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## Spectral analysis of light intensity fluctuations caused by orientational fluctuations in nematics

D. C. Van Eck and R. J. J. Zijlstra

Fysisch Laboratorium, Rijksuniversiteit Utrecht, The Netherlands

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**Résumé.** — Dans cet article nous discutons la technique homodyne et hétérodyne pour la détection de la densité spectrale de la lumière de laser diffusée par les cristaux liquides nématiques (n.l.c.). Nous avons étudié l'effet du bruit de fond du laser sur les expériences homodynes (section 2) et hétérodynes (section 3). Il paraît que les fluctuations d'intensité du laser n'ont d'importance que dans le cas hétérodyne. Nous montrons expérimentalement que les largeurs des lignes spectrales homodynes sont deux fois plus grandes que celles des lignes spectrales hétérodynes, ce qui indique que le processus statistique des fluctuations du directeur est Gaussien (section 5). Dans les sections 4 et 5 nous discutons brièvement les effets que la lumière diffusée superflue et le nombre des surfaces de cohérence sous-tendues par le photodétecteur ont sur nos mesures. Ni l'un ni l'autre élément perturbateur n'affecte les largeurs des lignes spectrales de la lumière diffusée. Finalement, dans la section 6, nous discutons et résumons les résultats principaux que nous avons obtenus.

**Abstract.** — In this paper we discuss the homodyne and heterodyne experimental technique for the detection of noise intensity spectra of laser light scattered by nematic liquid crystals (n.l.c.). We have studied the influence of laser noise on both homodyne (section 2) and heterodyne (section 3) experiments. It turns out that the laser intensity fluctuations are of importance only in the heterodyne case. It is shown experimentally that the linewidths of homodyne spectra are twice those of heterodyne spectra, which indicates that the statistical process of the director fluctuations is Gaussian (section 5). The influence on the measurements of stray light and the number of coherence areas subtended by the photodetector is discussed briefly in sections 4 and 5. Neither disturbing factor affects the linewidths of the light intensity spectra. In section 6 we discuss and summarize the principal results obtained.

1. **Introduction.** — Light scattering is a good tool for obtaining information about visco-elastic properties of nematic liquid crystals (n.l.c.). The local thermal fluctuations of the director give rise to an intense scattering of light. The noise spectrum of these light fluctuations is directly connected with the spectrum of the director fluctuations [1].

The light intensity fluctuations are detected with a photoelectric detector (photomultiplier). We have used two experimental techniques : the technique of homodyne detection known as self-beat spectroscopy and the technique of heterodyne detection, where the scattered light is optically mixed with the original incident laser light (local oscillator) on the detector surface [2].

The optical spectrum  $S_E(\nu)$  of laser light scattered by the long-range orientational fluctuations of the elongated molecules of a nematic is given by [1]

$$S_E(\nu) = \beta^2 E_0^2 \sum_{\alpha=1,2} G_\alpha^2 \frac{4 k_B T}{V K_\alpha(\mathbf{q})} \times \frac{\Delta\omega_\alpha}{(\Delta\omega_\alpha)^2 + (\omega - \omega_0)^2} \quad (1)$$

where  $\beta = (\epsilon_\alpha V/8 \pi R) (\omega_0/c)^2$ , a dimensionless quantity with  $\epsilon_\alpha$  the dielectric anisotropy,  $V$  the scattering volume,  $R$  the distance between  $V$  and the point of observation,  $\omega_0$  the angular frequency of the incoming field and  $c$  the velocity of light ;  $G_\alpha$  is a geometrical factor involving the director, the scattering vector  $\mathbf{q}$  and the polarizations of the incoming and scattered field ;  $k_B$  is Boltzmann's constant ;  $T$  the absolute temperature ;  $\Delta\omega = K_\alpha(\mathbf{q})/\eta_\alpha(\mathbf{q})$  with  $K_\alpha(\mathbf{q}) = K_{xx} q_\perp^2 + K_{33} q_\parallel^2$ , the elastic torque density, and  $\eta_\alpha(\mathbf{q})$  a viscosity constant of a nematic [1].

The optical line (eq. (1)) reflects the broadening of the original laser line with centre angular frequency  $\omega_0$  and consists of two Lorentzian shaped modes. Experimentally it is impossible to detect such an optical line by conventional optical apparatus, because the optical line has a centre frequency of  $5 \times 10^{14}$  Hz and a width of the order of  $10^2$  Hz. For this reason we analyze the noise spectrum  $S_{\Delta I}(\nu)$  of the intensity fluctuations of the scattered light with a photoelectric detector [3]. This noise spectrum of the light fluctuations can be calculated from the optical line (eq. (1)) with the help of an autoconvolution [4]. Hence

$$S_{\Delta I}(\nu) = \sum_{\alpha=1,2} F_{\alpha}^2 \cdot \frac{2 \Delta \omega_{\alpha}}{(2 \Delta \omega_{\alpha})^2 + \omega^2} + F_1 F_2 \frac{2(\Delta \omega_1 + \Delta \omega_2)}{(\Delta \omega_1 + \Delta \omega_2)^2 + \omega^2} \quad (2)$$

where  $F_{\alpha} = \beta^2 E_0^2 G_{\alpha}^2 k_B T/V \cdot K_{\alpha}(\mathbf{q})$  ( $\alpha = 1, 2$ ).

Note that the spectrum described by eq. (2) consists of two Lorentzians, with widths equal to those of the corresponding optical lines and a third Lorentzian with a width equal to the sum of those in the optical spectrum.

With the homodyne measuring technique we obtain the spectrum of the light intensity fluctuations. With heterodyne measurements the observed noise spectrum is proportional to the optical spectrum  $S_E(\nu)$  (cf. eq. (1)).

In the derivation of eq. (2) it was tacitly assumed that the laser is an ideal oscillator without laser light fluctuations. In addition it was assumed that the light detector subtended one coherence area. In this paper we shall discuss the influence of laser fluctuations on the measurements as well as the effect of the number of coherence areas subtended by the photomultiplier. Also the presence of background light from unwanted sources will be considered.

Using a photomultiplier (P.M.T.), we detect an anode current,  $i_a(t)$ , which is proportional to the light intensity  $I(t)$ . The relation between the noise spectrum,  $S_{\Delta i_a}(\nu)$ , of the anode current fluctuations and that of the fluctuations in the light intensity,  $S_{\Delta I}(\nu)$ , is given by [2]

$$S_{\Delta i_a}(\nu) = \langle i_a \rangle^2 \hat{S}_{\Delta I}(\nu) + S_0 \quad (3)$$

where  $\hat{S}_{\Delta I}(\nu) \equiv S_{\Delta I}(\nu)/\langle I \rangle^2$ , the normalized spectrum of the intensity fluctuations and where  $S_0$  is the frequency independent contribution to the spectral noise intensity of the anode current fluctuations. This term is associated with the fact that the photoelectric effect is subject to chance and that the number of secondary electrons per primary electron fluctuates [2]. The term  $\langle i_a \rangle^2 \cdot \hat{S}_{\Delta I}(\nu)$  in eq. (3) is the frequency independent, excess noise term.

The shot noise component  $S_0$  can be distinguished from the excess noise by two experiments. Firstly, the spectral intensity of the anode current fluctuations is measured while the P.M.T. is irradiated by scattered light. Secondly, a measurement is done with an incandescent light source that gives the same anode current. In the latter case, only shot noise will be observed (cf.  $S_0$  in eq. (3)).

**2. The effects of laser noise on the homodyne measurements.** — In practice the output of a laser fluctuates. For gas lasers these fluctuations are mainly due to gas-discharge noise [5]. In addition the excess noise spectrum,  $S_{\Delta I_L}(\nu)$ , of the laser intensity fluctuations may be influenced by line voltage.

We evaluate the influence of laser light fluctuations

on the outcome of a light scattering experiment by expressing the laser field as follows

$$E_L(t) = E_0(t) \exp \{ i(\omega_0 t + \mathbf{k}_0 \cdot \mathbf{r} + \zeta(t)) \}, \quad (4)$$

where we distinguish phase and amplitude fluctuations, described by  $\zeta(t)$  and  $E_0(t)$ , respectively. Notice that the laser field is a scalar field because the laser is linearly polarized.

Now the scattered field  $E_s(t)$  is given by

$$E_s(t) = E_0(t) \exp \{ i\zeta(t) \} \cdot \exp \{ i(\mathbf{k}_s \cdot \mathbf{r}' - \omega_0 t) \} \cdot \xi(t) \quad (5)$$

where  $\xi(t)$  describes the fluctuations of the scattering medium [6] (director-fluctuations) given by

$$\xi(t) = \beta \sum_{\alpha=1,2} G_{\alpha} n_{\alpha}(\mathbf{q}, t).$$

It should be noticed that  $\xi(t)$  is a dimensionless quantity for which it holds that  $|\xi(t)| \ll 1$  because the intensity of the scattered radiation,

$$I_s(t) \equiv \frac{1}{2} E_s(t) \cdot E_s^*(t) = \frac{1}{2} E_0^2(t) \cdot \xi^2(t),$$

is much smaller (for all scattering angles) than the original laser intensity

$$I_L(t) = \frac{1}{2} E_L(t) \cdot E_L^*(t) = \frac{1}{2} E_0^2(t).$$

In eq. (3) we are interested in the normalized spectrum of the intensity fluctuations of the scattered light. This spectrum can be derived easily with the help of eq. (5), the use of auto-correlation functions and the Wiener-Khinchine theorem [7, 8]. One finds (cf. appendix)

$$\hat{S}_{\Delta I}(\nu) = \hat{S}_{\Delta I_L}(\nu) + \hat{S}_{\Delta \xi^2}(\nu) + \hat{S}_{\Delta I_L} \circledast \hat{S}_{\Delta \xi^2} \quad (6)$$

where  $\circledast$  denotes a convolution defined by

$$\hat{S}_A \circledast \hat{S}_B = \frac{1}{2} \int_{-\infty}^{\infty} \hat{S}_A(\nu - \nu') \cdot \hat{S}_B(\nu') d\nu'.$$

We notice that the excess noise of the anode current spectrum (cf. eq. (6)) consists of three parts : i) the normalized spectrum of the laser fluctuations,  $\hat{S}_{\Delta I_L}(\nu)$ , ii) the normalized spectrum of the director fluctuations,  $\hat{S}_{\Delta \xi^2}(\nu)$ , and iii) a convolution of both spectra. If we take one mode into consideration ( $\alpha = 1$  or  $2$ ) it can easily be shown that (cf. eq. (2))

$$\hat{S}_{\Delta \xi^2}(\nu) = 4 \cdot \frac{2 \Delta \omega}{(2 \Delta \omega)^2 + \omega^2} \quad (7)$$

where  $\Delta \omega$  is the angular frequency-width at half maximum of the corresponding optical line in rad/s.

In the homodyne experiments [4] the maximum bandwidth  $\Delta \omega$  of  $\hat{S}_{\Delta \xi^2}(\nu)$  turned out to be about 60 krad/s. This means that the lowest relative spectral component of  $\hat{S}_{\Delta \xi^2}(\nu)$  (cf. eq. (7)) in which we are

interested is 10 % ( $\approx 10^{-5}$  s) of the corresponding plateau level. The maximum value of  $\hat{S}_{\Delta I_L}(v)$  we have measured for the 5 mW He-Ne laser (Spectra Physics 120) used in our experiments is  $5 \times 10^{-7}$  s. This means that we can neglect  $\hat{S}_{\Delta I_L}(v)$  with respect to  $\hat{S}_{\Delta \xi^2}(v)$  in the frequency range in which we are interested. We can neglect the convolution in eq. (6) with respect to  $\hat{S}_{\Delta \xi^2}(v)$  by looking at an upper limit

$$\begin{aligned} \hat{S}_{\Delta I_L} \circledast \hat{S}_{\Delta \xi^2} &\leq \max [\hat{S}_{\Delta I_L}(v)] \times \\ &\times \int_0^\infty \hat{S}_{\Delta \xi^2}(v') dv' = \max [\hat{S}_{\Delta I_L}(v)] \\ &= 5 \times 10^{-7} \text{ s} \end{aligned} \quad (8)$$

which is much smaller than the minimum value of  $\hat{S}_{\Delta \xi^2}(v)$  in which we were interested ( $10^{-5}$  s).

Qualitatively this result is immediately clear since the director fluctuations modulate the incoming light. The fluctuations in the incoming light are modulated also of course but this will be a second order effect.

**3. The effect of laser noise on the heterodyne measurements.** — In the case of heterodyne detection we shall follow the same procedure as before. The detected field consists of two parts, one being the scattered field and the other being the field of the local oscillator. Hence

$$E(t) = E_0(t) \cdot \exp \{ i(\mathbf{k}_s \cdot \mathbf{r}' - \omega_0 t + \zeta(t)) \} [\zeta(t) + \alpha] \quad (9)$$

where  $\alpha$  denotes a fraction of the amplitude of the original field (cf. eq. (4)). In heterodyne experiments it is important that the contribution of the local oscillator should be chosen much larger than the contribution of the scattered field [2]; thus we assume that for the ratio  $A$  of the corresponding intensities it holds that

$$A \equiv (\langle \zeta(t)^2 \rangle / \alpha^2) \ll 1.$$

With the help of eq. (9) the intensity of the local field becomes

$$I(t) = \frac{1}{2} E(t) E^*(t) = I_L(t) [\zeta^2(t) + \alpha^2 + 2\alpha\zeta(t)]. \quad (10)$$

Again we can calculate the normalized excess noise spectrum of the detected light intensity. One obtains [7, 8] (cf. appendix)

$$\begin{aligned} \hat{S}_{\Delta I}(v) &= \hat{S}_{\Delta I_L}(v) + 4A\hat{S}_{\Delta \xi}(v) + A^2\hat{S}_{\Delta \xi^2}(v) + \\ &+ A^2\hat{S}_{\Delta I_L} \circledast \hat{S}_{\Delta \xi^2} + 4A\hat{S}_{\Delta I_L} \circledast \hat{S}_{\Delta \xi}. \end{aligned} \quad (11)$$

Now we meet five contributions to the total normalized excess spectrum of the light intensity fluctuations, the second term of which on the right hand side of eq. (11) is the one we are interested in.

If  $A \ll 1$ , we can neglect both the homodyne term  $\propto \hat{S}_{\Delta \xi^2}$  and the convolution term  $\propto \hat{S}_{\Delta I_L} \circledast \hat{S}_{\Delta \xi^2}$ . Hence the spectrum of the anode current fluctuations (cf. eq. (3)) now becomes

$$S_{\Delta i_a} = S_0 + \langle i_a \rangle^2 [4A \{ \hat{S}_{\Delta \xi}(v) + \hat{S}_{\Delta I_L} \circledast \hat{S}_{\Delta \xi} \} + \hat{S}_{\Delta I_L}(v)]. \quad (12)$$

From eq. (12) it is obvious that when the intensity of the local oscillator is chosen too large with respect to the scattered intensity, «  $A$  » will decrease so much that the normalized laser intensity spectrum  $\hat{S}_{\Delta I_L}(v)$  dominates the whole intensity spectrum.

At first sight the convolution term in eq. (12) cannot be neglected (cf. eq. (8)). In order to prove that the contribution of the convolution term in eq. (12) can be ignored in practice, we define a spectral bandwidth  $\Delta v$  for an arbitrary spectrum as follows

$$\Delta v = \int_0^\infty \hat{S}_{\Delta I}(v') dv' / \hat{S}_{\Delta I}(0). \quad (13)$$

In most practical cases it holds that the bandwidth of the laser field fluctuations (read : local oscillator) is much smaller than the bandwidth of the scattered field. This means we can approach the convolution as follows (with eq. (13))

$$\begin{aligned} \hat{S}_{\Delta I_L} \circledast \hat{S}_{\Delta \xi} &\approx \hat{S}_{\Delta \xi}(v) \int_0^\infty \hat{S}_{\Delta I_L}(v') dv' = \\ &= \hat{S}_{\Delta \xi}(v) \cdot \Delta v_L \cdot \hat{S}_{\Delta I_L}(0). \end{aligned} \quad (14)$$

Experimentally it turns out that

$$\Delta v_L \cdot S_{\Delta I_L}(0) \approx 10^{-4} \ll 1.$$

The spectrum for the anode current fluctuations now becomes (cf. eq. (12))

$$S_{\Delta i_a}(v) \approx S_0 + \langle i_a \rangle^2 \{ 4A\hat{S}_{\Delta \xi}(v) + \hat{S}_{\Delta I_L}(v) \}. \quad (15)$$

In section 5 we shall use this expression for the discussion of the experimental results.

**4. Effect of stray light and of coherence area.** — A disturbing factor in light scattering experiments is the occurrence of unwanted stray light that is detected by the photoelectric detector. We can divide this stray light into three categories : i) Daylight, ii) light from electric lamps which is modulated with the line frequency, iii) stray laser light.

Daylight : If we detect very low light intensities, stray light of daylight may play a role. We suppose that this light is not correlated with the scattered light, and gives rise only to shot noise. So the light intensity  $\langle I \rangle$ , observed by the detector is

$$\langle I \rangle = \langle I_s \rangle + \langle I_d \rangle,$$

where  $I_s$  the scattered light intensity and  $I_d$  the intensity of the daylight. Detecting this total light intensity with a P.M.T. we obtain for the fluctuations of the associated anode current the following spectrum (cf. eq. (3))

$$S_{\Delta i_a}(v) = S_0 + \langle i_a \rangle^2 \frac{S_{\Delta I_s}(v)}{(\langle I_s \rangle + \langle I_d \rangle)^2}. \quad (16)$$

We see that the frequency dependence of the excess term is not influenced although its absolute value is. Stray light of electric light sources : The detector can be irradiated by light from electric sources. This stray light shows an excess noise spectrum  $S_{\Delta I_e}(v)$  with in most cases a strong 50 Hz component, due to the line frequency. The intensity  $I(t)$  now becomes

$$I(t) = I_s(t) + I_e(t).$$

So the anode current noise spectrum becomes

$$S_{\Delta i_a}(v) = S_0 + \frac{S_{\Delta I_s}(v) + S_{\Delta I_e}(v)}{(\langle I_s \rangle + \langle I_e \rangle)^2} \langle i_a \rangle^2. \quad (17)$$

Now we observe two independent light intensity spectra. By applying appropriate screening, however, the influence of this type of stray light could be made negligibly small.

Stray laser light : The homodyne experiments can be disturbed by weak stray light from the laser, which is correlated with the scattered light we are interested in. In this case besides homodyne mixing also heterodyne mixing of the scattered light may occur. This would give rise to additional noise spectra in the anode current (cf. eqs. (11) and (3)). In practice, however, this stray light can be kept small enough so that these effects cannot be observed, at least, for scattering

angles larger than 5 degrees. At small angles, however, these effects become apparent and cannot be avoided, since stray light also preferentially occurs in the forward direction.

Coherence area : Finally we shall discuss the influence of the number ( $N_c$ ) of coherence areas  $A_c$  subtended by the photo-sensitive area  $A_d$  of the photomultiplier ( $N_c = A_d/A_c$ ). Taking this number into account Van Rijswijk [9] and Fijnaut [10] found that for homodyne scattering experiments the expression for the spectral noise intensity of the current fluctuations becomes (cf. eqs. (3) and (7))

$$S_{\Delta i_a}(v) = S_0 + \langle i_a \rangle^2 H(N_c) \hat{S}_{\Delta \xi^2}(v) \quad (18)$$

where  $H(N_c)$  is a weighting function with a complicated behaviour. In extreme cases however, this weighting function becomes simple [10]

$$H(N_c) \begin{cases} = 1 & \text{if } N_c \ll 1 \\ = \frac{1}{N_c} & \text{if } N_c \gg 1. \end{cases} \quad (19)$$

It implies that the contribution of different coherence areas are statistically independent so that the corresponding noise intensities can be added quadratically. Studying the normalized current spectrum for low frequencies ( $\omega \ll \Delta\omega$ ), we obtain

$$\begin{aligned} \frac{S_{\Delta i_a}(0) - S_0}{\langle i_a \rangle^2} &= H(N_c) \hat{S}_{\Delta \xi^2}(0) \\ &= \begin{cases} \hat{S}_{\Delta \xi^2}(0) & \text{if } N_c \ll 1 \\ \frac{1}{N_c} \hat{S}_{\Delta \xi^2}(0) = \frac{A_c}{A_d} \hat{S}_{\Delta \xi^2}(0) & \text{if } N_c \gg 1. \end{cases} \end{aligned} \quad (20)$$

We see for large detector areas the excess spectrum becomes inversely proportional to  $A_d$ .

**5. Experimental tests.** — **5.1 HOMODYNE EXPERIMENTS.** — We describe some test experiments with a nematic liquid crystal for which we have chosen the nematic p-p'-dibutylazoxybenzene (DIBAB) which has its mesomorphic range at room temperature. According to eq. (1.6) we expect a set of three Lorentzians in the spectrum intensity of the noise in the scattered light. However, in our test experiment we have selected one mode ( $\alpha = 2$ ) corresponding to one of these Lorentzians, using crossed polarizers, the director being chosen in the plane of observation [1, 4]. We have measured the spectrum of the anode current fluctuations of the P.M.T. as a function of the scattering angle  $\theta_L$ , where  $\theta_L$  was varied from  $5^\circ$  to  $60^\circ$ . The relative excess noise,  $S_{\Delta i_a}^R(v)$ , which is the noise in excess of shot noise relative to shot noise (cf. eq. (3)),

$$S_{\Delta i_a}^R(v) \equiv (S_{\Delta i_a}(v) - S_0)/S_0,$$

is logarithmically plotted *versus* frequency in figure 1. Since the excess noise is proportional to  $\langle I_s \rangle^2$  and  $S_0$  is proportional to  $\langle I_s \rangle$ , for the relative excess noise it holds (cf. eqs. (3) and (7))

$$S_{\Delta i_a}^R(v) \propto \langle I_s \rangle \cdot \frac{2 \Delta\omega_2}{(2 \Delta\omega_2)^2 + \omega^2} \quad (21)$$

where  $2 \Delta\omega_2$  is defined as the bandwidth of the corresponding optical line (cf. eq. (1)).

From figure 1 it is apparent that the low-frequency plateaus of the noise spectra as well as the frequency widths of the spectra depend strongly on the scattering angle. This can be explained as follows. For low frequency ( $\omega \ll \Delta\omega_2$ ) the spectral values  $S_{\Delta i_a}^R(0)$  are in first approximation, inversely proportional to  $q^4$ , where  $\mathbf{q} \equiv \mathbf{k}_0 - \mathbf{k}_s$ , the scattering vector, where  $\mathbf{k}_0$  and  $\mathbf{k}_s$  are the wave vectors of the incident and scattered laser light respectively [4]. This behaviour can be explained as follows. For low frequencies it holds that (cf. eq. (21)).

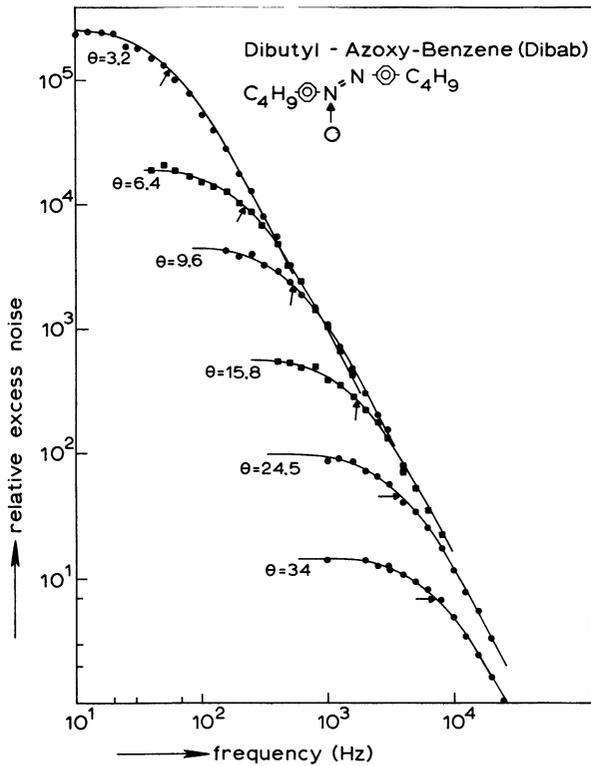


Fig. 1. — The relative excess noise  $S_{\Delta i_a}^R(\nu)$  versus frequency for different scattering angles  $\theta$ .

$$S_{\Delta i_a}^R(0) \propto \langle I_s \rangle \cdot \frac{1}{2 \Delta \nu_2} \quad (22)$$

Making a rough estimate for the elasticities ( $K_x(\mathbf{q}) \approx \bar{K}q^2$ ) and viscosities ( $\eta_x(\mathbf{q}) \approx \bar{\eta}$ ), we may write  $\langle I_s \rangle \propto (\bar{K}q^2)^{-1}$  and  $\Delta \nu \approx \bar{\eta}/\bar{K}q^2$ . Now the relative excess noise,  $S_{\Delta i_a}^R(0)$ , becomes

$$S_{\Delta i_a}^R(0) \propto \frac{\bar{\eta}}{\bar{K}^2 q^4} \quad (23)$$

If the amplitude of the scattering vector  $\mathbf{q}$  is approximated by  $2\bar{n}(\omega_0/c) \sin(\frac{1}{2}\theta)$ , where  $\bar{n}$  is the averaged refractive index, the relative excess noise becomes inversely proportional to  $\sin^4(\frac{1}{2}\theta)$ . This is illustrated in figure 2 where we have plotted  $S_{\Delta i_a}^R(0)$  versus  $\sin^4(\frac{1}{2}\theta)$  double logarithmically. We observe in first approximation a good agreement with theory.

For high frequencies ( $\omega \gg \Delta\omega_2$ ) the relative excess noise plotted in figure 1 shows a tendency to become independent of the scattering angle. With eq. (21) this is explained as follows. For high frequencies ( $\omega \gg \Delta\omega_2$ ) it holds that

$$S_{\Delta i_a}^R(\nu) \propto \frac{1}{\bar{K} \cdot q^2} \cdot \frac{\bar{K} \cdot q^2}{\bar{\eta}} \cdot \frac{1}{\nu^2} \propto \frac{1}{\nu^2} \quad (24)$$

which is independent of  $|\mathbf{q}|$  or the scattering angle  $\theta$ .

5.2 HETERODYNE EXPERIMENTS. — In section 1 we have shown theoretically that the linewidth of the

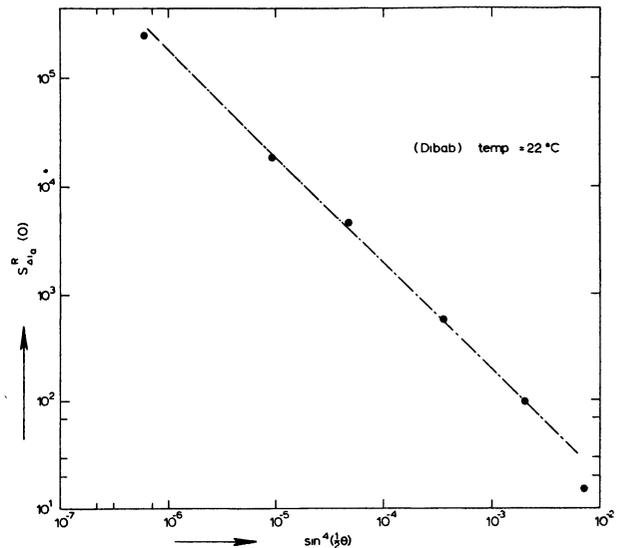


Fig. 2. — The low frequency plateau levels of the relative excess noise ( $S_{\Delta i_a}^R(0)$ ) versus  $\sin^4(\frac{1}{2}\theta)$ , where  $\theta$  is the scattering angle inside the scattering medium. The slope of the dashed line is  $-1$ .

homodyne spectrum is twice that of the heterodyne spectrum on the assumption that the scattering field is Gaussian. In order to check this supposition we did a heterodyne experiment in addition to a homodyne one. We have used the same geometrical configuration, as far as polarizations of the light and the director are concerned, as in the homodyne experiment. In order to make a heterodyne experimental set-up we have inserted in the homodyne experimental set-up [4] a block-prism that splits the original laser beam into two beams numbered (1) and (2) (cf. Fig. 3) which both cross the sample at the same point. As a consequence the detected light is partly scattered light originating from beam (1) and partly unscattered light from beam (2). The latter serves as a local oscillator signal in the photoelectric heterodyne mixing experiment. In this way we get optimum mixing on the detector surface. With this set-up we detect an anode current spectrum which is a mixture of a homodyne, a heterodyne and a laser intensity spectrum. Therefore (cf. eq. (11))

$$S_{\Delta i_a}(\nu) = S_0 + \langle i_a \rangle^2 [\hat{S}_{\Delta i_L}(\nu) + 4 A \hat{S}_{\Delta i_L} \otimes \hat{S}_{\Delta \xi} + 4 A \hat{S}_{\Delta \xi}(\nu) + A^2 \hat{S}_{\Delta \xi^2}(\nu)] \quad (25)$$

where  $A = \langle \xi^2 \rangle / \alpha^2 \approx 1.25 \times 10^{-3} \ll 1$ .

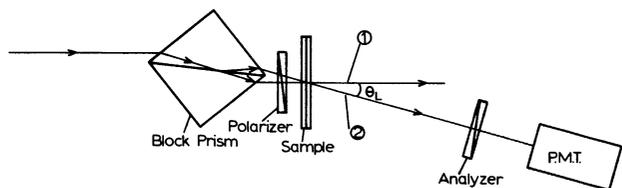


Fig. 3. — Experimental arrangement for heterodyne detection. (1) Denotes the incident beam and (2) the beam of the local oscillator.

In figure 4 we have plotted the spectrum of the light fluctuations (dots) for a heterodyne experiment. For low frequencies we expect a frequency independent level. However, we observe here a deviation from a Lorentzian shaped spectrum. This is due to the fact that other spectra interfere with the optical line  $\hat{S}_{\Delta\xi}(\nu)$  (cf. eq. (25)). For this reason we also measured the spectrum of the local oscillator fluctuations (open circles in Fig. 4) by extinguishing the incident beam (1 in Fig. 3). We would point out that this spectrum is

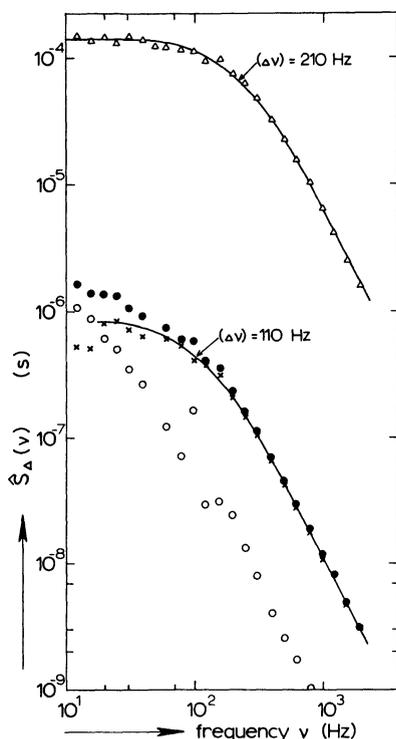


Fig. 4. — The homodyne spectrum  $\hat{S}_{\Delta\xi^2}(\nu)$  (triangles), the heterodyne spectrum  $\hat{S}_{\Delta I}(\nu)$  (dots), the spectrum of the local oscillator  $\hat{S}_{\Delta I_L}(\nu)$  (open circles) and the optical spectrum  $\hat{S}_{\Delta I}(\nu) - \hat{S}_{\Delta I_L}(\nu)$  (crosses) versus frequency. Two theoretical Lorentzians (full curves) are added for comparison.

not the same as the spectrum of the laser intensity fluctuations we mentioned in section 2, where

$$\max \{ S_{\Delta I_L}(\nu) \} \approx 5 \times 10^{-7} \text{ s}.$$

This is due to the fact that in this configuration the observed noise is a mixture of laser noise and the result of heterodyne mixing of the local oscillator signal and light scattered in the forward direction. As a consequence the normalized noise intensity is determined by the forward scattering rather than by fluctuations in the laser output (cf. eq. (15)). In addition, besides single scattering, double scattering may occur as well as scattering from other sources than the nematic.

Fortunately the bandwidth  $\Delta\nu$  of the normalized spectrum of the local oscillator is much smaller than the bandwidth of the normalized spectrum of the

director fluctuations (cf. eq. (8) and the spectra in figure 4 (dots and open circles)). For this reason we neglect the convolution term in eq. (12) and obtain the normalized optical spectrum by subtracting the normalized spectrum of the local oscillator from the total heterodyne normalized spectrum (cf. eq. (25)). This corrected normalized spectrum is represented in figure 4 (crosses). The full curve represents the best fitted Lorentzian spectrum. This curve and the experimental data are in good agreement.

For comparison we have added the corresponding homodyne normalized spectrum of the scattered light intensity fluctuations  $\hat{S}_{\Delta\xi^2}(\nu)$  in figure 4 (triangles). This spectrum is obtained by extinguishing the beam of the local oscillator (i.e. beam (2) in figure 3). The plateau level of the normalized intensity of these fluctuations is much higher ( $\hat{S}_{\Delta\xi^2}(0) \approx 1.4 \times 10^{-4} \text{ s}$ ) than that of the normalized heterodyne spectrum ( $\hat{S}_{\Delta\xi}(0) \approx 8 \times 10^{-7} \text{ s}$ ) which is explained by the fact that (cf. eq. (25)) in the homodyne case  $A = 1$  and terms  $\propto \hat{S}_{\Delta\xi}(\nu)$  are equal to zero. With regard to the bandwidth, we notice that the width of the homodyne spectrum is twice that of the heterodyne spectrum ( $210 \pm 10$  instead of  $110 \pm 5 \text{ Hz}$  in figure 4), which is in good agreement with the theoretical assumption that the statistical process of the director fluctuations is Gaussian.

5.3 THE RATIO OF COHERENCE AREA TO THE EFFECTIVE DETECTOR AREA. — In order to check eq. (20) we have measured the spectral noise intensity of the anode current fluctuations for low frequencies (i.e.  $\nu \ll \Delta\nu$ ) in a homodyne light scattering experiment on the nematic p-methoxy-p'-n-butylazoxybenzene ( $N_4$ ) at room temperature, using several detector areas  $A_d$ . Again we have used the geometrical configuration where only the second mode ( $\alpha = 2$ ) is involved.

In figure 5 we plotted the quantity (cf. eq. (18))

$$\frac{(S_{\Delta i_a}(0) - S_0) / \langle i_a \rangle^2}{\hat{S}_{\Delta\xi^2}(0)} = H(N_c) \quad (26)$$

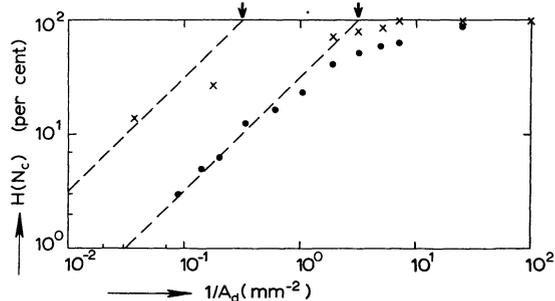


Fig. 5. — The percentage ratio

$$H(N_c) = \{ (S_{\Delta i_a}(0) - S_0) / \langle i_0 \rangle^2 \} / \hat{S}_{\Delta\xi^2}(0)$$

versus the reciprocal detector area. The dots and crosses represent the results of experiments done with a large and a small scattering volume respectively. The arrows indicate the corresponding effective reciprocal coherence areas.

double logarithmically *versus* the reciprocal detector area  $A_d$ . The numerator on the left hand side of eq. (26) follows directly from the measured data, whereas the denominator is determined theoretically with the help of eq. (21). In the experiment we changed the effective detector area  $A_d$  by placing diaphragms in front of the detector. The diameter of the diaphragms placed at a distance  $R = 0.47$  m from the scattering volume varied from 0.113 to 5.9 mm; the coherence area as determined by the dimensions of the scattering volume was kept constant. Since  $A_d$  is proportional to the number  $N_c$  of the subtended coherence areas of the detector, one expects that  $H(N_c)$  will become 100% at large values of  $1/A_d$  and will be proportional to  $1/A_d$  at small values of  $1/A_d$  (cf. eq. (20)). This behaviour is demonstrated in figure 5 (dots) where the slope of the double logarithmic plot at small values of  $1/A_d$  is equal to one. For the coherence area chosen, it was not possible, however, to fully reach the 100% level at large values of  $1/A_d$  because the detected light intensity became inconveniently small and stray light (cf. eq. (17)) disturbed the experiment. Therefore we repeated the same measurement for a large coherence area which could be obtained by choosing smaller dimensions for the scattering volume. The results are plotted as crosses. Note that in this case the value of  $H(N_c)$  does indeed become equal to 100% at high values of  $1/A_d$ . So eq. (20) is fully vindicated by these experimental results.

Inversely these kinds of experiments allow us to determine the effective area of coherence ( $A_c$ ) from the measured curves (cf. Fig. 5). The intersection of the extrapolated linear part with the 100% axis immediately gives us the reciprocal value  $A_c^{-1}$  of the coherence area (indicated by the arrow in figure 5). Thence we obtained  $A_c = 0.31$  mm<sup>2</sup> for the large scattering volume (dots). In order to complete the linear part for a small scattering volume (crosses) one simply applies a translation of the dots along the  $1/A_d$  axis. It was found in this case (crosses) that  $A_c = 3.03$  mm<sup>2</sup>. This method is preferable to one that used elementary formulas known from optics, which are not always applicable.

**6. Discussion and conclusions.** — The homodyne experiments have proved that the spectra of the intensity fluctuations are consistent with the theoretical predictions if we assume that the director fluctuations are Gaussian. This assumption is justified by the experimental proof that the linewidth of the homodyne spectrum is twice (within 5%) the linewidth obtained from a spectrum by heterodyne experiments. A second experimental proof has been given recently by Timmermans [11] who measured photon time interval distributions [12]. He was able to calculate the higher-order moments of the light intensity fluctuations [13], caused by laser light scattering on nematics. The observations he has made so far show again that the

light intensity fluctuations are due to Gaussian director fluctuations.

The homodyne experiments, which are much easier than heterodyne ones, can be used for the determination of the optical line spectrum. The optical linewidth  $\Delta\omega = 2\pi\Delta\nu$  contains information about the viscoelastic properties of nematic liquid crystals.

The effect of the laser noise can be neglected in the homodyne experiments (cf. sections 2 and 5) if the normalized intensity fluctuations  $\hat{S}_{\Delta I_L}(\nu)$  of the laser are much smaller than the normalized spectrum of the scattered light intensity  $\hat{S}_{\Delta \xi^2}(\nu)$ . In our experiments it must hold (cf. eq. (7)) that  $\hat{S}_{\Delta I_L}(\nu) \ll 10^{-5}$  s if frequencies up to 10 kHz are used. Note that the observed shot noise (cf. eq. (3)) does not play any role in this discussion.

In the heterodyne experiments another factor comes into play, i.e. the ratio  $A$  between the intensity of the scattered light and the intensity of the local oscillator light. Equation (12) shows that for this ratio it should hold  $1 \ll A^{-1} \ll \hat{S}_{\Delta \xi^2}(\nu)/\hat{S}_{\Delta I_L}(\nu)$ . If not, the spectrum obtained from heterodyne experiments must be corrected for the laser intensity fluctuations, as is illustrated in section 5.2.

The number of coherence areas subtended by the detector affects only the height of the normalized spectra of the scattered light, not their shape. Neither is the relative excess noise affected by the number of coherence areas subtended by the photoelectric detector.

**Appendix.** — A. DERIVATION OF THE NORMALIZED SPECTRUM OF THE LIGHT INTENSITY FLUCTUATIONS IN THE HOMODYNE CASE. — Starting from eq. (5) we can evaluate the correlation function,

$$\varphi_{I_s}(\tau) \equiv \langle I_s(\tau) \cdot I_s(t + \tau) \rangle,$$

of the scattered light intensity. Hence

$$\varphi_{I_s}(\tau) = \frac{1}{4} \langle E_0^2(t) \cdot E_0^2(t + \tau) \cdot \xi^2(t) \cdot \xi^2(t + \tau) \rangle. \quad (\text{A.1})$$

If the laser and orientational fluctuations are statistically independent we may write

$$\varphi_{I_s}(\tau) = \varphi_{I_L}(\tau) \varphi_{\xi^2}(\tau). \quad (\text{A.2})$$

If stationarity of the light intensity is assumed, it holds for the intensity fluctuations,  $\Delta I_s(t) = I_s(t) - \langle I_s(t) \rangle$ , that

$$\varphi_{\Delta I_s}(\tau) = \varphi_{I_s}(\tau) - \langle I_s \rangle^2.$$

Defining a normalized correlation function,

$$\psi_{\Delta I}^{(v)} \equiv \varphi_{\Delta I}^{(v)} / \langle I \rangle^2$$

we can write with eq. (A.2) for the fluctuations of the scattered light intensity

$$\psi_{\Delta I_s}(\tau) = \frac{(1 + \psi_{\Delta I_L}(\tau)) (1 + \psi_{\Delta \xi^2}(\tau)) \cdot \langle \xi^2 \rangle^2 \cdot \langle I_L \rangle^2}{\langle I_s \rangle^2} - 1 \quad (\text{A.3})$$

where  $\psi_{\Delta I_L}(\tau)$  and  $\psi_{\Delta \xi^2}(\tau)$  denote the normalized correlation functions of the laser and director fluctuations, respectively. Because  $\langle I_s \rangle^2 = \langle \xi^2 \rangle \cdot \langle I_L \rangle^2$  eq. (A.3) becomes

$$\psi_{\Delta I_s}(\tau) = \psi_{\Delta I_L}(\tau) + \psi_{\Delta \xi^2}(\tau) + \psi_{\Delta I_L}(\tau) \cdot \psi_{\Delta \xi^2}(\tau). \quad (\text{A.4})$$

With the help of the Wiener-Khinchine theorem [7], the normalized spectrum of the scattered light intensity fluctuations becomes (cf. eq. (6))

$$\hat{S}_{\Delta I_s}(v) = \hat{S}_{\Delta I_L}(v) + \hat{S}_{\Delta \xi^2}(v) + \hat{S}_{\Delta I_L} \circledast \hat{S}_{\Delta \xi^2} \quad (\text{A.5})$$

$$\text{where } \hat{S}_A \circledast \hat{S}_B \equiv \frac{1}{2} \int_{-\infty}^{\infty} S_A(v-v') S_B(v') dv'$$

**B. DERIVATION OF THE NORMALIZED INTENSITY SPECTRUM IN THE HETERODYNE CASE.** — From eq. (10) it follows for the correlation function,  $\varphi_I(\tau)$ , for the light intensity

$$\varphi_I(\tau) = \varphi_{I_L}(\tau) [\varphi_{\xi^2}(\tau) + \varphi_{\xi}(\tau) \cdot 4\alpha^2 + \alpha^4]. \quad (\text{B.1})$$

In eq. (B.1) we have omitted the terms containing  $\langle \xi(t+\tau) \rangle$ ,  $\langle \xi^2(t) \xi(t+\tau) \rangle$  and  $\langle \xi^2(t+\tau) \xi(t) \rangle$ ,

$\langle \xi(t) \rangle$ , which are zero. With eq. (B.1) we can express  $\psi_{\Delta I}(\tau)$  in the other normalized correlation functions i.e.  $\psi_{\Delta I_L}(\tau)$ ,  $\psi_{\Delta \xi^2}(\tau)$  and  $\psi_{\Delta \xi}(\tau)$ . Hence (cf. eq. (A.3))

$$\psi_{\Delta I}(\tau) = \frac{\langle I_L \rangle^2}{\langle I \rangle^2} (1 + \psi_{\Delta I_L}(\tau)) [(1 + \psi_{\Delta \xi^2}(\tau)) \langle \xi^2 \rangle^2 + (1 + \psi_{\Delta \xi}(\tau)) \langle \xi^2 \rangle 4\alpha^2 + \alpha^4] - 1. \quad (\text{B.2})$$

Because  $\langle \xi^2 \rangle / \alpha^2 \ll 1$  we may write for the intensity

$$\langle I \rangle \approx \langle I_L(t) \rangle \alpha^2 [1 + A] \approx \langle I_L(t) \rangle \cdot \alpha^2. \quad (\text{B.3})$$

With eq. (B.2) we obtain

$$\psi_{\Delta I}(\tau) = \psi_{\Delta I_L}(\tau) + 4A\psi_{\Delta \xi}(\tau) + A^2 \{ \psi_{\Delta \xi^2}(\tau) + \psi_{\Delta I_L}(\tau) \cdot \psi_{\Delta \xi^2}(\tau) \} + 4A\psi_{\Delta I_L}(\tau) \cdot \psi_{\Delta \xi}(\tau). \quad (\text{B.4})$$

Again we use the Wiener-Khinchine theorem [7] to obtain the normalized spectrum of the intensity fluctuations. Hence

$$\hat{S}_{\Delta I}(v) = \hat{S}_{\Delta I_L}(v) + 4A\hat{S}_{\Delta \xi}(v) + A^2 \hat{S}_{\Delta \xi^2}(v) + A^2 \hat{S}_{\Delta I_L} \circledast \hat{S}_{\Delta \xi^2} + 4A\hat{S}_{\Delta I_L} \circledast \hat{S}_{\Delta \xi}. \quad (\text{B.5})$$

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