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Visco-Elastic Properties of Some Nematic Liquid Crystals

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We report on some temperature dependent visco-elastic properties of di-butyl-azoxy-benzene (Dibab) and 4 methoxy-4'-azoxy-benzene (N_4) which have their nematic temperature range from 19.5 to 31.5°C and 16 to 76°C respectively. The results are obtained by studying the noise intensity spectrum of Rayleigh scattered laser light. Experiments are done using both homodyne and heterodyne techniques.

From the configuration where the director is perpendicular to the plane of observation we obtained both the splay-bend and the twist-bend slow mode.

INTRODUCTION

Light-scattering is a good way of obtaining information about viscoelastic properties of nematic liquid crystals. The local thermal fluctuations of the director (**n**) give rise to an intense scattering of light. The bend, twist and splay visco-elastic ratios follow from the linewidth of the Lorentzian-shaped spectra of the light intensity fluctuations. The general formulas for these spectra have been derived by the Orsay group.[3] In order to collect our data we have used our orientated sample and the polarization of the incoming and scattered light in two geometric configurations. In the first configuration (1) **n** is perpendicular to the plane of observation (**k**,**k**') which is defined by the wave vector of the incoming polarized laser beam **k** and the wave vector of the polarized scattered light **k**'. In the second configuration (2), **n** lies in the plane of observation.

The experiments are similar to those which were done previously by the Orsay group[6] and by Fellner *et al.*[1] However, we have made a very close study of configuration (1). We have measured very carefully the homodyne noise intensity spectrum at different scattering angles θ, where θ is the angle between **k** and **k**' inside the scattering medium. It turns out that we can

deduce both the twist and splay visco-elastic ratios as a function of temperature.

We verified whether the width of the homodyne light intensity spectrum was twice the optical linewidth by measuring a heterodyne spectrum under the same conditions and comparing the two spectra. We measured the heterodyne spectrum by mixing the scattered laser light with a local oscillator, i.e. the original split laser beam.[2] In Figure 1 we plotted logarithmically relative excess noise, i.e. the noise in excess of shot noise relative to the shot noise level, versus frequency for the homodyne as well as for the heterodyne case. (The figure shows the result of our measurements which were done with configuration (2)). The results obtained are consistent with the assumption of the Gaussian character of the scattered light field, i.e. the half-width of the heterodyne spectrum is half that of the homodyne spectrum.

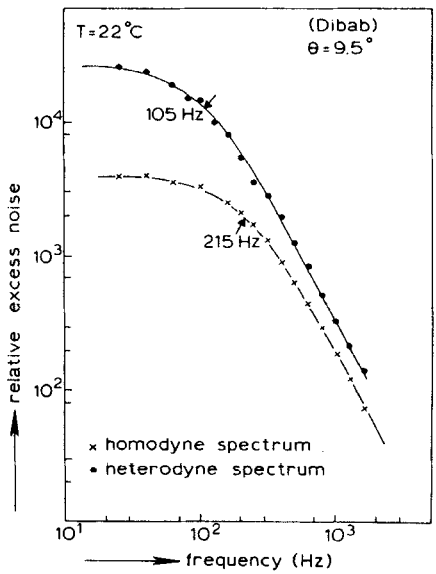

FIGURE 1 Relative excess noise intensity versus frequency. The spectra illustrate the Gaussian character of the optical field.

THEORY

The general expression for the optical spectrum was derived by Orsay[3] (Eqs. IV.23′, IV.25[a,b]). If we assume that the fluctuating process of the optical field is Gaussian, then the spectral intensity of the light intensity fluctuations, $S_{\Delta I}(\omega)$, is equal to the auto-convolution integral over the optical spectrum

(see Alkemade[5]). Thence

$$S_{\Delta I}(\omega) = M^2\left\{\sum_{\alpha=1,2}\frac{G_\alpha^4(\theta)(2u_{s\alpha})}{K_\alpha^2(q)[\omega^2+(2u_{s\alpha})^2]} + \frac{G_1^2G_2^2(u_{s1}+u_{s2})}{K_1K_2(\omega^2+(u_{s1}+u_{s2})^2)}\right\} \quad (1)$$

where G_α is a geometrical factor equal[3] to $(i_\alpha f_0 + i_0 f_\alpha)$ and M a constant. For the significance of the other symbols the reader is referred to Ref. 3.

The spectrum $S_{\Delta I}(\omega)$ consists of three parts associated with a splay-bend mode, a twist-bend mode and a cross-term of both modes.

If we use configuration (1) the linewidths of the optical spectrum u_{s1} and u_{s2} are given by

$$u_{s1} = \frac{K_{11}}{\eta_{\text{splay}}}\cdot q_\perp^2 \quad \text{and} \quad u_{s2} = \frac{K_{22}}{\eta_{\text{twist}}}\cdot q_\perp^2 \quad (2)$$

with $q_\perp^2 = (\omega_0/c)^2(\Delta n^2 + 4n_e n_0 \sin^2 \frac{1}{2}\theta)$, where ω_0 is the angular frequency of the monochromatic laser light and Δn the difference between the ordinary (n_0) and extraordinary (n_e) refractive index.

The contribution to the spectrum from both modes will be determined mainly by the value of the geometric $G_\alpha(\theta)$ terms in Eq. (1). These terms are strongly dependent on the scattering angle θ; for small angles it is the twist-bend mode that delivers the main contribution and for large angles it is the splay-bend mode.

If we choose configuration (2), only the twist-bend mode is detected. In one particular small region the normal part (with respect to the director) of the scattering vector $\mathbf{q}$ ($\mathbf{q} \equiv \mathbf{k} - \mathbf{k}'$) will vanish if the polarization of the incoming beam is perpendicular to $\mathbf{n}$ and the polarization of the scattered light is in the plane of observation. In this case the linewidth of the optical spectrum associated with the twist-bend mode becomes a pure bend mode:

$$u_{s2} = \frac{K_{33}}{\eta_{\text{bend}}}\cdot q_\parallel^2$$

with $q_\parallel^2 = (\omega_0/c)^2 n_\theta^2 \sin^2\theta$ where n_θ is the angular-dependent extraordinary refractive index.

EXPERIMENTAL ARRANGEMENT

The experimental arrangement shown in Figure 2 is similar to that used for common homodyne self beat experiments described by Ford and Benedek.[4] The light source is a 5 mW He-Ne laser (Spectra-Physics). The scattered light is detected by a photo-multiplier tube (P.M.T.) placed at a distance of about 80 cm from the scattering centre. A 2.5 mm pinhole is placed just in

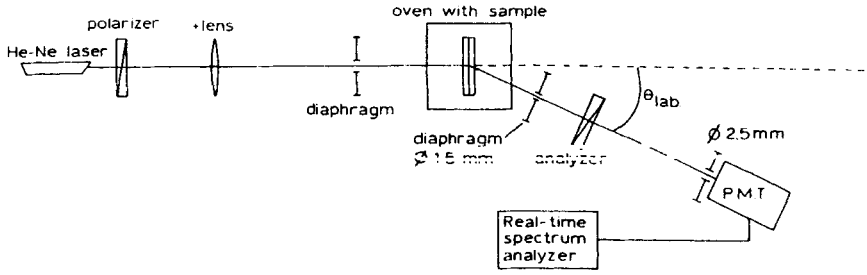

FIGURE 2 A block scheme of the experimental arrangement.

front of the detector surface; this is done to obtain a maximum value of the relative excess noise. The fluctuating anode current of the P.M.T. is fed into a frequency analyzer (Brüel & Kjaer) displaying the noise spectrum in the frequency range 12.5 Hz to 63 kHz.

The sample we used contains planar orientated liquid crystals obtained by rubbing the glass walls of the sample. The thickness of the sample is about 20 μm and is maintained by mylar spacers between the glass walls. The sample was placed in an oven stabilized with a water-pumping system which kept the temperature constant within 0.1 degree centigrade. The thermometer probe we used was a calibrated platinum resistor.

For the interpretation of our results it is necessary to know the refractive indices very precisely. They were measured by using the hollow prism method of Chatelain.[7] We measured the ordinary and extraordinary refractive indices by determining the deviation angles.

The experimental arrangement, especially the oven, limited the laboratory scattering angle θ_L, i.e. the angle outside the scattering medium, to the range 0° to 50°.

EXPERIMENTAL RESULTS

With the help of the measured refractive indices[8] we have plotted for configuration (1) the optical linewidth versus $\sin^2 \frac{1}{2}\theta$ in Figures 3 and 4 for Dibab and N_4 respectively. We can distinguish several regions, namely a splay mode region for large angles ($\theta > 10°$), a twist-bend region for small angles ($3.5° < \theta < 5.2°$), a mixture of both of these modes ($5.5° < \theta < 9°$) and a mixture of heterodyne and homodyne spectra for very small angles ($\theta < 3.5°$). We found that both types of nematic liquid crystals behave similarly in the entire nematic temperature range. A point of interest is the

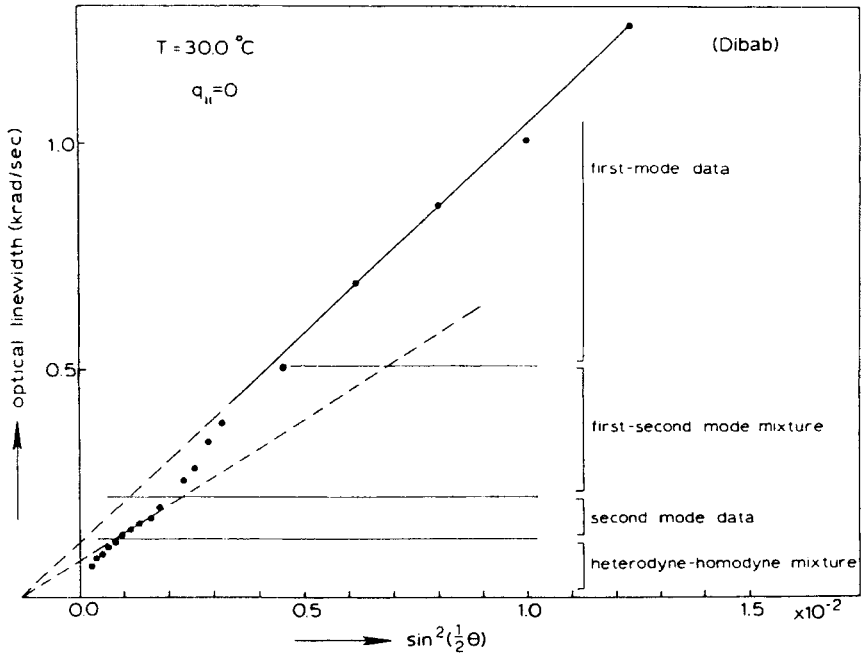

FIGURE 3 The optical linewidth plotted versus $\sin^2 \frac{1}{2}\theta$ in configuration (1), which illustrates the angle regions that can be distinguished in the case of Dibab.

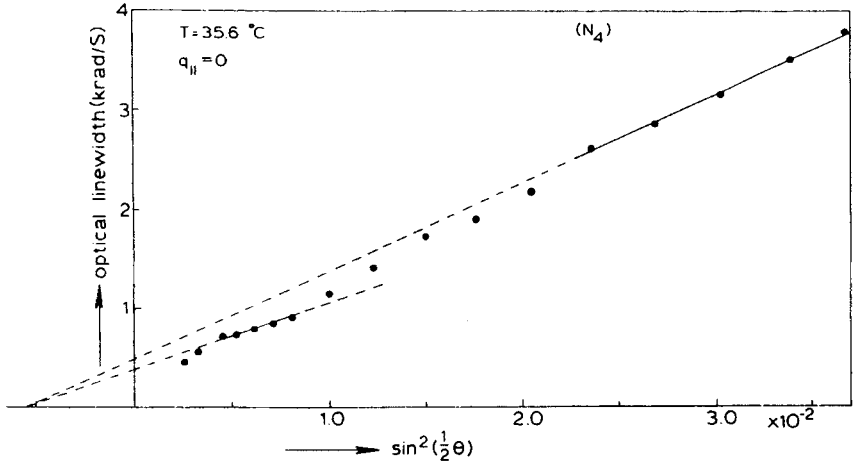

FIGURE 4 The plot of the optical linewidth versus $\sin^2 \frac{1}{2}\theta$ for N_4. Note the similarity to Dibab in Figure 3.

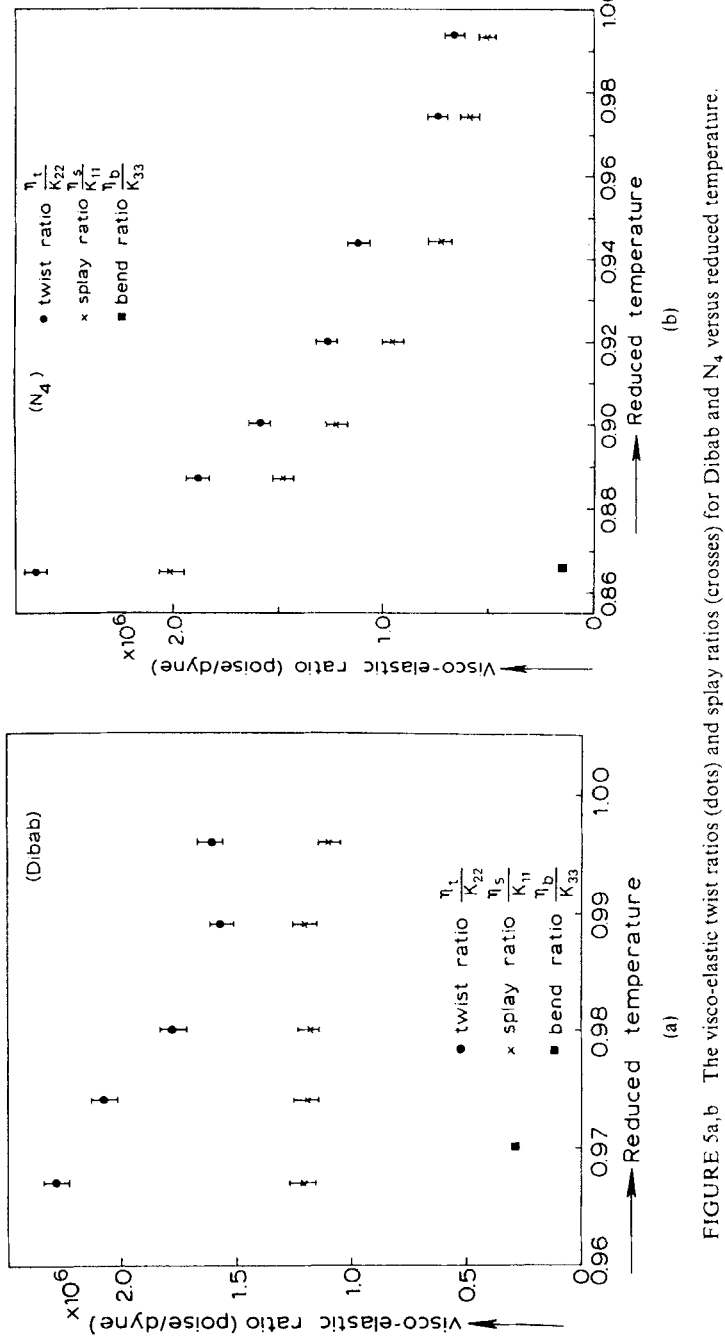

FIGURE 5a,b The visco-elastic twist ratios (dots) and splay ratios (crosses) for Dibab and N_4 versus reduced temperature.

intercept with the abscissa of the straight lines in the figures. For this point it holds that

$$\sin^2 \tfrac{1}{2}\theta = \frac{\Delta n^2}{4 n_{\parallel} n_{\perp}}$$

independent of the fluctuation mode, which is illustrated in Figures 3 and 4.

From the slope of the lines drawn we derive both the twist and splay visco-elastic ratios (cf. Eq. (2)). Figure 5 shows the thus obtained visco-elastic ratios for Dibab and N_4, where these ratios are plotted versus the reduced temperature T_{red}; T_{red} is the ratio of the measured temperature and the nematic–isotropic transition temperature in Kelvin.

The bend visco-elastic ratios are obtained by plotting the optical linewidth versus $\sin^2 \theta_L$ as is shown in Figure 6 for Dibab. In a small region ($\theta_L \approx 40°$) we do in fact expect a pure bend mode ($q_\perp = 0$). In Figure 7a,b we plotted the visco-elastic bend ratio versus the reduced temperature for N_4 and Dibab.

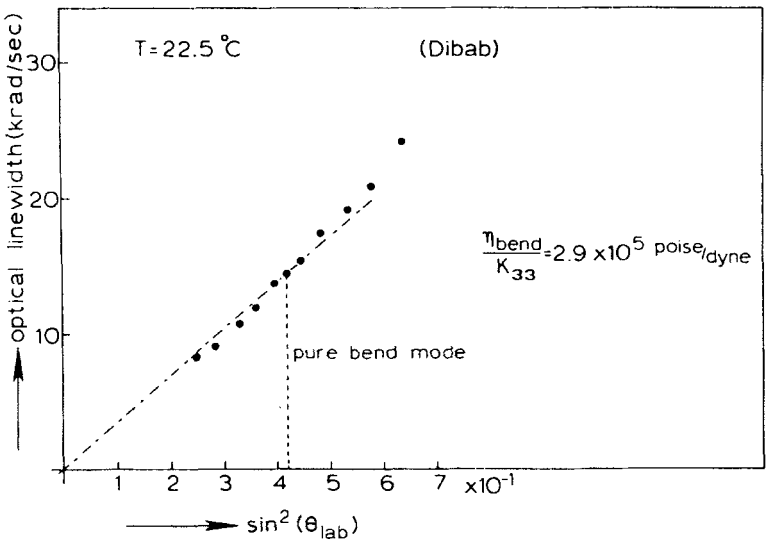

FIGURE 6 The optical linewidth plotted versus $\sin^2 \theta_L$ in the base of Dibab.

DISCUSSION

It is possible to determine the twist mode from configuration (1), but there must be very accurate measurement of the linewidths. We did this with the help of the real-time frequency analyzer.

The temperature dependent behaviour of N_4 is similar to the results reported for M.B.B.A.[1] with a strong decrease of the twist and splay ratios and

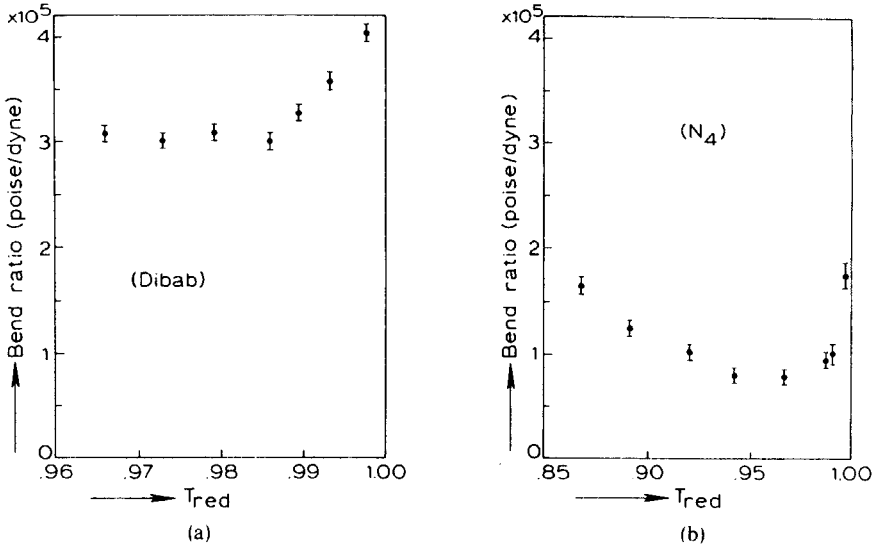

FIGURE 7a,b The visco-elastic bend ratios for Dibab and N_4 versus reduced temperature.

a strong increase of the bend ratios near the transition temperatures as a function of temperature.

Dibab, on the other hand, behaves differently. With Dibab both the bend and twist ratios are fairly constant as a function of temperature except near the transition region.

It would be interesting to measure the linewidths near the transition temperature; this would require a very good temperature stabilized system. The latter is in preparation.

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References

1. H. Fellner, W. Franklin, and S. Christensen, *Physical Review*, **A11**, no. 4, 1440 (1975).
2. M. Z. Cummins and H. L. Swinney, *Light-beat Spectroscopy, Progress in Optics*, **8**, 133 (1970).
3. Orsay Group, *The Journal of Chemical Physics*, **51**, no. 2, 816 (1969).
4. N. C. Ford and G. B. Benedek, *Physical Review Letters*, **15**, no. 16, 649 (1965).
5. C. Th. J. Alkemade, *Physica*, **25**, 1545 (1959).
6. Orsay Group, *Physical Review Letters*, **22**, no. 25, 1361 (1969).
7. O. Pellet and P. Chatelain, *Bull. Soc. Franc. Minér. Crist.*, **73**, 154 (1950).
8. D. C. van Eck, Internal report, Fysisch Laboratorium, Utrecht University.