Lithium niobate laser with frequency-degenerate pumping

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For the first time, lasing has been achieved in a frequency-degenerate four-wave interaction of beams from a helium-cadmium laser in lithium niobate crystals with nonlocal nonlinearities. The nonlinearities result from a redistribution of space charge during diffusion of the photoexcited carriers or from photovoltaic currents that oscillate over space.

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1. The interaction of two coherent light beams in a medium with a nonlocal response gives rise to a steady-state energy exchange even in the frequency-degenerate case.[1] This effect underlies the amplification of coherent light beams when they are diffracted by a dynamic grating hologram which they themselves record (see Ref. 2, for example), with a gain Γ ranging from tens of reciprocal centimeters ($LiNBO_3$, $LiTaO_3$) to hundreds of reciprocal centimeters ($BaTiO_3$, $SrBaNbO_3$).[2–4] The uniquely high gain in $BaTiO_3$, $\Gamma \gtrsim 10^2$ cm^{-1}, has recently made it possible to achieve self-

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pumped oscillation in this material in an external resonator with one coherent pump beam.[5–7] In the present letter we describe a similar oscillation achieved in iron-doped lithium niobate crystals pumped with a cw beam from a He-Cd laser. In the present experiments we make use of the same "diffusion" nonlinearity[8,9] as in $BaTiO_3$ crystals or a nonlocal nonlinearity, peculiar to activated crystals, which results from spatial oscillations of the photovoltaic currents.[10–12] Arrangements with both transmission and reflection dynamic gratings turn out to be capable of oscillation.

2. In order to achieve oscillation, it is necessary to arrange positive feedback and to offset the loss by means of an amplification (phase and amplitude conditions). In the present case the resonator is formed by the polished faces of the crystal (reflection coefficients $R = 15\%$ and 17% for the waves with the ordinary and extraordinary polarization). The pump beam is focused into the sample by a lens with a focal length of 25 cm. The nonlinear lens which arises in the illuminated region promotes the formation of high-Q modes with frequencies precisely equal to the pump frequency.

The threshold condition for a holographic laser is

$$R_1R_2\exp[(\Gamma_t - 2\alpha - 2\gamma)l] = 1, \tag{1}$$

where R_1 and R_2 are the mirror reflection coefficients, Γ_t is the threshold gain, α is the absorption coefficient, γ represents the other types of loss, and l is the thickness of the sample. Condition (1) reflects the circumstance that in the arrangements used the amplification occurs in only a single pass because of the anisotropy of the mechanism by which the gratings are recorded and because of the asymmetry of the pumping. For our laser we have $\gamma \ll \alpha \simeq 1\ \text{cm}^{-1}$; hence $\Gamma_t \simeq 11\ \text{cm}^{-1}$.

3. In photorefractive crystals the gain is

$$\Gamma = 2\pi\Delta n/\lambda\cos\theta = -\pi r n^3 E_{sc}/\lambda\cos\theta, \tag{2}$$

where r and n are the corresponding components of the linear electrooptic tensor and the refractive-index tensor, λ is the wavelength, and 2θ is the angle at which the beams meet in the medium. In contrast with the situation in ordinary media with a cubic nonlinearity, the magnitude of the refractive-index modulation, Δn, does not depend on the light intensity but does depend on the space-charge field E_{sc}. For a diffusive nonlinearity, we have $E_{sc}^d = 2\pi kT/e\Lambda$, where e is the electron charge, and $\Lambda = \lambda/2n\sin\theta$ is the period of the grating. A gain $\Gamma \simeq 10^2\ \text{cm}^{-1}$ has been achieved in $BaTiO_3$ crystals in a comparatively weak diffusion field, $E_{sc}^d = 1.2$ kV/cm ($2\theta = 8°$), by virtue of the exceedingly high electrooptic coefficient $r_{42} = 920 \times 10^{-10}$ cm/V. In order to achieve the necessary gain levels in the case of the diffusion nonlinearity in lithium niobate, with a maximum electrooptic coefficient $r_{33} = 30 \times 10^{-10}$ cm/V, it is necessary to reduce Λ.

In an arrangement in which the transmission gratings are recorded in a Y-cut crystal, the grating vector lies in a plane which contains the C axis of the crystal, and the pump wave and the excited wave are extraordinarily polarized (Fig. 1a). For incidence of the pump beam on the crystal at the Brewster angle, $2\theta = 22°$, the calculated value is $\Gamma = 18\ \text{cm}^{-1}$. The strongest diffusion field, $E_{sc}^d = 19$ kV/cm at $\Lambda = \lambda/2n = 0.09\ \mu$m, arose during the recording of reflection gratings in Z-cut crystals (Fig.

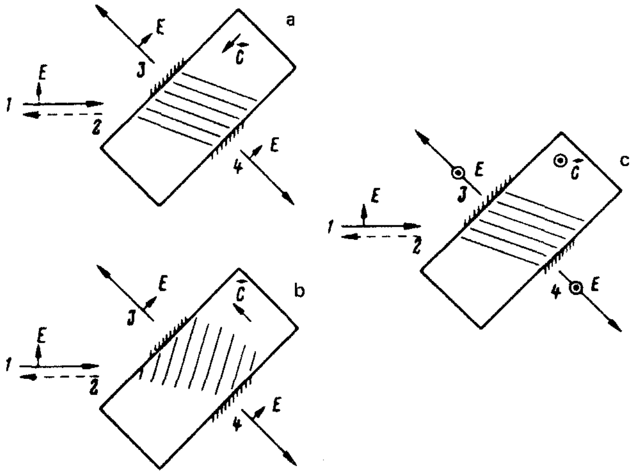

FIG. 1. Arrangement for lasing in $LiNbO_3$:Fe. Initially, only pump wave 1 is incident on the crystal; in the steady state, waves 3 and 4 appear (the excited waves), along with wave 2, which is propagating opposite the pump wave.

1b). Although we have $r_{13} = 8.6\times 10^{-10}$ cm/V in this configuration, the calculated value if $\Gamma = 64$ cm^{-1}

Another way to achieve high steady-state gain levels in lithium niobate is to use the strong fields E_{sc}^{phg} which result from the excitation of spatially oscillating currents by virtue of the imaginary component of the photovoltaic tensor,[10,11] β_{15}^{a} (the arrangement in Fig. 1c):

$$E_{sc}^{phg} = -j^{phg}/\sigma = -(\beta_{15}^{a}\sqrt{I_1 I_2}\cos\varphi)/\sigma\,, \qquad (3)$$

where I_1 and I_2 are the intensities of the orthogonally polarized beams, and $\sigma = \kappa(I_1 + I_2)$ is the photoconductivity of the crystal. The angle (ϕ) between the current direction and the grating vector is $\varphi = \mathrm{arctg}\times[(n_0 - n_e\cos 2\theta)/n_e \sin 2\theta]$. According to Ref. 12, we have $\beta_{15}^{a} \simeq 10^{-9}$ A/W; hence, using $r_{51} = 28\times 10^{-10}$ cm/V for $2\theta = 10°$ ($\varphi = 20°$), we find $\Gamma = 20$ cm^{-1}.

Consequently, threshold condition (1) should hold with an ample margin in all the oscillation schemes considered (Fig. 1).

4. Oscillation was achieved in all three of these arrangements in plane-parallel samples of reduced lithium niobate crystals with a thickness of 0.4 cm and an iron impurity of 0.03% by weight. In all cases we first observed a sharp intensification of the photo-induced scattering of the pump beam—an analog of superradiance in ordinary laser systems. Then, against the background of this scattered light, a brighter spot corresponding to the lasing beam gradually developed. This beam was oriented along the normal to the ends of the sample. The intensity of the light scattered at other angles fell off sharply. The angular divergence of the excited beam was $\sim 5°$ in most cases, but it had a structure and frequently contained a distinct core with a divergence near the divergence of the pump beam, $\simeq 30'$.

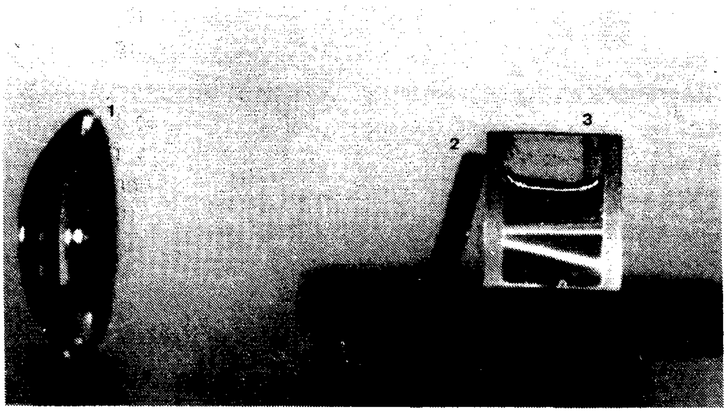

FIG. 2.

Figure 2 shows the lasing arrangement corresponding to Fig. 1c. The pump beam, polarized in the plane of the figure, propagates from left to right and is focused by lens 1 onto tilted crystal 2, whose optic axis is oriented along the normal to the figure. On the table behind the crystal is a cell (3) holding a dye solution, in which we can see the paths traced out by the transmitted pump beam and by the excited beam, which propagates along the normal to the surfaces of the sample. We see that the excited beam has the greater part of the pump intensity (35% of the intensity incident on the crystal at the angle $2\theta = 10°$, in comparison with 4% in the transmitted pump beam).

This approximately steady state is interrupted from time to time by abrupt changes in the intensity of the transmitted pump beam and the excited beams and by a comparatively rapid return to the original state. The most probable reason for these interruptions is dielectric breakdown in the sample due to the buildup of large-scale space-charge fields,[13,14] but we do not rule out the possibility that the interruptions are due instead to the multivaluedness of the solutions for the intensity of the conjugate wave in the four-wave interaction (an optical bistability).[15,16]

5. In the case of a frequency-degenerate counterpropagating interaction, the amplitude of the fourth wave is proportional to the product of the amplitudes of the pump waves, E_1E_2, and the signal wave, E_3^*. In the case at hand, the waves E_3 and E_4 convert into each other upon reflection from the resonator mirrors; i.e., the weak-signal diffraction efficiency of the dynamic grating that arises depends linearly on the prevailing intensity in the mode being excited. Here there is analogy with stimulated emission in a medium with a population inversion, where the probability for the stimulated process is proportional to the number of photons in the mode. This analogy becomes even more complete when we note that each photon which is produced is emitted in the same direction, set by the condition for Bragg diffraction, and with the appropriate phase, since for nonlocal nonlinearity mechanisms all four of the interacting waves are related by the simple phase condition

$$\varphi_4 = -\varphi_3 + \varphi_1 + \varphi_2 + p\pi \text{, where } p = 0.1 \quad \text{for} \quad \Gamma \gtrless 0, \tag{4}$$

regardless of their intensity ratios.[17]

The laser which is most closely related to this one in terms of physical principles is the Brillouin-scattering laser, in which the diffraction is caused by traveling dynamic gratings, so that a frequency shift arises.

The output frequency of this holographic laser is precisely equal to the pump frequency, but it can also be tuned over a broad range (over essentially the entire visible range for lithium niobate with a diffusive recording process). Furthermore, for these lasers there are completely definite gain bands in terms of the spatial frequency, i.e., intervals of angles in which a specific nonlinearity mechanism provides an adequate gain.

We wish to emphasize that lasers operating by dynamic-holography principles open up some new opportunities for correcting the wave-fronts of laser beams in real time and for efficiently exciting beams with a given spatial-angular structure.

[1]D. I. Stasel'ko and V. G. Sidorovich, Zh. Tekh. Fiz. **44**, 580 (1974).
[2]V. Markov, S. Odoulov, and M. Soskin, Opt. Laser Tech. **11**, 95 (1979).
[3]J. Feinberg and R. Hellwarth, Opt. Lett. **5**, 519 (1980).
[4]V. Voronov, I. Dorosh, Yu. Kuz'minov, and N. Tkachenko, Kvant. Elektron. (Moscow) **7**, 2313 (1980) [Sov. J. Quantum Electron. **10**, 1346 (1980)].
[5]J. O. White, M. Cronin-Golomb, B. Fisher, and A. Yariv, Appl. Phys. Lett. **40**, 450 (1982).
[6]M. Cronin-Golomb, B. Fisher, J. Nilsen, J. O. White, and A. Yariv, Appl. Phys. Lett. **41**, 219 (1982).
[7]M. Cronin-Golomb, B. Fisher, J. O. White, and A. Yariv, Appl. Phys. Lett. **41**, 689 (1982).
[8]D. L. Staebler and J. J. Amodei, J. Appl. Phys. **43**, 1042 (1973).
[9]N. Kukhtarev, V. Markov, S. Odoulov, M. Soskin, and V. Vinetski, Ferroelectrics **22**, 949 (1979).
[10]V. Belinitcher, Phys. Lett. **66A**, 213 (1978).
[11]B. I. Sturman, Kvant. Elektron. (Moscow) **7**, 483 (1980) [Sov. J. Quantum Electron. **10**, 276 (1980)].
[12]S. G. Odulov, Pis'ma Zh. Eksp. Teor. Fiz. **35**, 10 (1982) [JETP Lett. **35**, 10 (1982)].
[13]E. M. Avakian, K. G. Bielabaev, A. A. Kaminski, and V. K. Sarkisov, Phys. Status Solidi **A36**, K25 (1976).
[14]K. K. Shvarts, Izv. Akad, Nauk SSSR, Ser. Fiz. **41**, 788 (1977).
[15]H. Winful and J. Marburger, Appl. Phys. Lett. **36**, 613 (1980).
[16]N. V. Kukhtarev and V. N. Starkov, Pis'ma Zh. Tekh. Fiz. **7**, 692 (1981) [Sov. Tech. Phys. Lett. **7**, 296 (1981)].
[17]N. V. Kukhtarev and S. G. Odulov, Pis'ma Zh. Eksp. Teor. Fiz. **30**, 6 (1979) [JETP Lett. **30**, 4 (1979)].

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