcal{E}_2 = i\,\frac{\omega_1}{cn_2}\,\frac{d_m}{2} \times \exp(+i\Delta k z)\mathcal{E}_1{}^2\,, \quad (1b)$$

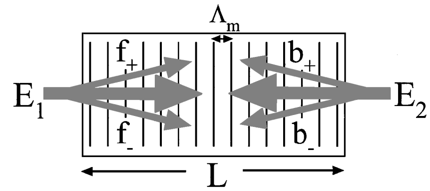

Fig. 1. Interaction of counterpropagating fundamental E_1 and second harmonic E_2, which are beams in a $\chi^{(2)}$ medium poled with period Λ_m. Spatial sidebands of amplitude $f_\pm$ and $b_\pm$ are generated inside the medium.

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where $\nabla_\perp^2$ operates on the transverse coordinates $\mathbf{r} = (x, y)$. Before proceeding it is convenient to rewrite Eqs. (1) in the scaled dimensionless form

$$\left(\frac{\partial}{\partial t} + \frac{\partial}{\partial z} - i\nabla_\perp^2\right)A_1 = A_1^* A_2\,, \tag{2a}$$

$$\left(\frac{\partial}{\partial t} - \frac{\partial}{\partial z} - \frac{i}{2}\nabla_\perp^2\right)A_2 = +i\beta A_2 - A_1^2\,, \tag{2b}$$

where we have made the substitutions $\mathscr{E}_1 = (2cn_1/\omega_1 d_m L)A_1$, $\mathscr{E}_2 = -i(2cn_1/\omega_1 d_m L)A_2 \exp(i\Delta k z)$, $t \to (n_1 L/c)t$, $z \to Lz$, $\mathbf{r} \to \sqrt{L/2k_1}\,\mathbf{r}$, $\beta = \Delta k L$, and $n_2 \approx n_1$.

Equations (2) have stationary plane-wave solutions

$$\overline{A}_1 = a_1 \exp(ia_2 z)\,, \tag{3a}$$

$$\overline{A}_2 = ia_2 \exp(2ia_2 z)\,, \tag{3b}$$

with real amplitudes a_1 and a_2 and positive a_1. Solutions (3) exist provided that the phase mismatch takes the value $\beta = -[2a_2 + (a_1^2/a_2)]$.

We then look for modulational instability, using the ansatz

$$A_1 = \overline{A}_1[1 + f_+(z)\exp(ik_\perp \cdot \mathbf{r}) + f_-(z) \times \exp(-ik_\perp \cdot \mathbf{r})]\exp(\nu t)\,, \tag{4a}$$

$$A_2 = \overline{A}_2[1 + b_+(z)\exp(ik_\perp \cdot \mathbf{r}) + b_-(z) \times \exp(-ik_\perp \cdot \mathbf{r})]\exp(\nu t)\,, \tag{4b}$$

where $\pm\mathbf{k}_\perp$ is the transverse wave vector of the sidebands in scaled dimensionless form. Linearization of Eqs. (2) in the amplitudes of the sidebands $f_\pm$ and $b_\pm$ gives the set

$$\left(\frac{\mathrm{d}}{\mathrm{d}z} + \nu + ik_d\right)f_+ = ia_2(-f_+ + f_-^* + b_+)\,,$$

$$\left(\frac{\mathrm{d}}{\mathrm{d}z} + \nu - ik_d\right)f_-^* = -ia_2(f_+ - f_-^* + b_-^*)\,,$$

$$\left(\frac{\mathrm{d}}{\mathrm{d}z} - \nu - i\,\frac{k_d}{2}\right)b_+ = -i\,\frac{a_1^2}{a_2}\,(2f_+ - b_+)\,,$$

$$\left(\frac{\mathrm{d}}{\mathrm{d}z} - \nu + i\,\frac{k_d}{2}\right)b_-^* = i\,\frac{a_1^2}{a_2}\,(2f_-^* - b_-^*)\,, \tag{5}$$

where $k_d = k_\perp^2$. Equations (5) together with the boundary conditions

$$f_\pm(0) = b_\pm(1) = 0 \tag{6}$$

form a well-posed boundary-value problem for the eigenvalue ν, which is the instability growth rate of the sidebands. The solvability condition for the boundary-value problem defined by Eqs. (5) and (6), subject to the requirement that $\mathrm{Re}(\nu) = 0$, gives an equation for the instability threshold. For general values of the parameters of the problem the resulting expression is cumbersome. In the restricted case of ground-state amplitudes with equal moduli $a_1 = \pm a_2$, and assuming further that at threshold the sidebands are not frequency shifted with respect to the ground state $[\mathrm{Im}(\nu) = 0]$, we find a compact form for the threshold condition:

$$2(2a_2 - k_d)\cos\omega_p \cos\omega_m + \frac{12a_2^3 - 4a_2^2 k_d + a_2 k_d^2 + k_d^3}{\omega_p \omega_m}\sin\omega_p \sin\omega_m + \frac{40a_2^3 - 36a_2^2 k_d + 6a_2 k_d^2 + 9k_d^3}{8a_2^2} = 0\,, \tag{7}$$

where

$$\omega_{p,m} = \left(\frac{-12a_2^2 + 12a_2 k_d + 5k_d^2 \pm \omega}{8}\right)^{1/2},$$

$$\omega = (144a_2^4 - 32a_2^3 k_d - 40a_2^2 k_d^2 + 24a_2 k_d^3 + 9k_d^4)^{1/2}.$$

The dashed curve in Fig. 2 shows a_2 as a function of k_d found by solution of Eq. (7) for the lowest branch of the transverse instability threshold. The minimum threshold $a_2 \simeq 1.94$ occurs for $k_d \simeq 2.79$. Note that in addition to the threshold curve shown in Fig. 2 other solutions of Eq. (7) exist that correspond to higher-lying threshold curves. For negative a_2 Eq. (7) has no solutions. Note that in the cascading limit of large β Eq. (2b) reduces to $A_2 = -(i/\beta)A_1^2$, which allows Eqs. (2) to be rewritten as a single equation for A_1 with an effective cubic nonlinearity that takes the form $-(i/\beta)|A_1|^2 A_1$. The nonlinearity is self-focusing for a_2 positive and self-defocusing for a_2 negative. We can thus state that modulational instability without frequency shifts occurs only under conditions corresponding to a self-focusing nonlinearity.

To verify that the analytic solution obtained for $\mathrm{Im}(\nu) = 0$ corresponds to the lowest threshold, we solved Eqs. (5) and (6) numerically for $a_2 = \pm a_1$ and arbitrary complex ν. It turns out that for $a_2 > 0$ and $k_d < 4.61$ solution of Eq. (7) gives the lowest instability threshold. At $a_2 \simeq 3.26$ and $k_d \simeq 4.61$ a bifurcation occurs, and for larger k_d the lowest instability threshold is obtained in the presence of frequency detuning $[\mathrm{Im}(\nu) \neq 0]$, as shown by the solid and the dotted curves in Fig. 2.

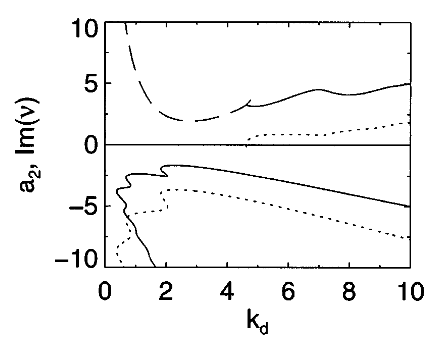

Fig. 2. Pump amplitude a_2 and frequency shifts $\mathrm{Im}(\nu)$ at the threshold for transverse instability. The dashed curve is a_2 found from Eq. (7), and the solid and dotted curves are a_2 and $\mathrm{Im}(\nu)$, respectively, found from numerical solution of Eqs. (5) and (6).

For $a_2 < 0$ all solutions were found numerically from Eqs. (5) and (6), and they are accompanied by frequency shifts as shown in Fig. 2. The lowest instability threshold occurs on the solution branch shown in Fig. 2 at $a_2 \simeq -1.68$ and $k_d \simeq 2.28$. Additional branches with higher thresholds are not shown in the figure, although they cross the depicted branch at several places for $k_d \lesssim 0.7$. Note that for both signs of a_2 the solution branches are duplicated since the sign of $\mathrm{Im}(\nu)$ is arbitrary.

It is of interest to estimate the pump power that is necessary for experimental observation of transverse instability. The pump-beam irradiance is given by $I = (2\epsilon_0 c^3 n_1{}^3/\omega_1{}^2 d_m{}^2 L^2) a_1{}^2$ in watts per meter squared. The characteristic transverse spatial scale of the instability is $\Lambda = 2\pi(L/2k_1k_d)^{1/2}$. Assuming a Gaussian-type pump beam and defining the ratio of the Gaussian beam diameter d_g to the spatial scale Λ by $m = d_g/\Lambda$, we find an expression for the required pump power $P = (\pi^3 m^2 cL/4n_1\omega_1 k_d)I$. Note that m cannot be too small, since that would imply that the generated sidebands do not overlap spatially with the pump beams over the length L of the crystal. Simple geometrical arguments lead to the approximate requirement that $m > (2/\pi)k_d$. Although the minimum instability threshold found in Fig. 2 occurs at $|a_2| \simeq 1.7$, it is accompanied by frequency shifts that can complicate experimental observations. We therefore obtain our estimate on the basis of the slightly higher threshold at $k_d \simeq 2.7$ and $a_1 = a_2 \simeq 1.9$. Assuming $m = 5$, a pump wavelength of 1.06 μm, a crystal length of 1 cm, $n_1 = 2.2$, and $d_m \sim 30$ pm/V, which corresponds to $LiNbO_3$, we find $I \sim 6.4$ MW/cm^2, $\Lambda \sim 75$ μm, and $P \sim 3.5$ kW. This level of pump power is readily available with a nanosecond-pulsed Nd:YAG laser. Note that in the scheme considered here the same pump intensity must be provided at both the fundamental and the second-harmonic frequencies.

Currently available poling techniques cannot, however, meet the requirement that $\Lambda_m = 2\pi/k_m \sim 120$ nm. An alternative is to quasi-phase match the beams to a high order of a longer-period poling. This technique was used recently in an experimental demonstration of backward second-harmonic generation.[18] For square-shaped modulation of the nonlinear coefficient the effective nonlinearity is $d_{m,\mathrm{eff}} = (4/\pi)d_m/p$, where p is the order of the phase matching. A realizable poling period of 3.5 μm gives $p = 29$, $P \sim 1.8$ MW, and peak irradiance $I \sim 3.3$ GW/cm^2; these values are still well below damage thresholds for $LiNbO_3$.

Finally, it should be mentioned that at these irradiance levels two-photon absorption of the second-harmonic beam can increase the threshold for observation of instability. For a second-harmonic beam at 0.53 μm recent measurements of the two-photon absorption coefficient in bulk $LiNbO_3$ indicated a value of $\beta_a = 2.5 \times 10^{-12}$ m/W.[19] Thus at the suggested irradiance of $I = 3.3$ GW/cm^2 the irradiance reduction experienced in a crystal of length $L = 1$ cm is roughly $\exp(-L\beta_a I) = 0.43$, or 57%. Note that the absorption occurs on the second-harmonic beam but not on the counterpropagating fundamental. We expect that the power requirements for experimental observations will be increased by no more than a factor of 2–3.

In conclusion, we have demonstrated the presence of transverse modulational instability of couterpropagating beams in a bulk $\chi^{(2)}$ medium without cavity feedback. By analogy with similar absolute instabilities in media with a $\chi^{(3)}$ nonlinearity, we expect that the nonlinear stage of the instability will result in the formation of spatial patterns. Numerical estimates indicate that the effect should be observable with currently available quasi-phase-matched media.

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