Induced light scattering in the mesophase of a nematic liquid crystal (NLC)

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(Submitted 29 August 1979)
Pis'ma Zh. Eksp. Teor. Fiz. **30**, No. 8, 510–513 (20 October 1979)

The process of induced scattering (IS) of light in the mesophase of an NLC, i.e., the amplification process of a weak field, frequency-shifted relative to the monochromatic strong field, is discussed. Two IS mechanisms are considered: 1) nonstationary rotation of the direction of the magnetic field by the interfering fields and 2) nonstationary heating of weakly absorbing NLC by the interfering fields. A calculation yields an extremely high gain coefficient and a low IS threshold power.

PACS numbers: 61.30.Gd, 42.65.Gv

Recent years have witnessed considerable interest in nonlinear optical phenomena in liquid crystals (LC's), see, for example, the review in Ref. 1. We recently investigated strong self-stimulation effects (cubic optical nonlinearity) of monochromatic light fields in the oriented mesophase of nematic and cholesteric LC's (see also Ref. 2).

This paper discusses two possible mechanisms for the induced scattering (IS) of light in the oriented mesophase of an NLC.

1. **IS due to nonstationary rotation of the director**. A calculation of the perturbations $\delta\mathbf{n}(\mathbf{r},t)$ of the normal relative to the unperturbed magnetic field direction $\mathbf{n}$ [so that $(\delta\mathbf{n}\mathbf{n}) = 0$] due to the action of the anisotropic part of the interaction energy $\epsilon_0|(\mathbf{E}\mathbf{n})|^2/16\pi$ of an NLC with a light field gives for the variations of the dielectric

constant tensor $\delta\epsilon_{ik}(\mathbf{r},t)$ at the light frequency:

$$\delta\epsilon_{ik}(\mathbf{r},t) = 4\pi e^{i\mathbf{qr} - i\Omega t}\chi_{iklm}(\Omega,\mathbf{q})(\mathbf{E}_L)_l(\mathbf{E}_S^*)_m + 4\pi e^{-i\mathbf{qr} + i\Omega t}\chi^*_{iklm}(\Omega,\mathbf{q})(\mathbf{E}_L^*)_l(\mathbf{E}_S)_m,$$

$$\chi_{iklm}(\Omega,\mathbf{q}) = \frac{\epsilon_a^2}{64\pi^2}\frac{1}{q^2-(\mathbf{qn})^2}\{\Gamma_2[f_{il}n_kn_m + f_{mi}n_kn_l + f_{lk}n_in_m + f_{km}n_in_l - 4q^2n_in_kn_ln_m] - \Gamma_1[p_iq_ln_kn_m + p_mq_in_kn_l + p_lq_kn_in_m + p_kq_mn_in_l - 4(\mathbf{qn})^2n_in_kn_ln_m]\}, \quad (1)$$

$$\mathbf{q} = \mathbf{k}_L - \mathbf{k}_S, \quad \Omega = \omega_L - \omega_S,$$

$$\Gamma_1 = [-i\eta\Omega + K_{11}q^2 + (K_{33} - K_{11})(\mathbf{qn})^2 + \kappa_a H^2]^{-1},$$

$$\Gamma_2 = [-i\eta\Omega + K_{22}q^2 + (K_{33} - K_{22})(\mathbf{qn})^2 + \kappa_a H^2]^{-1},$$

$$\mathbf{p} = 2\mathbf{n}(\mathbf{qn}) - \mathbf{q}, \quad f_{ik} = [q^2 - (\mathbf{qn})^2]\delta_{ik} + p_iq_k \neq f_{ki}.$$

Here K_{11}, K_{22}, K_{33} are the Frank constants, $\kappa_a H^2/2$ is the anisotropic part of the interaction energy of the static magnetic field $\mathbf{H} = \mathbf{n}H$, as a result of which the normal orientation $\mathbf{n} = \mathbf{n}^0$ is maintained uniform within the entire volume. (For orientation by a static electric field $\mathbf{E}_{\text{stat}}$ the quantity $\kappa_a H^2$ is replaced by $\epsilon_a^{\text{stat}}E^2_{\text{stat}}/4\pi$). In the derivation of (1) it was assumed that two light waves are acting on the NLC:

$$\mathbf{E}(\mathbf{r},t) = 0.5\mathbf{E}_L\exp(i\mathbf{k}_L\mathbf{r} - i\omega_L t) + 0.5\mathbf{E}_S\exp(i\mathbf{k}_S\mathbf{r} - i\omega_S t) + \text{c.c.},$$

and only the interference terms in $\mathbf{E}_L$, $\mathbf{E}_S$ are left in $\delta\epsilon_{ik}(r,t)$. The quantity $\epsilon_a = \epsilon_\parallel - \epsilon_\perp$ is the anisotropy of the tensor $\hat{\epsilon}$ at the light frequency, η (g/cm·sec) is the relaxation constant of the NLC. The gain coefficient g_s (cm^{-1}, with respect to intensity) of the signal wave $\mathbf{E}_S = \mathbf{e}_S E_S$ in the presence of the pumping field $\mathbf{E}_L = \mathbf{e}_L E_L$ is given by the expression

$$g_S = -\frac{\omega_S^2}{c^2}\frac{4\pi}{k_S}|E_L|^2\mathrm{Im}[e_{Si}e_{Sl}e_{Lm}e_{Lk}\chi_{iklm}(\Omega,\mathbf{q})]. \quad (2)$$

The physical mechanism of the amplification in the presence of IS consists of the following. Interference of the fields $\mathbf{E}_L^*$ and $\mathbf{E}_S$ excites, because of the orientation forces, a traveling wave of local normal rotations, $\delta\mathbf{n} \propto \exp(-i\mathbf{q}r + i\Omega t)$ (as though it is recording a dynamic hologram). The scattering of the field E_L by this hologram gives the field $\mathbf{E}_S$ again. It is significant that in the presence of a phase delay $\delta\epsilon_{ik}$ of the grating with respect to the fields $E_L^* E_S$ (in the presence of Im $\hat{\chi}^*$) the scattered waves interfere with the already existing field $\mathbf{E}_S(r)$, weakening or enhancing it, depending on whether Im $\chi^* > 0$ or Im $\chi^* < 0$. Equation (2) with (1) taken into consideration can also be obtained by means of a conversion from the spontaneous light scattering cross section, a differential in terms of frequency,[3] using the coupling formulas from Refs. 2, 4. It also follows from the coupling formulas that the amplification is positive only when the signal lies in the Stokes region relative to the pumping $\omega_L - \omega_S = \Omega > 0$. The quantity g_S from (2), (1) depends in a rather complicated manner on the polarizations e_L and e_S of the interacting waves and the orientation of the vector $\mathbf{q} = \mathbf{k}_L - \mathbf{k}_S$ with respect to the normal. Let us consider a specific case when the wave vectors $\mathbf{k}_L$ and $\mathbf{k}_S$ lie in a plane perpendicular to the normal $\mathbf{n}^0$. Let us assume, in addition, that $\mathbf{e}_L$ corresponds to the extraordinary wave, $\mathbf{e}_L \| \mathbf{n}^0$. Then the amplification will be different from zero only for the ordinary-type signal wave, $\mathbf{e}_S \perp \mathbf{n}^0$. In this case only the structure of $\Gamma_2(\mathbf{\Omega},\mathbf{q})$ from (1) plays a role, and the gain coefficient is equal to

$$g_S = g_{max}(q)\,\frac{2\Omega\gamma}{\Omega^2 + \gamma^2}, \qquad \gamma = \frac{K_{22}q^2 + \kappa_a H^2}{\eta},$$

$$g_{max} = \frac{\epsilon_a^2}{32\pi^2}\,\frac{\omega_S^2}{c^2}\,\frac{1}{k_S\eta\gamma}\,|E_L|^2. \tag{3}$$

For backward IS ($\mathbf{k}_S = -\mathbf{k}_L$) we have $q = (\omega/c)(n_0 + n_e) = 3.3\times10^5$ cm^{-1} (for $\lambda = 623$ nm), and the optimum frequency shift is $\Omega_{opt}/2\pi = \gamma/2\pi \approx 1\times10^5$ Hz. Here we used the values $K_{22} = 2.9\times10^{-7}$ dynes, $\eta = 5\times10^{-2}$ g·cm^{-1}·sec^{-1}. For the nematic PAA one can assume $\epsilon_\alpha \approx 1$ (at $T = 125°$ C). Then $g_{max} = Gcn_e|E_L|^2/8\pi$, and, if the power density of the laser beam $P_L = cn_e|E_L|^2/8\pi$ is expressed in megawatts per cm^2, then $G \approx 80$ cm/mW. For a medium with length $z = 10^{-1}$ cm a gain of $\exp(GP_L z) = \exp(10)$ is achieved for a pumping power density of $P_L = 1.25$ MW/cm^2. A time $\tau = GP_L z/2\gamma$ is required to establish the IS process (see, for example, Ref. 5). In this example $\tau = 0.8\times10^{-5}$ sec.

For forward IS $q = (\omega/c)(n_e - n_0) \approx 2.9\times10^4$ cm^{-1}, and with all other conditions the same we have $G \approx 10^4$ cm/MW, $\gamma/2\pi = 7.6\times10^2$ Hz. A gain of $\exp(GP_L z) = \exp(10)$ is achieved in a length $z = 0.1$ cm for $P_L \approx 10^4$ W/cm^2, and $\tau_{estab} \approx 10^{-3}$ sec. Thus, IS of this type should be quite easily detectable in an NLC. In the case of sample orientation by the walls (instead of a magnetic field) Eq. (3) will be valid only at a distance $l \gtrsim q^{-1}$ from the walls. Since $q \gtrsim 10^4$ cm^{-1} always, this restriction has little significance.

2. Temperature induced scattering in an NLC, caused by absorption (TIS-A).

Temperature induced scattering of light, caused by absorption (TIS-A), is well known in nonlinear optics (see Ref. 6 and the reviews in Refs. 4, 7). The recording mechanism of a traveling grating $\delta\hat{\epsilon}(\mathbf{r},t) \sim \exp(i\Omega t - i\mathbf{qr})$ in TIS-A is based on an evolution of heat through the absorption of the energy of the interfering light fields. In this case the gain coefficient can be positive in either the Stokes or in the anti-Stokes region, depending on whether $\partial\epsilon/\partial T > 0$ or $\partial\epsilon/\partial T < 0$. The specific property of an NLC, leading to an anomalously low TIS-A threshold and to an anomalously high gain coefficient, is a very large value of $\partial\epsilon/\partial T$. Thus, in the mesophase of MBBA at $T \approx 33°$ C, i.e., at 10° C from the phase transition point ($T_K \approx 43°$ C), the quantity $\partial\epsilon_{\parallel}/\partial T \approx -0.9 \times 10^{-2}$ deg^{-1} and $\partial\epsilon_{\perp}/\partial T \approx -0.2 \times 10^{-3}$ deg^{-1} (see Ref. 8). Let us recall that for most liquids $\partial\epsilon/\partial T$ amounts to 10^{-5} deg^{-1}. Our consideration of TIS-A in an NLC also takes account of the light absorption anisotropy in the NLC (dichroism) and the heat conductivity anisotropy. The expression for the gain coefficient of the wave $\mathbf{E}_S = \mathbf{e}_S E_S$ in the presence of a strong wave $\mathbf{E}_L = \mathbf{e}_L E_L$ has the form

$$g_S = A \frac{\omega_S}{8\pi} |E_L|^2 \beta_{lm} e_{Sl} e_{Lm} - \beta_{lm} e_{Sl} e_{Sm} , \tag{4a}$$

$$A = \frac{\Omega}{\rho C_p (\Omega^2 + \gamma^2)} \left[\frac{\partial \epsilon_{\perp}}{\partial T} (\mathbf{e}_S \mathbf{e}_L) + \frac{\partial \epsilon_a}{\partial T} (\mathbf{e}_S n)(\mathbf{e}_L n) \right]. \tag{4b}$$

Here $\gamma = \kappa_{ik} q_i q_k$; in addition, $\kappa_{ik} = \kappa_{\perp}\delta_{ik} + (\kappa_{\parallel} - \kappa_{\perp}) n_i n_k$ is the thermal diffusivity tensor, $\beta_{ik} = \beta_{\perp}\delta_{ik} + (\beta_{\parallel} - \beta_{\perp}) n_i n_k$ is the tensor $\hat{\beta} \approx (\omega/cn)\,\text{Im}\,\hat{\epsilon}$, determining the light absorption. We assume the anisotropy of the real part of $\hat{\epsilon}$ is not too large and therefore it can be assumed $n \approx (n_0 + n_e)/2$; $\beta_{\parallel}$ and $\beta_{\perp}$ are equal to the absorption coefficients for the *o*- and *e*-waves, respectively, for propagation perpendicular to the axis. The second term in (4a) corresponds to the absorption coefficient β_S of the E_S wave, and therefore amplification begins only when the pumping exceeds the threshold power density P_{thr}. In the general case the quantity P_{thr} depends on the ratio $\beta_{\parallel}/\beta_{\perp}$, but not on the absolute magnitude of the absorption, and the gain coefficient is equal to $g_S = \beta_S [P_L/P_{\text{thr}} - 1]$. Let us give numerical estimates for TIS-A in the substance MBBA for the above-mentioned value $T \approx 33°$ C. For backward TIS-A ($\mathbf{K}_S \approx -\mathbf{K}_L$), for $\lambda = 632.8$ nm and for the case when both waves are *e*-type, with a thermal diffusivity coefficient $\kappa \approx 0.6 \times 10^{-4}$ cm^2/sec we obtain $P_{\text{thr}} \approx 20$ kW/cm^2, and the optimum frequency shift is $\Omega/2\pi = \gamma/2\pi \approx 9 \times 10^5$ Hz. For an absorption coefficient (for which it is not advisable to take too small a value here) of the order $\beta_S \approx 5$ cm^{-1} and a medium with a thickness $z = 10^{-1}$ cm a gain of exp(10) is achieved for $P_L \approx 20 P_{\text{thr}}$ ≈ 400 kW/cm^2; the process establishment time $\tau = \beta_S P_L z / 2\gamma P_{\text{thr}}$ amounts to $\tau \approx 0.8 \times 10^{-6}$ sec.

Thus, two physical mechanisms have been indicated in this paper for induced scattering of light in the mesophase of an NLC (orientation and thermal), leading to an extremely large gain coefficient value. The detection of the predicted types of IS is, in our opinion, of considerable interest, including dealing with the problem of the wavefront rotation of the light for backward IS, see Ref. 9. In conclusion the authors wish to thank S.M. Arakelyan, E.I. Kats and Yu.S. Chilingaryan for valuable discussions.

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