Parametric generation and phase conjugation of intersecting laser beams in a layer of a nematic liquid crystal containing a dye

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Parametric generation has been obtained for stimulated forward scattering of counterpropagating beams of radiation from a Nd^{3+}:YAG laser in a layer of nematic liquid crystal containing a dye. Calculations show the possibility of self-pumped phase conjugation of the wave front of a beam with a diffraction spreading length that is comparable to the dimension of the region of self-intersection in the nematic liquid crystal with a thermal nonlinearity. Self-pumped phase conjugation of the wavefront of quasi-cw radiation has been obtained.

Investigations of parametric generation in the field of intersecting light beams have demonstrated the possibility of obtaining self-pumped phase conjugation and mutual conjugation of low-intensity cw laser beams in media with photorefractive nonlocal nonlinearity.[1-3] The possibility of observing these effects in other nonlinear media is an open question. In this paper we report the first observation of parametric generation and phase conjugation of counterpropagating beams (intersecting at a small angle θ_n) of a quasi-cw Nd^{3+}:YAG laser as a result of their combined Rayleigh forward scattering by the thermal nonlinearity of a nematic liquid crystal with a dye near the nematic-isotropic liquid phase transition.

Plane-polarized radiation consisting of a single mode in the transverse index from a cw Nd^{3+}:YAG laser with an output power $P_0 < 2$ W was modulated by means of a chopper (the pulse length τ_m was varied from 0.5 to 5 μs and the repetition period was 100 τ_m) and focused into the nematic liquid crystal cell. Counterpropagating beams were formed either by means of the specularly reflecting wall of the nonlinear layer (in cells with a liquid crystal layer thickness $\lambda = 1$, 0.5, or 2 mm), or with an external semitransparent mirror, with subsequent focusing of the beams into a cell with transparent walls and $l = 1$ mm. The planar orientation of the nematic mixture in tolanes with a dye, having an absorption $\alpha l \approx 0.4$, in a cell with $l = 0.5$ mm, was fixed by a surface orientant. The homeotropic orientation in cells with $l = 1$ and 2 mm, filled with a nematic mixture based on 5CB with a dye ($\alpha l \approx 0.8$ and 0.5, respectively), was obtained by means of an external ac electric field produced by a voltage of 30 to 500 V at a frequency of 50 Hz, applied to the current-carrying walls. When the pump power exceeded a threshold level, which depends on the spatial structures of the beams and the temperature of the cell, we observed in all the cells an abrupt increase in the backward-propagating scattered radiation (the lower oscilloscope trace in Fig. 1a), which appears after a delay time relative to the beginning of the pump pulse (the

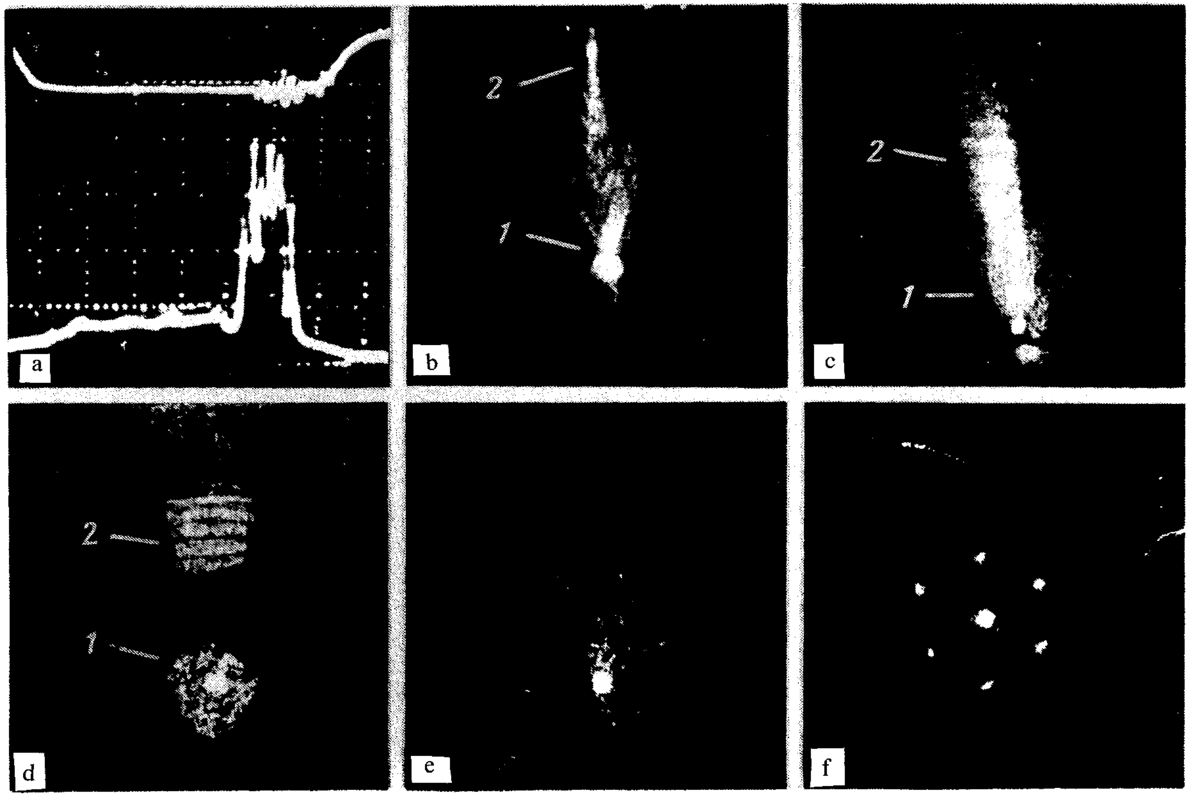

FIG. 1. a) Oscilloscope traces (sweep time 100 μs/div) and spatial spectra of the generated radiation: b) in a cell with $l = 0.5$ mm and $d/f \approx 0.1$, $\theta_i \approx 5$–6°; c) same parameters, with $\theta_i \approx 3$–4°; d) in a cell with $l = 2$ mm with $d/f \approx 0.05$ and $\theta_i \approx 5°$. e) For a pump beam transmitted through a phase plate for $d/f \approx 0.1$ and $\theta_i \approx 3°$; f) or through a "grid" amplitude transparency and an aberrator at the plane conjugate to the transparency. The arrows indicate the generated wave (1) and the reflected pump beam (2).

upper oscilloscope trace in Fig. 1a). The delay time decreased with a decrease in the angle of incidence of the pump beam on the cell and with an increase in the pump power and in the temperature of the cell, which was placed in a constant-temperature system. The temperature dependences of the time of development of generation and the pump threshold power, which are particularly pronounced near the phase transition between the nematic liquid crystal and the isotropic liquid, indicate that the effect is due to the thermal nonlinearity of the liquid crystal. In fact, the thermal nonlinearity coefficients of a nematic liquid crystal $(\partial n_{0,e}/\partial T)$ increase near the nematic/isotropic phase transition (at a temperature of the medium within 2–3 K of the critical temperature T_c) by more than an order of magnitude, reaching values of $\approx 10^{-2}$ K^{-1} (Ref. 4). It is estimated that already for an effective value of $(\partial n/\partial T) \approx 2\times 10^{-3} K^{-1}$ for a pump intensity $I_p \approx 5$ kW/cm^2 the growth rate of the thermal scattering is $G = \alpha l(\partial n/\partial T) I_p [8\pi k D \sin^2(\theta_p/2)]$ (where D is the thermal conductivity of the medium) at a scattering angle $\theta_s \approx 4°$ and $\alpha l \approx 0.5$ attains a value of $G_{th} \approx 17$–20, which is sufficient (as shown below; Fig. 2b) for the development of generation in the field of beams intersecting at an angle $\theta_i \approx \theta_s$. If we also take into account the heating of the medium during the time of the pump pulse (the steady-state temperature rise of the medium on the axis of a 0.5-W beam for an absorption coefficient $\alpha = 3$ cm^{-1} is ≈ 20 K), we can easily see that for these experimental parameters the conditions for generation in the nematic liquid crystal with $T_c \approx 320$ K are at-

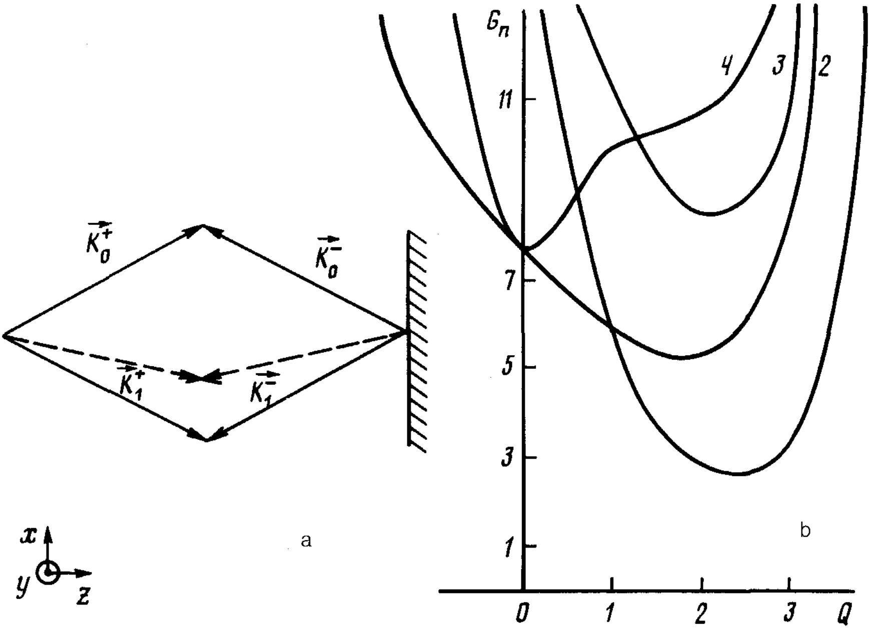

FIG. 2. (a) Geometry of the wave vectors of the interacting pump waves ($\mathbf{k}_0^{\pm}$) and the generated waves $\mathbf{k}_1^{\pm}$) in a nonlinear layer with specular boundaries. (b) Dependence of the generation threshold G_{th} on the detuning parameter for plane pump waves for $\alpha l \ll 1$ and (1) $R = 0.75$; (2) $R = 0.1$; (3) $\alpha l = 1$ and $R = 0.95$; (4) for beams with $z_d = l$, $R = 0.1$, $\alpha l \ll 1$.

tained when the wall of the cell is heated in the constant-temperature system to $T_{ct} \approx 300$–310 K.

The spatial structure of the generated radiation, which was studied visually and with the use of an image converter, and was recorded on I-1060 photographic film, exhibited substantial changes with variations in the angle of intersection and length z_f of the focal region of the pump beam in the nematic liquid crystal layer. At large angles $\theta_i \gg \theta_2 \equiv d/f$ (where d is the diameter of the pump beam at the lens and f is the focal length of the lens) the generated radiation was considerably different from the conjugated radiation: the spectrum of the generated radiation (recorded at the focal plane of a supplementary lens with a focal length $f \approx 200$ mm) is smeared out near the plane of incidence and elongated in the direction of specular reflection (Fig. 1b). As the angle of incidence was reduced (in air $\theta_i < \theta_2 n_0$), while the focal region was embedded in the nonlinear medium ($z_f < 2l$) a core was observed in the scattering spectrum (Fig. 1c,d) simultaneously with the development of generation in the backward direction (monitored with a photodetector), with dimensions corresponding to a divergence close to the diffraction limited divergence of the pump beam, θ_d. The maximum power of this radiation emitted in the angle $(1.5$–$2.0)\theta_d$ was $\approx 10\%$ of the power of the incident pump beam. In addition to the core, the integrated scattering spectrum (summed over the time of the pump pulse) also exhibited a nonconjugated component. The generation of the conjugated wave was observed in experiments with a phase plate, which increases the divergence of the pump beam to $(10$–$15)\theta_d$: for sufficiently strong focusing of the pump beam in the nematic liquid crystal layer with a mirror ($z_f < 2l$) the core in the spatial spectrum of the generated beam (after it had passed back through the phase plate) contained up to 20% of the reflected power (Fig. 1e). With this same focusing we observed phase conjugation of a beam carrying the image of an amplitude transparency distorted by an aberrator: after the beam passed back through the aberrator it reproduced the spectrum of the transparency (Fig. 1f).

The effects of parametric generation observed in the experiments can be clarified by considering the example of plane pump waves $E_0^{\pm} \exp(i\omega_0 t + i\mathbf{k}_0^{\pm}\mathbf{r})$ which intersect in a medium with a thermal nonlinearity. Noise (seed noise) of the light waves of the form $A_1^{\pm} \cos(qy)\exp(i\omega_0 t + i\mathbf{k}_1^{\pm}\mathbf{r} + \beta t)$, for which the wave vector $\mathbf{k}_1^{\pm}$ lies in the plane of intersection (Fig. 2a), can grow as a result of combined scattering of the pump beams by the thermal grating that the pump beams produce: $T_g \approx (E_0^{+*}A_1^{+} + E_0^{-*}A_1^{-} \exp(i\mathbf{k}\mathbf{r}))\cos(qy)\exp(i(\mathbf{k}_1^{\pm} - \mathbf{k}_0^{+})\mathbf{r})$, where $\kappa = \mathbf{k}_0^{+} - \mathbf{k}_1^{+} - \mathbf{k}_0^{-} + \mathbf{k}_1^{+}$. If the thermal grating grows faster than it is smoothed out by thermal conduction, then this growth becomes exponential in time and scattering waves are generated $[\mathrm{Re}(\beta) > 0]$. The minimum growth rate G_{th} of the stimulated scattering necessary for generation depends on the direction of propagation and the structure of the wave fronts of the seed waves. These factors are governed by the detuning parameter: $Q = q^2 l/(2k) + \kappa l$. The dependence of the threshold growth rate $G_{th}(Q)$ for the value of $\mathrm{Im}(\beta) \equiv \Omega$ for each value of Q of detuning of the scattering wave frequency, obtained in the approximation of an undepleted pump for constant $(\partial n/\partial T)$, is shown in Fig. 2b for the case where the counterpropagating waves are formed by reflection from the rear reflecting boundary of the nonlinear layer. The detuning Q, for which the lasing threshold is a minimum, varied in magnitude with variations in the coefficient of reflection R of the mirror and the absorption of the medium (cf. curves 1–4 in Fig. 2b) and changes sign when the coefficient $(\partial n/\partial T)$ changes sign, but for plane pump waves it remains nonzero.

In the other limiting case, for counterpropagating speckle pump beams, the complex amplitudes of the perturbation waves (by analogy with the theory of phase conjugation by backward stimulated scattering[5]) can be sought in the form $E_1^{\pm} \approx E_0^{\mp *} \exp(i\mathbf{qr} + \beta t)$. Within the approximations of small amplification of the noise at the wavelength of one speckle nonuniformity of the pump beams and for a given average curvature of their wave fronts, the dependence of the threshold growth rate on the detuning $Q = q^2 l/(2k)$, obtained by a decomposition of the beams into the Fourier components of their plane waves, is given by the expression (for $\alpha l \ll 1$)

$$\frac{G_1(1+3R) - 2Q(1 - H(R_1 + 1/R^2)) - B}{G_1(1+3R) - 2Q(1 - H(R_1 + R)) + B} = \exp(iB),$$

where $B = \sqrt{(1-R)^2 G_1^2 + 4Q^2(1 - HR_1)}$, $G_1 = G/(1 + ir_g\Omega)$, $R_1 = (1+R^2)/R$, $H = 1/z_d$, and z_d is the diffraction-limited divergence length of the pump beam. For $H \ll 1$ this relation gives the dependence $G_{th}(Q)$ for plane pump waves. For $H > 0.5$ and $R < 0.2$ the minimum threshold is reached for $Q = 0$ (curve 4 of Fig. 2b). Thus, the lowest lasing threshold is obtained for waves with fronts that are turned toward the counterpropagating pump waves, when the diffraction length of the pump beams is comparable to the dimensions of their region of intersection in the nonlinear medium, and their intensities differ by a factor of several units or the absorption is sufficiently strong $(\alpha l \approx 1)$.

Another factor that may have an influence on the spatial structure of the generated radiation is the increase of the coefficient of nonlinearity $(\partial n/\partial T)$ of the nematic liquid crystal near the nematic/isotropic phase transition. If the temperature approaches the critical temperature at the maxima of the heating effect due to the pump beams but there is no overheating, then the greatest amplification is obtained for a conjugated wave whose maxima are correlated with the maxima of the intensity of the pumping.

These factors can determine the spatial structure of the generated beams until the length of the intersection region of the pump beams is commensurate with z_d and l. At large angles of incidence the geometry of the intersection region provides a greater amplification for the scattering wave that propagates at an angle that is smaller than θ_i. This fact may explain the experimentally observed scattering spectrum for $\theta_i \gg \theta_2$ (Fig. 1b).

In summary, in the parametric generation of laser beams focused in a nematic liquid crystal with a dye and intersecting at a small angle, the beams can undergo phase conjugation. The important distinction between this generation involving forward Rayleigh stimulated scattering and the analogous processes based on nonlocal photorefractive nonlinearity is the possibility of decreasing the time of development of generation by increasing the pump power. This time τ is determined from the condition that the scattering wave grows from the noise level to an observable level. For low-angle scattering in a nematic liquid crystal this means that the growth rate of the generated wave, which is described by the expression $G/G_{th} - 1)\tau/\tau_p$ (where τ_r is the relaxation time of the thermal grating), should reach values between 6 and 10 (Ref. 6). The minimum delay time obtained experimentally near the nematic/isotropic phase transition ($\approx$50–100 μs) is much shorter than the analogous times in most photorefractive crystals.

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