Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl16

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Version of record first published: 28 Mar 2007.

To cite this article: B. Ya. Zel'Dovich , A. V. Sukhov & N. V. Tabiryan (1987): Theory and Experiment on Stimulated Scatterig of Light in Nematics, Molecular Crystals and Liquid Crystals, 143:1, 7-15

To link to this article: http://dx.doi.org/10.1080/15421408708084605

Mol. Cryst. Liq. Cryst., 1987, Vol. 143, pp. 7-15
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Printed in the United States of America

THEORY AND EXPERIMENT ON STIMULATED SCATTERING OF LIGHT IN NEMATICS

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Abstract Theory for transient gain by stimulated scattering (SS) is rederived in simple terms. Orientational SS in forward direction has been achieved experimentally in a planar cell of nematic 5CB in a free-running pulse from a ruby laser. Measurements of the time evolution of SS signal and of its angular spectrum confirm theoretical predictions.

INTRODUCTION

Stimulated scattering(SS) of light by thin nematic liquid crystal (NLC) cell in the self-diffraction mode was predicted in [1]. SS in the volume of NLC was predicted [2,3] to appear as a result of excitation of a travelling wave of the director orientation, see also our review [4]. The emphasis in[2,3] was made mostly on the planar cell geometry. However, our first experimental observation of SS was done[5] for the near-to-normal incidence of the cwargon laser o-wave pump onto the homeotropic cell with NLC 5CB, see also[4]. The results of experimental observation[6] of forward SS of ruby laser pump in a planar 5CB cell are presented in this report along with

simple derivation of transient SS formulae. Experiment on SS in the self-diffraction mode was presented on Naples 1986 "Optics of Liquid Crystals" Meeting by Y.C.Khoo and coworkers.

THEORY

Let an intense pump wave $\vec{E}_{\perp}$ of the extraordinary polarization is propagating through a planar sample of a nematic liquid crystal along the direction normal to the unperturbed director $\vec{n}$ of this crystal. An ordinary-polarization wave $\vec{E}_S$, with a slight frequency shift in the Stokes direction, is propagating collinear with $\vec{E}_{\perp}$. The interaction of these two waves with the dipole moment, which they induce in the medium, gives rise to a traveling wave of a reorientation of the director, $\delta\vec{n}$, by virtue of the anisotropy of the dielectric constant of the nematic liquid crystal. The amplitude of this reorientation traveling wave is proportional to $E_{\perp}^{*}E_S$; its wave vector q is equal to the difference $|\vec{k}_S - \vec{k}_{\perp}| = 2\pi\Delta n/\lambda$, where $\Delta n = n_{\parallel} - n_{\perp}$ is the anisotropy of the refractive index of the liquid crystal, and λ is the wavelength of the light in a vacuum (we are ignoring the small difference between the wavelengths of $\vec{E}_{\perp}$ and $\vec{E}_S$ which results from their frequency difference). This reorientation wave in turn leads to the writing of a traveling grating of the dielectric tensor in the medium. The scattering of the pump wave $\vec{E}_{\perp}$ by this grating leads in the steady-state case, i.e., with

a steady-state amplitude $\delta\vec{n}(\vec{r})$, to an exponential spatial amplification of the wave $\vec{E}_s$, i.e., to a stimulated scattering of $\vec{E}_L$ into $\vec{E}_s$: $|E_s|^2 \sim \exp(GLP_L)$, where G is the steady-state gain, L is the interaction length (the thickness of the cell), and $P_L = cn_\parallel |E_L|^2/8\pi$ is the pump power density. The steady-state gain, the frequency shift of the stimulated scattering, and its relaxation time τ are[2,3]

$$G = \frac{10^{13}\lambda (n_\parallel + n_\perp)^2}{8\pi n_\parallel n_\perp K_{22} c}\left(\frac{cm}{MW}\right); \quad \Omega = \Gamma = \frac{K_{22}q^2}{\eta}; \quad \tau \simeq \Gamma^{-1} \quad (1)$$

Here K_{22} is the Frank constant, c is the velocity of light, and c is the velocity of light, and η is the orientational viscosity of the nematic liquid crystal.

When the stimulated scattering is excited by a laser pulse with duration $\tau_p \ll \Gamma^{-1}$, transient theory of SS is necessary. In this case the equation for the Stokes amplitude $E_s(z,t) = E_L(t) \times S(z,t)$ is[7,8]

$$\frac{\partial^2 S}{\partial z \partial T} = i G \Gamma S(z,T); \quad T = \int_0^t P_L(t')\,dt' \quad (2)$$

If we seek for the solution of (2) in the form $S(z,t) = \exp f(z,t)$, then it follows from (2)

$$\frac{\partial f}{\partial z}\cdot\frac{\partial f}{\partial T} + \left[\frac{\partial^2 f}{\partial z \partial T}\right] = i G \Gamma \quad (3)$$

For the exponentially large gain $|f| \gg 1$, the square brackets term may be omitted in (3), and then $f \approx \sqrt{4iG\Gamma z T}$,

$$|E_s|^2 = \alpha |E_L|^2 \exp\left[2\sqrt{2}\left(G\Gamma z \int_0^t P_L(t')\,dt'\right)^{1/2}\right] \quad (4)$$

The factor $\alpha|E_L|^2$ arises in front of the exponential function because the stimulated scattering develops from nucleating noise which is the Fourier component of the spontaneous scattering of the pump by thermal fluctuations of the director with a suitable spatial and temporal period. This nucleating noise subsequently undergoes the exponential amplification described above.

EXPERIMENT

Here we report the experimental observation[6] of forward orientational SS in a planar sample of NLC 5CB, 70μm thick. The SS was excited by a free-running pulse from a ruby laser $\sim$800μs long. An estimation of the relaxation time of SS from (1) yields $\tau \approx 5$ ms, so that SS was definitely a transient process.

The experimental arrangement used to observe the stimulated scattering is shown in Fig.1. The light from a ruby laser (RL), whose energy is adjusted over the range $\sim$30-300 mJ by filters LF, passes through an Iceland spar wedge W_1, which singles out the polarization component $\vec{e}_L$ of this light, which corresponds to the e polarization for the liquid crystal, and through an iris diaphragm D_1. The light is then focused by lens L_1 (f=5 cm) into a cell C holding the liquid crystal. Lens L_2, along with L_1 and C, forms a telescope system. Most of the transmitted light is directed to a Glan prism G, which singles out the polarization component $\vec{e}_S$ corresponding

to the stimulated-scattering signal. This component is detected by photocell F_2. Part of the light transmitted through C is reflected from the glass wedge and passes through Iceland spar wedge W_2, which separates the light into the components $\vec{E}_L$ and $\vec{E}_S$; this light is recorded on film with the help of a two-mirror stepped attenuator SA. This approach makes it possible to determine the angular divergences of $\vec{E}_L$ and $\vec{E}_S$ as the ratio of the size of the spot on the film to the focal length of L_2. To monitor the energy and shape of the laser pulse, we use a standard (IMO-2) calorimeter Cal and photocell F_1. Both the $|\vec{E}_L(t)|^2$ signal and the instantaneous value $\int_0^t P_L\, dt'$ (with the help of an integrating circuit) are taken from this photocell.

It should be noted that in these experiments the laser is operated in two regimes: a regime of a single mode in terms of the transverse index and a multimode regime. All the energy measurements are carried out in the multimode regime, since the pulse in the single-mode regime has a microsecond peak structure, which makes it extremely difficult to carry out accurate quantitative measurements. In the multimode regime, on the other hand, by choosing an appropriate resonator geometry we obtain a pulse with a nearly smooth temporal envelope. Figure 2 shows some typical oscilloscope traces of the pump pulse and of the stimulated-scattering pulse (a and b, respectively) in this regime. The results of the measurements of the time evolu-

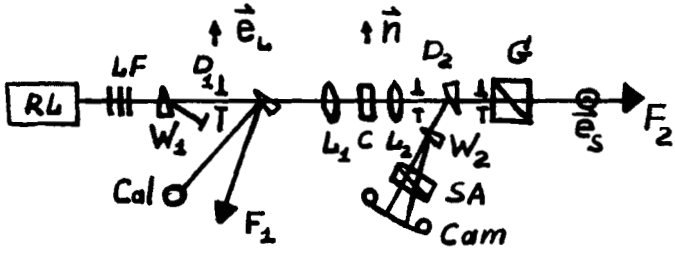

Figure 1

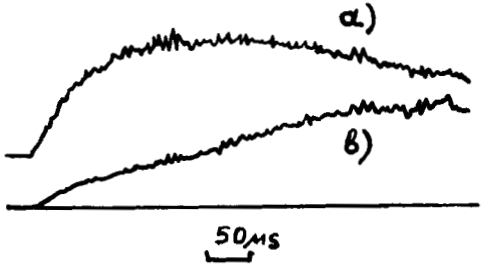

Figure 2

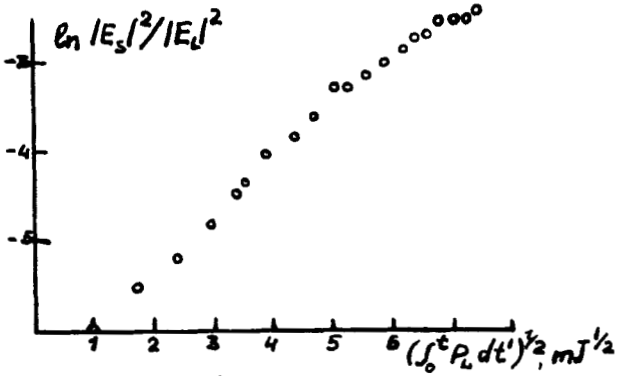

Figure 3

tion of the stimulatedscattering signal are shown in Fig.3 as a plot of $\ln|\vec{E}_S|^2/|\vec{E}_L|^2$ versus the instantaneous value of $(\int_0^t P_L dt')^{1/2}$. The obvious linearity of this dependence shows that the functional dependence of the results agrees well with Eq.(4). For a quantitative comparison of the proportionality factor of this dependence with the theoretical factor, we measured the transverse dimension of the beam in the crystal: a=FWHM$\approx$90μm. We found that the experimental proportionality factor is 1.2 times smaller than the theoretical factor calculated from $2\sqrt{2}(G\tau^{-1}(L/a^2))^{1/2}$. This result is again evidence of a good agreement of the theory with experiment within the experimental error. The experimental level of the spontaneous noise, α, is $\alpha\approx 5\times10^{-3}$. A theoretical estimate of this lefel from the known equations for the cross section for spontaneous scattering in nematic liquid crystals with allowance for the angular aperture of the measurement system, $\theta_p\simeq 1.5\times10^{-1}$ rad, is (in the single-constant approximation)

$$\alpha=\frac{\pi\Delta n^2(n_{11}+n_\perp)^2k_B T' L}{8\lambda^2 K_{22}}\int_0^{(\theta_p/\Delta n)^2}\frac{d\xi}{1+\xi}\approx 4.2\times10^{-3} \quad (5)$$

where k_B is the Boltzmann constant, and T^0 is the absolute temperature ($\sim$300 K). It can also be seen from (5) that the theoretical results agree well with the experimental results.

We turn now to the measurements of the angular spectrum of the stimulatedscattering signal and the pump signal. It should be noted that

the longitudinal and transverse dimensions of the interaction region are comparable in our experiments, so that the geometry allows not only collinear stimulated scattering but also stimulated scattering through quite large angles (on the order of 0.5 rad). In our previous study[5] of stimulated scattering in a homeotropic cell, the noncollinear stimulated scattering was discriminated against by a nonlinearity mechanism involving the sharp increase in the grating wave vector $\vec{q}$ upon a deviation from exactly collinear scattering. In this report $|\vec{q}|$ is on the order of 2×10^4 cm^{-1}, i.e., extremely large even for collinear scattering, so that the nonlinearity mechanism discriminates against the stimulated scattering only at angles $\gtrsim 0.2$rad. We therefore have every reason to believe that the angular spectrum of the stimulated scattering is much larger than that of the pump. To experimentally test this point, we used single-mode light with a divergence (after L_1) FWHM=5×10^{-2}rad. The experimental width of the angular spectrum of the stimulated scattering signal in this case was found to exceed the angular aperture of the measurement system ($\simeq 0.2$rad), and the angular spectrum was found to have an obvious speckle, not not reproducible in different cases. This result was to be expected for transient stimulated scattering (the pulse duration is much shorter than the scale time over which spontaneous fluctuations lose their time coherence).

In summary, we have experimentally detected a transient stimulated scattering with an extremely short interaction length, L=70 μm, and a quite moderate pump energy density, $\int P_L dt$=100 - -300 J/cm^2. The steady-state gain estimated from the experimental data is also extremely large, G=5x10^{3}cm/MW, at orientational viscosity $\eta \simeq 1$ P and $n_e^2 - n_o^2$=0.65.

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