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Light Scattering by Negative Nematics in the Near Infrared†

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The light scattering by negative dielectric anisotropy nematics submitted to high *ac* electric fields *E* has been studied in the near infrared range (up to a wavelength $\lambda = 2.5 \mu\text{m}$). The stabilizing field effect reduces the orientational fluctuations and therefore the scattered light intensity, more especially for the high wavelengths. For a given value of λ the reciprocal of the relative scattered intensity S^{-1} increases linearly with E^2 , the slope of the straight line obtained depending on the initial orientation (homogeneous or homeotropic) of the liquid crystal. At temperatures sufficiently far from that of nematic-isotropic transition the scattered intensity *S* decreases when the temperature increases. The relaxation time τ after a sharp cut-off of the electric field *E* has also been measured; as the scattered intensity, its reciprocal τ^{-1} increases following a linear law with E^2 . Applications of these results as a way of obtaining modulated infrared light are considered.

INTRODUCTION

Almost all of the experiments on the dynamic scattering mode (DSM) with nematic liquid crystals of negative dielectric anisotropy have been performed in the visible spectral domain. In this wavelength range, when an increasing voltage (*dc* or low frequency *ac*) exceeding

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a threshold of about 10 volts is applied to a thin nematic layer held between two transparent electrodes, the incident light is scattered due to electrohydrodynamic instabilities and the cell transmission decreases rapidly towards a low value (a few percent); then the transmitted intensity remains constant, even if the applied voltage reaches high values (80 volts for example).

Little attention has been given to the transmission of such cells in the infrared. Fray et al.¹ have determined for various applied voltages the light intensity scattered in the infrared (about $\lambda = 10 \mu\text{m}$) by a nematic of negative dielectric anisotropy; they have found three different regions of wavelengths attributed to the size of the scattering domains as compared with the incident wavelength. Recently we have shown² that the transmission in the near infrared by negative nematics increases as the incident wavelength and the applied voltage are higher as illustrated in Figure 1. Then the transmission can reach values as large as 80 to 90%; consequently the light scattering is attenuated or can disappear almost entirely.

We report here complementary results on this effect, in particular the electric field and temperature dependence of the scattering, the influence of the initial orientation and the variation of the relaxation time with the electric field.

EXPERIMENTAL

The transmission spectra for wavelengths λ from 0.4 to $2.6 \mu\text{m}$ have been determined for several negative ($\epsilon_a \approx -0.50$) nematics submitted to high *ac* (500 Hz) electric fields (up to 10^5 V.cm^{-1}), using a Beckmann spectrophotometer or a monochromator attached to a pyroelectric detector; the incident and emergent light beams are carefully collimated and are perpendicular to the glass plates. The cell transmission $\frac{I_v}{I_o}$ (I_v and I_o being the transmitted light intensities with and without applied voltage) and the relative scattered intensity $S = \frac{I_s}{I_o} = \frac{I_o - I_v}{I_o}$ are deduced from the experimental spectra for each wavelength.

The cells are conventional: thicknesses between 6 and $23 \mu\text{m}$, initial orientation homogeneous (by rubbing the electrodes) or homeotropic (by depositing a thin film of surfactant HTAB - hexadecyltrimethylammonium bromide -); their temperature is kept constant for each

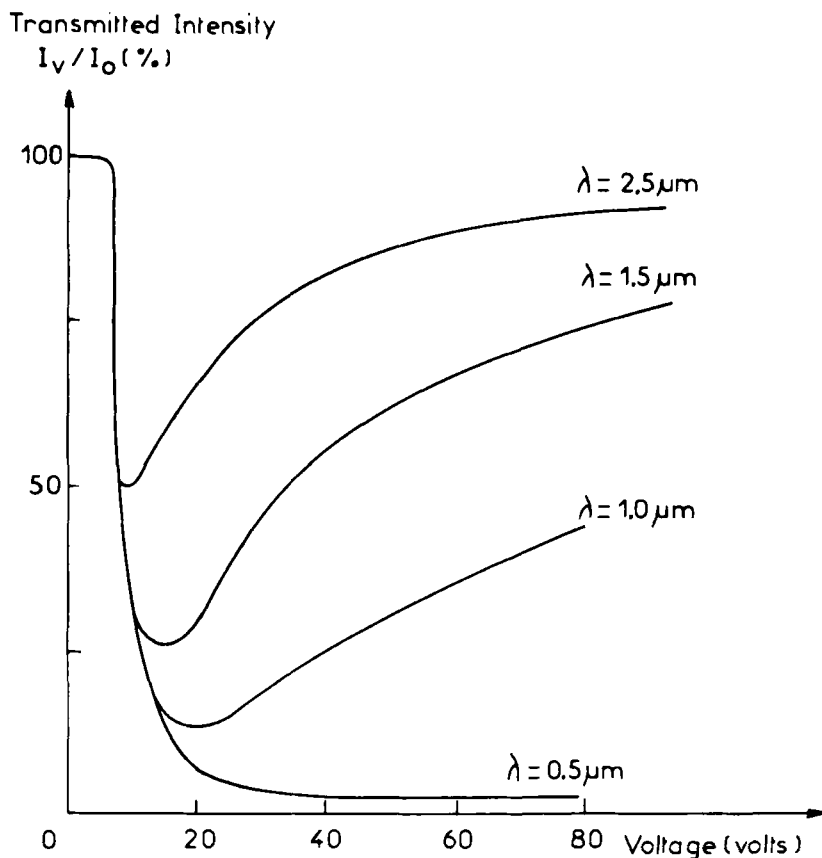


FIGURE 1 Relative transmitted intensity vs voltage for various wavelengths, in DSM ($\epsilon_a < 0$).

measurement but can be varied between 7 and 70°C. The following nematics were used: MBBA, and mixtures (from E. Merck) for DSM: NP 997, ZLI 1623 and ZLI 1735; they have dielectric anisotropies between -0.52 and -0.60 and, except for MBBA, relatively high nematic—isotropic transition temperatures (about 80°C). Their behavior concerning the DSM is approximately the same provided that the electrical conductivity is sufficient (in the case of pure MBBA it may be necessary to add small quantities of tetrabutylammonium chloride).

The relaxation time τ after a sharp cut-off of the applied voltage V (pulses of amplitude V) has also been measured in the infrared

range ($\lambda = 1.7 \mu\text{m}$) by using a monochromator attached to a germanium photodiode.

RESULTS

The experimental results obtained confirm the previous observations: the transmitted intensity increases with λ for wavelengths between $\lambda = 0.5$ and $1.0 \mu\text{m}$ according to the applied voltages, the higher the voltage the larger the increase of transmission. The light scattering in nematic liquid crystals is interpreted in terms of small amplitude orientational fluctuations, the density fluctuations giving a negligible contribution; the observed decrease of the scattering can be explained by the variation of these fluctuations under the effect of the electric field.

De Gennes³ has made an analysis of the scattering processes from which it comes out that the scattered intensity decreases with increasing wavelength λ . Under our conditions this analysis gives a linear variation with $\frac{1}{\lambda^2}$; this author has also shown that the application of a stabilizing field reduces the amplitude of the fluctuations: the scattered intensity S decreases for increasing fields and is given⁴ in the case of a stabilizing electric field E by the relation

$$\frac{1}{S} = a [1 + bE^2]$$

a being a constant and b a function of ϵ_a and of $\frac{\epsilon_{\perp}}{\epsilon_{\parallel}}$

A similar relation (S being replaced by τ) gives the damping time τ of the fluctuations.

Our results are interpreted in terms of the variation of the electric field E . In Figure 2 the linear increase of the relative scattered intensity S vs $\frac{1}{\lambda^2}$ is shown for a cell of MBBA and two values of the electric field E , in agreement with the data mentioned above. Similar results have been found with other nematics.

In Figure 3 the reciprocal of S is plotted vs E^2 for a given wavelength ($\lambda = 2.0 \mu\text{m}$). It should be noted that a linear graph would be obtained by plotting $\frac{1}{S}$ vs the cell current intensity; in fact we have

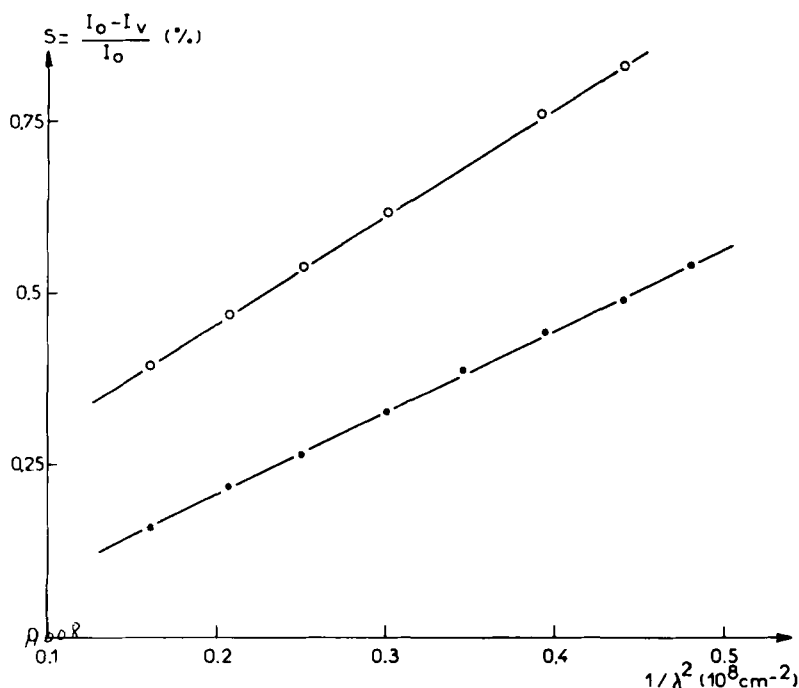


FIGURE 2 Relative scattered intensity S vs $\frac{1}{\lambda^2}$ for a 6 μm , MBBA, homogeneous cell at room temperature and for two values of electric field E : ○ $E = 5.10^4 \text{ V.cm}^{-1}$ ● $E = 10^3 \text{ V.cm}^{-1}$.

verified for the various products that the electric current is proportional to E^2 , depending on the electrical conductivity of the nematics, according to earlier observations.^{5,6} For the small thicknesses the slopes of the straight lines obtained in Figure 3 depend on the initial orientation: for a given value of E , $\frac{1}{S}$ is higher in homeotropic than in homogeneous alignment, corresponding to an increased stabilizing effect on fluctuations by the field, the initial ordering direction being coincident with that of E ; this difference disappears for thicknesses of about 10 to 13 μm . The influence of the anchoring forces is also shown by the variation of S with the thickness: the scattered intensity increases with the thickness.

The temperature dependence of the scattering has been also considered. In Figure 4, $\frac{1}{S}$ is plotted vs E^2 for $\lambda = 2.0 \mu\text{m}$ and for a cell of NP 997 ($d = 6 \mu\text{m}$, homogeneous orientation) at various tem-

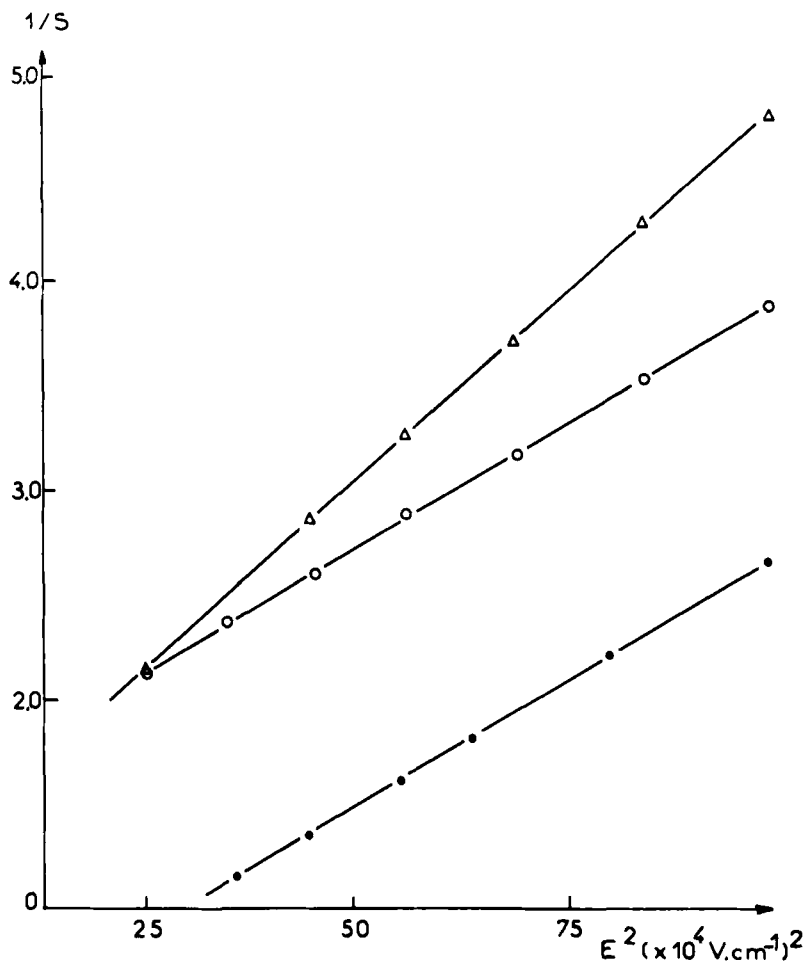


FIGURE 3 Reciprocal of the relative scattered intensity $\frac{1}{S}$ vs E^2 , for $\lambda = 2 \mu\text{m}$ in various cells of NP 997: Δ 6 μm homeotropic, \circ 6 μm homogeneous \bullet 10 μm homogeneous.

peratures T , sufficiently far from that of nematic-isotropic transition: the scattered intensity S decreases when the temperature increases for a given electric field. The relation giving the scattered intensity S indicates that it is essentially proportional to $\frac{\epsilon_a^2}{k_{11}}$ (ϵ_a dielectric anisotropy, k_{11} elastic constant); both parameters ϵ_a and k_{11} decrease with increasing temperature so that the temperature effects would be small. Instead of that a decrease of S and at the same time an im-

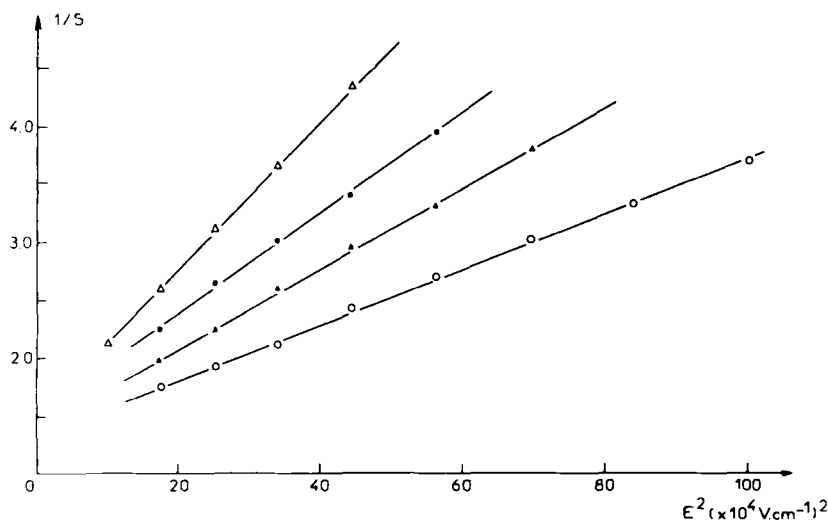


FIGURE 4 Reciprocal of the relative scattered intensity $\frac{1}{S}$ vs E^2 , for $\lambda = 2 \mu\text{m}$ in a homogeneous cell of NP 997 at various temperatures: ○ $T = 20^\circ\text{C}$, ▲ $T = 30^\circ\text{C}$, ● $T = 45^\circ\text{C}$, △ $T = 60^\circ\text{C}$.

portant increase of the current intensity i are observed experimentally. The linear graphs obtained by plotting $\log \left(\frac{1}{S} \right)$ and $\log i$ vs $\frac{1}{T}$ lead to think that the variations of S and i with the temperature are related to a decrease of the viscosity which makes the stabilizing field more effective.

As indicated above the relaxation times τ after a sharp cut-off of the electric field have been measured for various cells and for $\lambda = 1.7 \mu\text{m}$ (sensitivity limit of the germanium photodiode that was used); as expected the reciprocal of the time τ increases following a linear law, with E^2 as shown in example of Figure 5 for a $6 \mu\text{m}$ homogeneous cell of NP 997.

CONCLUSION

The results reported in this paper show that the variation of the scattered intensity of light with the electric field E which had been found for relatively low fields remains valid for higher values of E and also in the range of the near infrared. The increase of the transmission in the infrared and the relaxation times of a few tens of

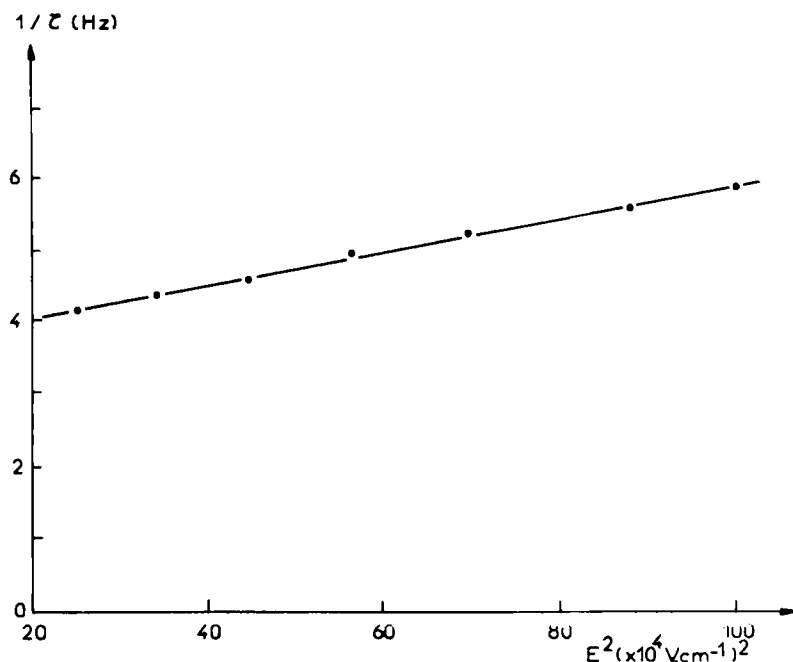


FIGURE 5 Reciprocal of the relaxation time τ vs E^2 for a 6 μm homogeneous NP 997 cell and for $\lambda = 1.7 \mu\text{m}$.

milliseconds found for some negative nematics lead to a way of obtaining modulated infrared light. The application to a slightly polarized cell (≈ 20 volts) and therefore opaque to the infrared radiation, of an electric pulse of sufficient amplitude (≈ 30 volts) will make the cell transparent to the infrared.

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