

Parametric generation in media with local response in unclosed resonators

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Steady-state generation has been achieved in counterpropagating four-wave mixing in an iron-doped lithium niobate crystal in a two-sided mirror layout and in a ring layout. A new calculation shows that generation can occur in a medium with a nonlinear local response when the excited waves are tuned away from the pump waves in terms of angle and frequency. The possibility of phase conjugation in the two-sided-mirror layout due to an additional frequency shift of one of the pump waves is discussed.

Research over the past decade on frequency-degenerate four-wave mixing of various types has revealed that generation can be arranged in unclosed resonators: in a semiopen linear resonator, in a ring resonator, and in a resonator of the cat-conjugation type.¹ It can also be arranged without any resonator at all, in layouts with two-sided conjugating mirrors.² It is generally believed that a necessary condition for steady-state generation in such layouts is a purely nonlocal linear response,³ i.e., a spatial shift of the phase grating in the crystal with respect to the interference pattern responsible for the writing.¹⁾

In this letter we are reporting the first observation of steady-state generation in a photorefractive crystal with a local nonlinear response in a layout with a two-sided conjugating mirror and in a ring layout. We also report the results of a calculation which show that generation can occur in any medium which has a local response if (1) the generated wave is detuned in angle to restore phase matching and (2) the frequency degeneracy is lifted.

The generation was achieved in the layouts shown in Fig. 1, through the use of the cw beam, consisting of a single mode in terms of transverse index, from an argon or helium-cadmium laser. The electric vectors of the pump waves (4 and 2) and those of the excited waves (3 and 1) oscillated in the plane of the figure. The optic axis of the crystal lay in the same plane. The *Y*-cut, iron-doped lithium niobate samples had dimensions of $6 \times 6 \times 3$ mm. The laser beams used for the pumping were not expanded.

In the two-sided mirror layout (Fig. 1a), pump waves 4 and 2, which are symmetric with respect to the *z* axis, are formed by two independent lasers. A diffuser pressed tightly against the crystal on the $z = l$ side increases the divergence of wave 2 to 3° inside the crystal. Under these conditions, excited beams 1 and 3 appear after 10 min, as a consequence of a diffraction of waves 2 and 4 by the transmitting phase grating which arises spontaneously in the crystal. In the time evolution of the onset of this generation we observe a prolonged prethreshold stage, an intense peak when generation arises, and then a transition to a quasisteady state is a fraction of 1%.

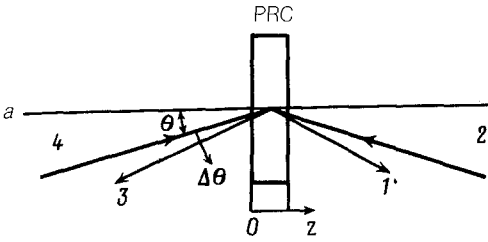
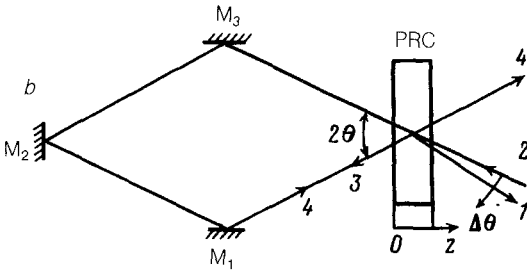


FIG. 1. *a*—Two-sided mirror layout; *b*—ring-generator layout. PRC) Photorefractive crystal.



The angular divergence of wave 3 and that of wave 1 after passage through an aberrator are approximately the same: $15'$ in the plane in which the beams converge and larger by a factor of about three or four in the perpendicular plane. Consequently, after wave 1 has undergone cancellation (after passage through the aberrator), its divergence is an order of magnitude smaller than that of wave 2 in the crystal, although it is not restored to the original divergence of wave 2.

Each of the excited waves propagates along a direction which is not exactly opposite that of the pump wave. The angular deviation $\Delta\theta$ increases as the pump beams approach an antiparallel orientation (i.e., as the angle θ is reduced; Fig. 2). In all the crystals used, with various iron contents, the generated waves deviate from the normal to the crystal surface by an angle larger than that of the pump waves; i.e., $\Delta\theta$ is positive.

Generation with corresponding characteristics was achieved in a ring arrangement (Fig. 1b), with the distinction that in this case it was not necessary to use an aberrator to produce pencil beams.

The fact that there is no sensitivity with respect to a reversal of the orientation of the polar axis of the crystal convinces us that the primary amplification mechanism in each case is the local response (which is dominant in $\text{LiNbO}_3:\text{Fe}$), not a possible contribution of a diffusion nonlocal nonlinearity.³

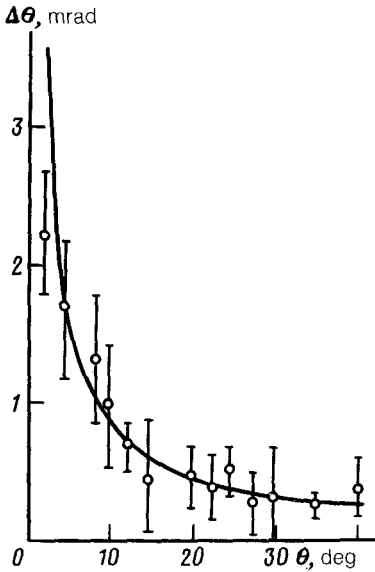


FIG. 2. Points—Angular detuning of the excited wave, $\Delta\theta$, versus the angle between the pump waves, θ , for an $\text{LiNbO}_3:\text{Fe}$ (0.02% by mass) sample; solid line—theoretical approximation.

To analyze the possibility of generation, we solved the standard system of simplified equations for the complex amplitudes A_i of the waves which mixed in the crystal (Ref. 1, for example). We used the approximation of a given pump wave field: $A_2 = \text{const}$, $A_4 = \text{const}$. Under these conditions, one can transform to a system of equations which describe the ratio of the amplitudes of the weak (excited) waves, $\eta(z) = |A_1/A_3|$, and the generalized phase $\phi(z) = \varphi_1 + \varphi_2 - \varphi_3 - \varphi_4 - \Delta k_z z$, which incorporates a possible angular deviation $\Delta k_z = (4\pi\Delta\theta\cos\theta\sin\theta/\lambda\sqrt{n^2 - \sin^2\theta})$ of the mixing waves

$$\frac{d\eta}{dz} = -\gamma''\eta - \frac{\gamma''\sqrt{R}}{R+1}(1+\eta^2)\cos\phi - \frac{\gamma'\sqrt{R}}{R+1}(1-\eta^2)\sin\phi,$$

$$\frac{d\phi}{dz} = \frac{\gamma''\sqrt{R}}{R+1}(1-\eta^2)\sin\phi - \frac{\gamma'\sqrt{R}}{R+1}(1+\eta^2)\cos\phi - \gamma'\eta - \Delta k_z z. \quad (1)$$

Here γ' and γ'' are the coupling constants for the local and nonlocal responses, respectively. We derived new generation solutions for a response of a mixed type. We assumed that the constants γ'' and γ' are related to the initial, purely local constant $\gamma^0 = (\pi r_{33} n_c^3 \beta_{33} / \kappa \lambda \cos\theta)$ (r_{33} and β_{33} are the components of the electrooptic and photovoltaic⁷ tensors, κ is the photoconductivity constant, and λ is the wavelength):

$$\gamma' + i\gamma'' = \frac{1}{1 + (\tau\delta\omega)^2} + \frac{i\tau\delta\omega}{1 + (\tau\delta\omega)^2}, \quad (2)$$

where τ is the Maxwellian relaxation time, and $\delta\omega = \omega_1 - \omega_4 = \omega_3 - \omega_2$ is the frequency deviation.

Under the assumption $\eta(z=0) = 0$ (wave 1 is not given at the entrance face, $z=0$), we can derive $\phi(0) = \tan^{-1}(\gamma'/\gamma'')$ from the second equation and then calculate the functional dependences $\eta(z)$ and $\phi(z)$ by the Runge-Kutta method. It is easy to show that if the sign of $\delta\omega$ is chosen correctly the quantity η will approach infinity at a finite critical thickness $z = l_{\text{thr}}$, which corresponds to mirrorless generation. The generation threshold in the ring arrangement is calculated from the condition $\eta = 1/\sqrt{R}$. A necessary condition for generation in this case is that the nonlinear detuning, which arises because of the presence of a local response, be cancelled by a geometric detuning: $\Delta k_z = -\gamma'$.

Some numerical estimates support the arguments above. Working from independent measurements concerning the kinetics of the non-Bragg erasure of the diffraction grating, we measured the photoconductivity constant, $\kappa = 1.6 \times 10^{-13} \text{ cm}/(\Omega \cdot \text{W})$. Working from the kinetics of the rise of the diffraction efficiency during the writing of a grating by two beams, we measured the photovoltaic constant: $\beta_{33} = 3.2 \times 10^{-9} \text{ A/W}$. From these figures we can find the optimum frequency shift of the excited wave, $\delta\omega = 3 \times 10^{-5} \text{ Hz}$, and the nonlocal coupling constant, $\gamma' = 30 \text{ cm}^{-1}$.

Experimentally, it is not possible to measure such a small frequency shift against the background of the instrumental noise stemming from the mechanical instability of the apparatus. On the other hand, by fitting the theoretical function $\Delta\theta = -(\gamma'\sqrt{n^2 - \sin^2\theta}/4\pi\sin\theta\cos\theta)$ to the experimental points in Fig. 2, we find the constant $\gamma' = 20 \pm 5 \text{ cm}^{-1}$. This figure agrees well with the result of the direct measurements. We wish to stress that the nonlinear change in the refractive index, Δn (and thus that in the interaction constant γ'), is negative in a crystal with a photovoltaic charge transport, so we would expect positive values of Δk_z and $\Delta\theta$, in complete agreement with the experimental observation.

It follows from these calculations and experiments that this generator is not a strictly phase-conjugating mirror, because of the angular deviation of the excited waves from the pump waves [see (3)]. This angular deviation is small in absolute value (on the order of a milliradian), so that one can largely compensate for the aberrations introduced in one of the pump beams by the diffuser pressed against the entrance face of the crystal. By varying the frequency of one of the pump waves, however, we can introduce an additional wave detuning and completely cancel the angular detuning for one of the excited waves. The necessary frequency detuning can be found from the relation $(\Delta\lambda/\lambda) = (\Delta n/n)\cot^2\theta$, where $\Delta n = \gamma'\lambda/2\pi$ is the nonlinear local change in the refractive index. For $\theta = 30^\circ$, $\lambda = 0.44 \mu\text{m}$, and $\Delta n \leq 10^{-3}$, a value $\Delta\lambda \leq 6 \text{ \AA}$ is sufficient for this cancellation. Media with a local response are thus suitable for developing a single-sided (not two-sided, as in the case of media with a nonlocal response) conjugating mirror with a mixing of two oppositely directed, mutually incoherent light beams.

¹⁾Time-varying generation in ring arrangements was discussed in Refs. 4 and 5.

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