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## Liquid Crystals

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### Determination of $K_i$ ( $i = 1-3$ ) and $\mu_j$ ( $j = 2-6$ ) in 5CB by observing the angular dependence of Rayleigh line spectral widths

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## Determination of $K_i$ ( $i = 1-3$ ) and $\mu_j$ ( $j = 2-6$ ) in 5CB by observing the angular dependence of Rayleigh line spectral widths

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Using Parodi's relation, all of the Leslie viscosity coefficients, except  $\mu_1$ , together with the Frank elastic constants have been measured successfully by the photon correlation spectroscopy of Rayleigh scattered light. The values so determined are in good agreement with those previously determined from shear flow experiments by Chmielewski and by Skarp *et al.* The polar angle dependence of mode 1 spectral width is proposed as a novel method for the measurement of  $\mu_1$  and for the experimental confirmation of Parodi's relation.

### 1. Introduction

Since the pioneering work by the Orsay Liquid Crystal Group [1-4], many Rayleigh light scattering experiments have been made to determine the Frank elastic constants and Leslie viscosity coefficients [5-26]. As far as we know, however, no one has been successful in determining all of the Leslie viscosity coefficients precisely by the Rayleigh light scattering method. One reason for this failure is the lack of accuracy in the elastic constant ratios,  $K_1/K_2$  and  $K_3/K_2$ , obtained from the scattering angle dependence of the intensity [20, 21, 23].

In this work, we use photon correlation spectroscopy and accurately measure the spectral widths to obtain not only the ratios of the viscosity coefficients and the elastic constants,  $\mu_i/K_n$ s, but also the ratios of the elastic constants,  $K_n/K_m$ s. Depolarized scattering geometries were used to measure  $\eta_{\text{splay}}/K_1$ , which greatly improve the accuracy; the inaccuracy in  $\eta_{\text{splay}}/K_1$  has been another obstacle [23, 27].

We then determine the twist elastic constant  $K_2$  by observing the scattered light intensity or spectral widths as a function of an applied electric field [25, 26]. Thus, using Parodi's relation, all of the Leslie viscosity coefficients, except  $\mu_1$ , are successfully measured by the Rayleigh light scattering method; the values so determined are in good agreement with those previously determined from shear flow experiments by Chmielewski [28] and by Skarp *et al.* [29]. Finally, a novel method is proposed for the determination of  $\mu_1$  and for the experimental confirmation of Parodi's relation: the polar angle dependence of the mode 1 spectral width, which can be performed by cell rotation about the optic axis. The importance of this has already been insisted in the intensity measurement [27].

### 2. Experimental procedures

The experimental set up for photon correlation spectroscopy and the preparation of sample cells have already been described in the previous paper [23]. The material, 4-*n*-pentyl-4'-cyanobiphenyl (5CB), was supplied by Merck Japan Limited; it was used without further purification. To illustrate the accuracy of our measurement, figure 1 shows an observed homodyne signal in linear and logarithmic scales.

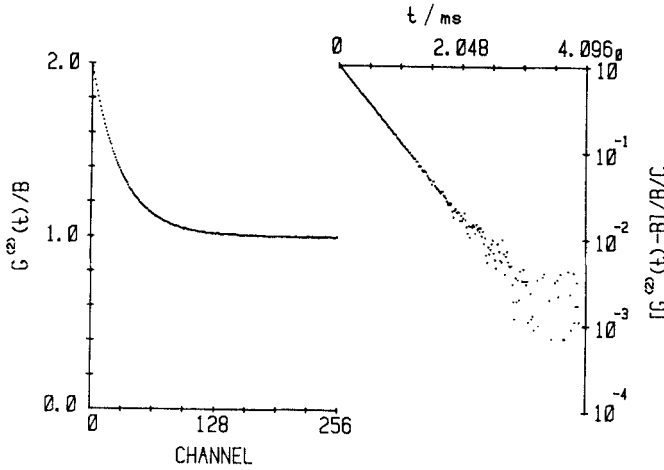


Figure 1. Example of an observed autocorrelation function.

The determination procedure is as follows

(1)  $\eta_{\text{splay}}/K_1$ ,  $\eta_{\text{twist}}/K_2$  and  $\eta_{\text{bend}}/K_3$  were obtained by using the scattering geometries with a pure single deformation summarized in figure 2. The accuracy of  $\eta_{\text{splay}}/K_1$  was greatly improved because the (O-E) and (E-O) depolarizations were used instead of the (E-E) polarization, as is clearly seen by comparing figure 3 with the corresponding figure 4 of [23]. Here, E and O refer to extraordinary and ordinary, respectively, and the first letter in parenthesis shows the state of polarization of the incident light and the second shows that of the scattered light.

(2)  $K_1/K_2$  was determined by the van der Meulen-Zijlstra method [12-14] using the mixed mode scattering geometries with  $q_{\parallel} = 0$  summarized in figure 2.

(3) The mode 2 spectral width is given by

$$\Gamma_2 = \frac{(K_3/K_2)q_{\parallel}^2 + q_{\perp}^2}{(\gamma_1/K_2) - \frac{q_{\parallel}^2}{aq_{\perp}^2 + cq_{\parallel}^2}}, \tag{1}$$

where

$$\begin{aligned} a &= K_2\eta_a/\mu_2^2, & c &= K_2\eta_c/\mu_2^2, \\ \eta_a &= \mu_4/2, & \eta_c &= (-\mu_2 + \mu_4 + \mu_5)/2, \\ \eta_{\text{twist}} &= \gamma_1, & \eta_{\text{bend}} &= \gamma_1 - \mu_2^2/\eta_c, \end{aligned}$$

and

$$\gamma_1 = -\mu_2 + \mu_3.$$

Since

$$\frac{1}{c} = \frac{\eta_{\text{twist}}}{K_2} - \frac{\eta_{\text{bend}}}{K_3} \frac{K_3}{K_2},$$

equation (1) contains two independent unknown parameters,  $K_3/K_2$  and  $a = \eta_a K_2/\mu_2^2$ . Hence two parameter fitting to equation (1) of the observed scattering angular

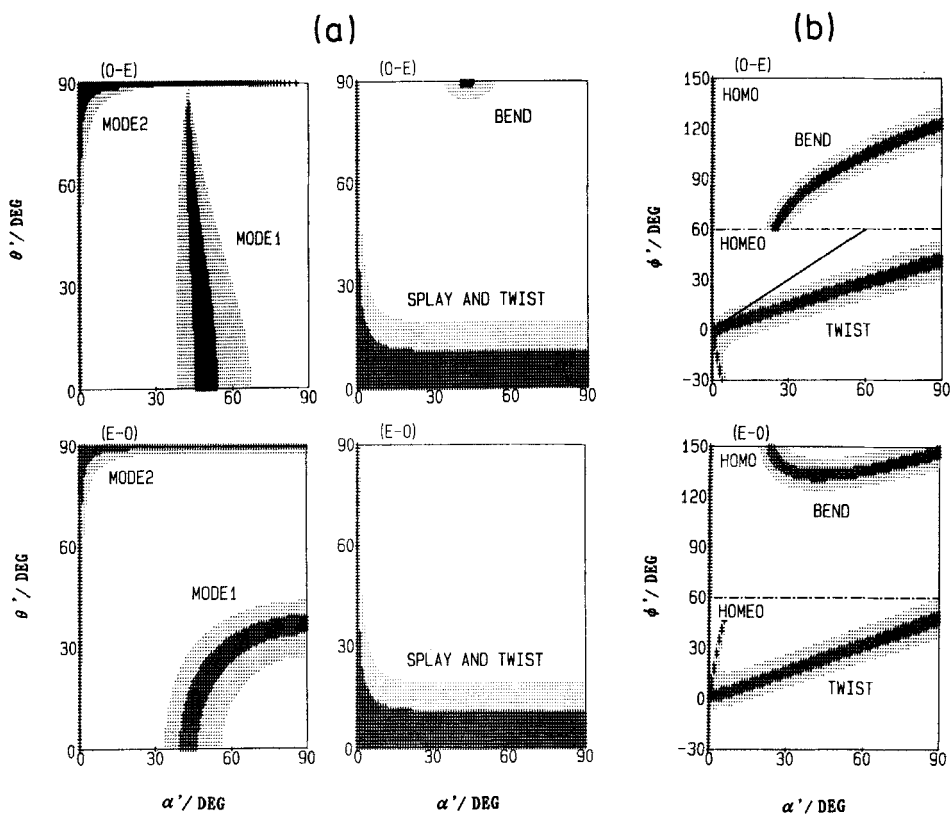


Figure 2. Maps drawn in (a) the  $(\theta', \phi' = 90^\circ, \alpha')$  space and (b) the  $(\theta' = 90^\circ, \phi', \alpha')$  space where at least one of  $(G_1/G_2)^2$ ,  $(G_2/G_1)^2$ ,  $(q_{\parallel}/q_{\perp})^2$  and  $(q_{\perp}/q_{\parallel})^2$  becomes negligibly small; darkly and lightly hatched regions indicate less than  $10^{-3}$  and  $10^{-2}$ , respectively. Here,  $\theta'$  and  $\phi'$  are polar and azimuthal angles defining the director orientation.  $\alpha'$  is the scattering angle. The primes indicate the angles observed outside the medium.  $G_1$  and  $G_2$  are geometrical parameters defined in equation (17) of [27]. The reduced temperature  $(T_{NI} - T)$  is  $5^\circ\text{C}$ .

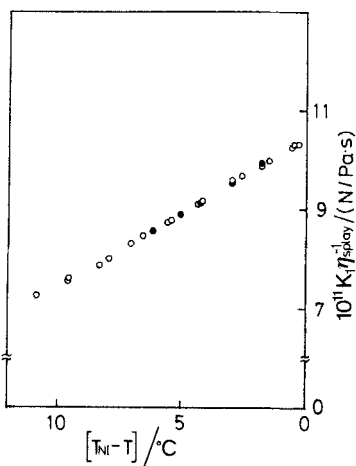


Figure 3. The temperature dependence of  $K_1/\eta_{\text{splay}}$  determined by using (O-E) indicated by  $\circ$  and (E-O) denoted by  $\bullet$  depolarizations in the space  $(\theta' = 0, \phi' = 0, \alpha')$ .

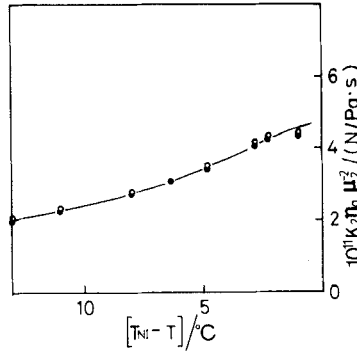


Figure 4. The temperature dependence of  $K_2\eta_a/\mu_2^2$ ; ● and ○ indicate (E-O) and (O-E) depolarizations, respectively.

dependence of  $\Gamma_2$ , allows us to determine  $K_3/K_2$  and  $a = K_2\eta_a/\mu_2^2$  as illustrated in figure 4.

(4) The mode 1 spectral width is given by

$$\Gamma_1 = \frac{(K_3/K_2)q_{||}^2 + (K_1/K_2)q_{\perp}^2}{(\gamma_1/K_2) - \frac{\{(\mu_3/\mu_2)q_{\perp}^2 - q_{||}^2\}^2}{bq_{\perp}^4 + dq_{\perp}^2q_{||}^2 + cq_{||}^4}}, \tag{2}$$

where

$$b = K_2\eta_b/\mu_2^2, \quad d = K_2\eta_a/\mu_2^2$$

$$\eta_b = (\mu_3 + \mu_4 + \mu_6)/2, \quad \eta_a = (\mu_1 + \mu_3 + \mu_4 + \mu_5)/2,$$

and

$$\eta_{\text{splay}} = \gamma_1 - \mu_3^2/\eta_b.$$

Since

$$\frac{1}{b} = \left(\frac{\mu_2}{\mu_3}\right)^2 \left(\frac{\eta_{\text{twist}}}{K_2} - \frac{\eta_{\text{splay}}}{K_1} \frac{K_1}{K_2}\right),$$

equation (2) contains two independent unknown parameters,  $\mu_2/\mu_3$  and  $d = K_2\eta_a/\mu_2^2$ . Hence two parameter fitting to equation (2) of the measured polar and scattering angle dependence, in principle, allows us to determine  $\mu_3/\mu_2$  and  $d = K_2\eta_a/\mu_2^2$ . Note that the depolarized scattering geometries can be used as shown in figure 2(a) [27].

(5) The twist elastic constant  $K_2$  was determined by observing the scattered light intensity or spectral width as a function of an applied electric field [26].

### 3. Results and discussion

Since the experimental setup we used was not suitable to measure the polar dependence of the spectral width, we could not perform procedure (4). Instead, we assumed Parodi's relation

$$\mu_2 + \mu_3 = -\mu_5 + \mu_6.$$

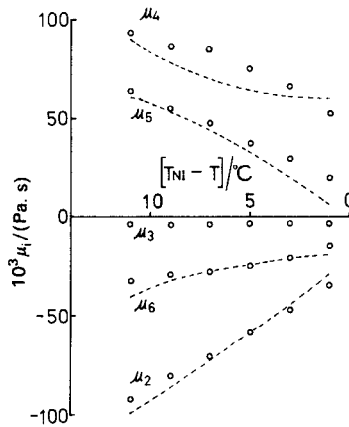


Figure 5. The temperature dependence of the Leslie viscosity coefficients  $\mu_i$  ( $i = 2-6$ ) determined in this work indicated by  $\circ$ , compared with the data (except  $\mu_3$ ) from shear flow experiments by Chmielewski [28] represented by the broken line.

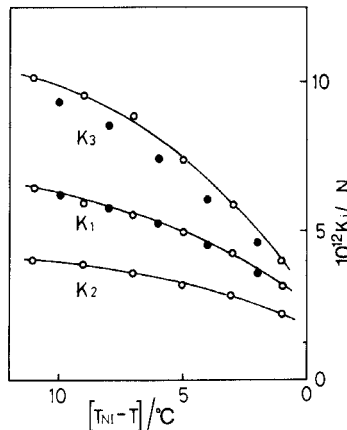


Figure 6. The temperature dependence of the Frank elastic constants  $K_i$  ( $i = 1-3$ ) where a rather large disagreement exists between the data of  $K_3$  determined in this work indicated by  $\circ$  and in the intensity measurement of scattering angle dependence represented by  $\bullet$  [27].

Thus, all of the Leslie viscosity coefficients except  $\mu_1$ , together with the Frank elastic constants were determined as shown in figures 5 and 6. The viscosity coefficients  $\mu_2$ ,  $\mu_4$ ,  $\mu_5$  and  $\mu_6$  so determined are in good agreement with those previously obtained by Chmielewski [28] using a shear flow method. Figure 7 shows the ratio  $\mu_3/\mu_2$  so determined together with that previously obtained by Skarp *et al.* using a shear flow method [29]; the agreement is rather good. The elastic constant  $K_3/K_2$  is systematically higher than that obtained from the intensity measurement of the scattering angle dependence [27], resulting in a higher  $K_3$  as shown in figure 6, although the agreement is almost equally close for  $K_1/K_2$ . The reason may be that the intensity measurement of the scattering angle dependence has a low reliability. Actually, figure 1 in [27] shows a larger data scatter and a larger systematic error for  $K_3/K_2$  than for  $K_1/K_2$ . Detailed measurements and full discussions will be reported shortly.

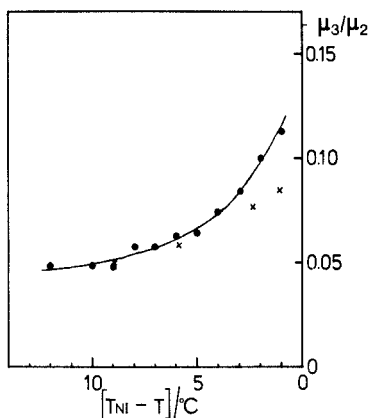


Figure 7. The temperature dependence of  $\mu_3/\mu_2$  determined in this work indicated by ●, compared with the data from shear flow experiments by Skarp *et al.* [29] represented by x.

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