"Explosive instability" and optical generation in photorefractive crystals

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A new type of resonatorless optical generation is predicted, and has been observed, in photorefractive crystals with a circular photovoltaic effect in a frequency-degenerate counterpropagating multiwave interaction.

1. For the counterpropagating four-wave interaction in media with a cubic nonlinearity, the intensities of the signal and conjugate waves grow in proportion to the square of the interaction length, l^2. Over large lengths, energy will sometimes be returned to the pump waves and sometimes retransferred to the signal and conjugate waves.[1,2] Solutions of this sort are known in the time domain in plasma physics and are associated with a decay instability.[3] In the present letter it is shown that in counterpropagating multiwave interactions in photorefractive crystals with a nonlinearity due to circular photovoltaic currents[4] the intensity of the signal wave at the exit from the crystal increases with increasing thickness as $(l - l_{cr})^{-2}$; i.e., this behavior is similar to the buildup of waves over time in the course of an explosive instability.[3] In a crystal with $l \geqslant l_{cr}$, a resonatorless optical generation should arise. This generation has been achieved experimentally in a $LiNbO_3$:Cu crystal.

2. We assume that two pump waves, p and p', are incident on a birefringent photorefractive crystal. These waves are propagating in opposite directions and in directions perpendicular to the polarized signal wave s, which lies in a plane perpendicular to the C axis of the crystal. As a result of the interaction, the following waves may also appear: (a) only a single wave s', which is the conjugate of the signal wave; (b) only a single wave, which is a phase-conjugated intermediate wave i, symmetric with respect to the pump wave; (c) three additional waves, specifically, a conjugate wave s' and a pair of mutually conjugate intermediate waves i and i'. If the last case (c) is to be achieved, the following matching conditions must be satisfied:

$$2\mathbf{k}_p = \mathbf{k}_s + \mathbf{k}_i; \quad 2\mathbf{k}_{p'} = \mathbf{k}_{s'} + \mathbf{k}_{p'}; \qquad (1)$$

$$\mathbf{k}_{p'} + \mathbf{k}_p = \mathbf{k}_{s'} + \mathbf{k}_s = \mathbf{k}_{i'} + \mathbf{k}_i. \qquad (2)$$

For a copropagating parametric interaction (b), a single pump wave and the satisfaction of matching condition (1) are sufficient. Finally, the achievement of the counterpropagating vector four-wave interaction (a) requires that the signal wave be far from matching conditions (1) but that condition (2) hold for the wave s,s' and p,p'. These processes are shown schematically in Fig. 1.

3. The simplified equations for the complex amplitudes (A) of the signal wave and the waves which are produced can be written in the approximation of a given field of the pump wave ($A_s, A_{s'}, A_i, A_{i'}, \ll A_p, A_{p'}$) as follows:

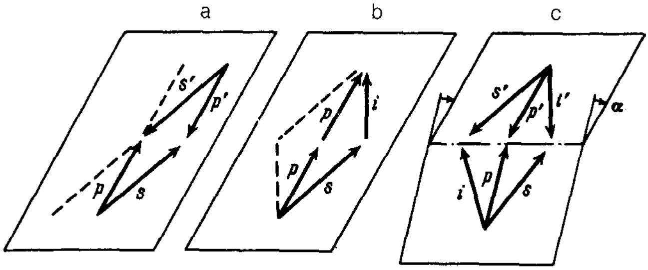

FIG. 1. a—Vector matching for the counterpropagating four-wave parametric interaction; b—for the copropagating three-wave parametric interaction; c—for the six-wave interaction. Processes a and c lead to an "explosive" singularity, but process b does not.

$$\frac{dA_s}{dx} = \gamma'' D(x) A_p, \quad \frac{dA_{s'}^*}{dx} = \gamma'' D(x) A_{p'}^*,$$
$$\frac{dA_i^*}{dx} = -\gamma'' D(x) A_p^*, \quad \frac{dA_{i'}}{dx} = -\gamma'' D(x) A_{p'}, \tag{3}$$

where

$$D(x) = A_p^* A_s + A_{p'}^* A_{s'} - A_p A_i^* - A_{p'} A_{i'}^*,$$

$$\gamma'' = \omega n^3 r_{51} \beta_{15}'' \cos\theta (2c\sigma)^{-1}.$$

Here r is a component of the electrooptic tensor, θ is the angle between the signal wave and the pump waves, and σ is the photoconductivity of the crystal. Equations (3) incorporate the effects of only the circular, spatially oscillating current, which is associated with an antisymmetric component of the photovoltaic tensor,[4,5] β_{15}''. This (nonlocal) nonlinearity is predominant for the $LiNbO_3$ crystals used in the experiments, and this nonlinearity leads to qualitatively new effects in the multiwave interactions.

The first term on the right side of each equation in (3) is responsible for a direct exchange of energy between a pump wave and a weak wave upon diffraction from a displacement grating.[4] The third term is responsible for the copropagating parametric interaction,[6,7] and the second and fourth terms are responsible for a counterpropagating parametric interaction. The solution of system (3) for the normalized intensity of the signal wave, $T = |A_s(e)/A_s(0)|^2$, is

$$T = \left[\frac{(1-2q^2)\exp[\Gamma l(1-q^2)(1+q^2)^{-1}] + 1}{2\{1 - q^2 \exp[\Gamma l(1-q^2)(1+q^2)^{-1}]\}} \right]^2, \tag{4}$$

where $q^2 = |A_{p'} A_p^*|^2$, and the ratio of the pump wave intensities is $\Gamma = 2\gamma'' |A_p|^2 (1+q^2)$.

In particular case (b), i.e., with $q = 0$, expression (4) reduces to the well-known expression for the parametric amplification of the signal beam.[7] In the case $q \neq 0$, new

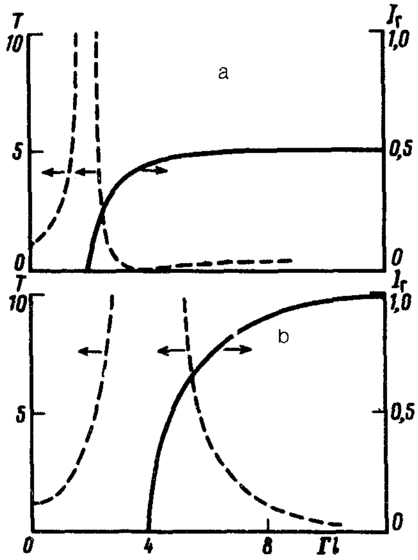

FIG. 2. Transmission of the signal beam, T (dashed lines), and intensity of the resonatorless generation, I_g (solid lines), as functions of the coupling constant Γl for (a) the six-wave interaction and (b) the four-wave interaction for the same intensities of the pump waves.

solutions, of an "explosive" type, appear. For these new solutions, the intensity of the new waves increases without bound at a finite value of the coupling constant Γl. For identical pump wave intensities, $q = 1$, we thus find T_6 for the six-wave interaction and T_4 for the four-wave interaction to be

$$T_6 = \left[\frac{1-(\Gamma l/4)}{1-(\Gamma l/2)}\right]^2, \quad T_4 = \left[\frac{1}{1-(\Gamma l/4)}\right]^2.$$

For a six-wave interaction, in which three distinct processes simultaneously result in an intensification of the signal wave (the direct two-wave energy exchange and the counterpropagating and copropagating parametric processes), the "explosion" occurs at a value $l_{cr} = 2/\Gamma$, lower than for the four-wave process, in which there is no copropagating parametric amplification, $l_{cr} = 4/\Gamma$ (dashed lines in Fig. 2).

Although there is a formal analogy between the effect which we are discussing here and the explosive instability in a plasma, there are also some important distinctions. The unbounded increase in the wave intensity, proportional to $(t-t_0)^{-1}$ in the case of the explosive instability in a plasma, follows from the solution of nonlinear equations describing a system containing waves with negative energy.[3] On the other hand, the result that the intensities of the signal and conjugate waves become infinite at $l = l_{cr}$ follows from the solution of the system of linear equations (3) and is a consequence of the ill-posed nature of the problem, in which the boundary conditions on the counterpropagating waves are specified at different entrance faces of the crystal[8]: $x = 0$ and l. The steady-state solution of the complex system of nonlinear equations shows that the "explosive" singularity in the approximate solution corresponds to the threshold for the onset of resonatorless generation. The intensity of the genera-

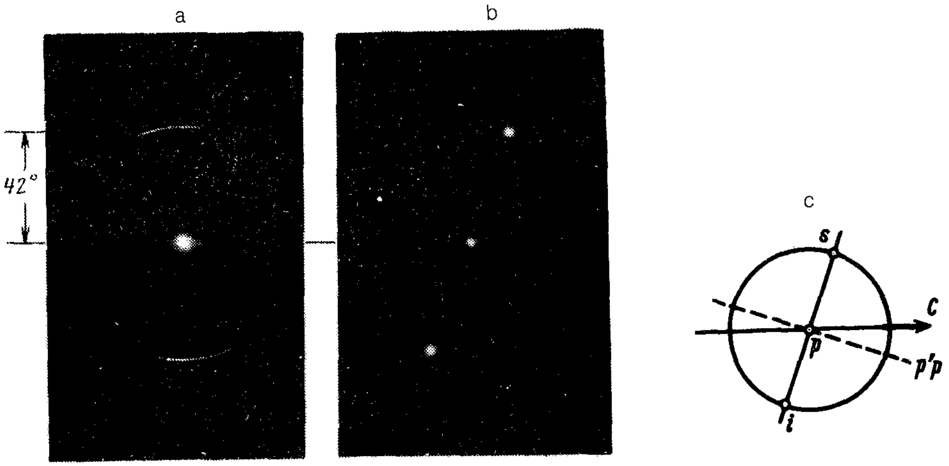

FIG. 3. Spatial distribution of the light waves emerging from the sample (a) in the initial stage of the exposure and (b) in the steady state. Diagram c shows a cross section of the matching cone and the positions of the planes containing the pump wave p, p' and the generation waves s,i.

ted beams, $I_\Gamma = |A_{s'}(0)/A_{p'}(l)|^2$ with $q^2 = 1$, is given for interactions (a) and (b) by (Fig. 2).

$$(\Gamma l)_4 = 2\sqrt{I_g}\,\ln\left[(1+\sqrt{I_g})(1-\sqrt{I_g})^{-1}\right], \tag{5}$$

$$(\Gamma l)_6 = \sqrt{2I_g}\,\ln\left[(1+\sqrt{2I_g} + \sqrt{1-2I_g})(1-\sqrt{2I_g}+\sqrt{1-2I_g})^{-1}\right]. \tag{6}$$

4. Experimentally, we have detected resonatorless parametric generation in a counterpropagting six-wave interaction (c) in $LiNbO_3$:Cu (0.02 wt. %) crystals 0.15 cm thick. The generation is excited by the unfocused beam from a single-frequency, single-mode argon laser (an e-wave in the crystal). The counterpropagating pump beam is formed by reflection from the opposite face of the crystal. At a power density on the order of 1 W/cm^2, an intense parametric scattering (b) appears after a few tens of seconds (Fig. 3a). A pair of brighter beams then begins to form at the surface of matching cone (1); as these beams intensity, they suppress the original scattering (b) almost completely (Fig. 3b).

The angular position of the generation beams is determined by matching conditions (1) and (2); the wave triads s, p, i and s', p', i' are propagating in respective planes; both of these planes are perpendicular to the plane in which the pump beams converge. Consequently, by varying the plane of misadjustment of the sample we were able to achieve generation in an arbitrary direction along matching cone (1). An exceptional case arises when the sample is tilted in the plane normal to the C axis of the crystal. In this case, the wave vector of the dynamic gratings corresponding to matching conditions (1) and (2) is oriented along the C axis, i.e., in a direction for which the circular photovoltaic current is zero,[4,5] and generation becomes impossible.

A strict spatial localization of the generation beams when there is a small angle between the pump beams and also the lower threshold for self-excitation are advantageous distinctions of the six-wave arrangement, in which it has proved possible to achieve this type of generation for the first time.

The generation is quite stable with respect to variations in the pump conditions. In our experiments, for example, this generation was observed at a pump-intensity unbalance $q^2 \cong 1:6$. This result agrees qualitatively well with a calculation according to which the generation threshold increases by a factor of only two at $q^2 = 1:50$.

In summary, this new nonlocal nonlinearity, associated with the circular photovoltaic effect,[4] is distinguished from the known[9] diffusive nonlocal nonlinearity in that the nature of the counterpropagating multiwave interactions is radically different. This new nonlinearity has made it possible to achieve resonatorless generation of new coherent light beams with a controllable spatial position.

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