

## Ultrasonic Light diffraction in liquid crystal in isotropic phase

液晶等方相での超音波光回折

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### 1. Introduction

Recently we have investigated polarization of diffracted light by ultrasound in triphenylphosphite.<sup>1</sup> Only phase retardation of the  $\pm 1$ st order diffracted lights was observed and it was proportional to the ultrasonic frequency. This was because the birefringence was much smaller than the isotropic modulation of the refractive index and the reorientational relaxation frequency was much larger than that of ultrasound in TPP. The obtained values were good agreement with theoretical estimation from the modified Klein and Fltts formula.

In this study, we used the isotropic phase of liquid crystal, 4-cyano-4'-pentylbiphenyl (5CB). The sample is appropriate for examination of the proposed method, because the reorientational relaxation frequencies can be changed by changing the temperature. In addition, the flow birefringence<sup>2</sup> and the reorientational relaxation time<sup>3</sup> for 5CB have already been reported; these are required for the theoretical estimation. Frequency dependences of the complex ratio of birefringence to isotropic refractive index variation were obtained. We compared them with those estimated by the theory.

### 2. Theory

Consider a light wave with wavenumber  $k_l$  and incident at an angle  $\theta$  on plane longitudinal ultrasound propagating along the  $x$  axis. The refractive index gratings produced by the ultrasound for the lights can be written as

$$n_c(x, t) = n_0 + n_c \operatorname{Re}[\exp[i(\omega t - kx + \phi_c)]] \quad (1)$$

Here  $c$  denotes  $x$  or  $y$ . The complex normalized amplitude of the  $j$ -th order diffracted light  $\Phi_{j,c}$  appears as the coefficients of the complex Fourier series of the diffracted light waves and it can be calculated from the general Raman-Nath theory.

$$\frac{d\Phi_{j,c}}{dz} + \frac{v_c}{2L}(\Phi_{j-1,c} - \Phi_{j+1,c}) = ij(j-2\alpha)\frac{Q}{2L}\Phi_{j,c} \quad (2)$$

Here  $c$  denotes  $x$  or  $y$ . Raman-Nath parameter  $v_c = kn_c L$ , Klein and Cook parameter  $Q = k^2 L / n_0 k_{\text{light}}$ , and  $\alpha = -(n_0 k_{\text{light}} / k) \sin \theta$ . At the normal incidence,

$\alpha = 0$  and the conditions that  $\alpha = 0.5, -0.5$  correspond to those for the Bragg incidence for the 1st and -1st order diffracted lights, respectively. The boundary condition is  $\Phi_{j,c} = \delta_{j,0}$ , where  $\delta_{j,0}$  is Kronecker delta. From the solution of eq.(2), we can calculate the intensity of diffracted light. From the complex ratio of electric field strength:

$$\frac{E_{j,y}}{E_{j,x}} = e^{ij(\phi_y - \phi_x)} \frac{\Phi_{j,y}(v_y)}{\Phi_{j,x}(v_x)} \quad (3)$$

the relative phase shifts of the  $x$ -polarization component from the  $y$ -polarization one  $\delta_j$  and the change of the azimuth angle,  $\Delta\chi_j$  are calculated from eq.(3). The refractive index change caused by ultrasound can be divided into the isotropic change in refractive index  $n_{\text{iso}}$  and birefringence  $\Delta n$ . From symmetry requirement of the anisotropy of the longitudinal wave, the change in refractive index grating in complex number due to the ultrasound can be written respectively as,  $n_x e^{i\phi_x} = n_{\text{iso}} - (2/3)\Delta n$ ,  $n_y e^{i\phi_y} = n_{\text{iso}} + (1/3)\Delta n$ .

Given the normalized density variation due to the longitudinal ultrasound,  $\delta\rho/\rho$ , an isotropic refractive index change  $n_{\text{iso}}$  and a birefringence  $\Delta n$  are obtained in liquids of anisotropic molecules as,

$$n_{\text{iso}} = \rho(\partial n_0 / \partial \rho)(\delta\rho/\rho), \quad (4)$$

$$\Delta n = \frac{\Delta n_f}{\gamma} \Gamma \frac{i\omega}{i\omega + \Gamma} \frac{\delta\rho}{\rho}, \quad (5)$$

where  $\Delta n_f / \gamma$  and  $\Gamma$  are the flow birefringence per shear rate and the reorientational relaxation rate, respectively. The  $\rho(\partial n_0 / \partial \rho)$  value is estimated from the refractive index using the empirical equation. The  $n_{\text{iso}}$ , and  $\Delta n$  per  $\delta\rho/\rho$  can be calculated using refractive index values measured by refractometer and the  $\Delta n_f / \gamma$  and  $\Gamma$  values in the literature, respectively. We will discuss consistency between experimental data and theory.

For an initially linearly polarized light with the polarization plane at  $\pi/4$  with respect to the  $x$  axis,  $\delta_j$  and  $\Delta\chi_j$  are obtained as

$$\delta_j = -\arg(E_{j,y} / E_{j,x}) \quad (6)$$

$$\Delta\chi_j = \frac{1}{2} \tan^{-1} \frac{2R_j \cos \delta_j}{R_j^2 - 1} + \gamma \frac{\pi}{2} - \frac{\pi}{4} \quad (7)$$

where  $R_j = |E_j(v_y) / E_j(v_x)|$  and  $\gamma = 1$  if  $R_j < 1$  and  $\gamma = 0$  otherwise. In the analysis of experimental data, we adjust the parameter to fit the dependence of the

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$\Delta\chi_j$  and  $\delta_j$  values on  $v_x$ . From the relationship of  $v_x = kn_x L$ ,  $\Delta n / n_{\text{iso}}$  will be a fitting parameter. Note that  $\Delta n / n_{\text{iso}}$  is represented in complex number. Once the  $\Delta n / n_{\text{iso}}$  value is obtained from the analysis, we can obtain birefringence per strain using the  $\rho(\partial n_0 / \partial \rho)$  value estimated from the refractive index measurement.

### 3. Results and discussions

Figure 1 shows Ramam-Nath parameter dependence normalized intensity  $I_{jc}$  of diffracted light by ultrasound in 5CB at 25 MHz, 60°C. Index  $j$  indicates diffraction order and the index  $c$ , which is  $x$  or  $y$ , indicates the polarization of incident light at Bragg angle condition. The amplitude used here is the Raman-Nath parameter, for  $x$ -polarization light,  $v_x$ . The difference in the intensity of diffracted light between the  $x$ - and  $y$ - polarization components come from the difference in the amplitude of refractive index grating induced by the ultrasound even if the ultrasonic amplitudes are the same. Substituting the  $\Delta n_f / \gamma$  value<sup>2</sup> and  $\Gamma$  value<sup>3</sup> in the literature into the eq. (5) gives birefringence,  $\Delta n$ . The  $n_{\text{iso}}$  is estimated using the refractive index. Once  $v_x$  is given, we can calculate the values of  $v_y$  at the same ultrasound amplitude and thus calculate intensity of  $y$ - polarization components. Agreements of diffraction light intensity between experiment and calculation are good.

Figure 2 shows Ramam-Nath parameter dependence of  $\delta_{+1}$  and  $\Delta\chi_{+1}$  for an initially linearly polarized light with the polarization plane at  $\pi/4$  with respect to the  $x$  axis in 5CB at 25 MHz, 60°C. Curves in Fig. 2 are the calculated  $\delta_{+1}$  and  $\Delta\chi_{+1}$  values by adjusting the  $\Delta n / n_{\text{iso}}$  value to fit to those obtained experimentally. The relation of  $\Delta n / n_{\text{iso}} = 0.103 + 0.089i$  is thus obtained.

Figure 3 shows the frequency versus the  $\Delta n / n_{\text{iso}}$

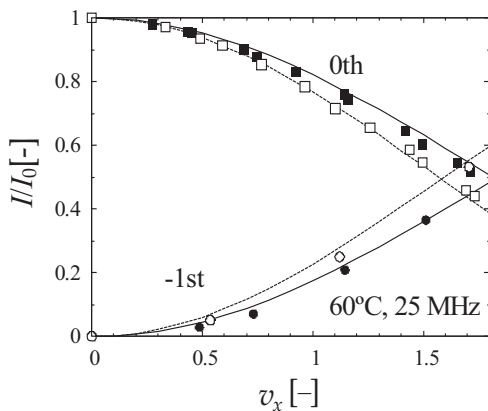


Fig.1 Ramam-Nath parameter dependence normalized intensity  $I_{jc}$  of diffracted light by ultrasound in 5CB. Index  $j$  indicates diffraction order and index  $c$ , which is  $x$  or  $y$ , indicates the polarization of the incident light.

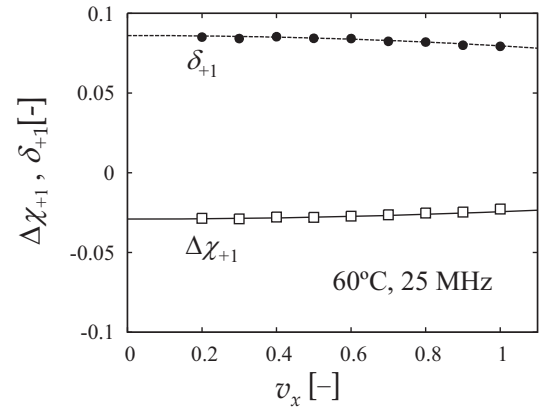


Fig.2 Ramam-Nath parameter dependence of change of the azimuthal angles and phase retardations at the Bragg angle condition in 5CB.

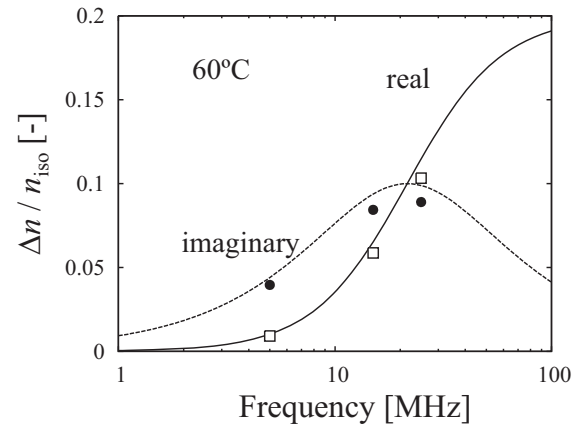


Fig. 3 Frequency dependence of the complex ratio of birefringence to isotropic refractive index variation  $\Delta n / n_{\text{iso}}$  in 5CB

values in 5CB at 60°C. Curves in Fig.3 are the calculated  $\Delta n / n_{\text{iso}}$  using the procedure described in the section of the theory. Required the  $\Delta n_f / \gamma$  value<sup>3</sup> and  $\Gamma$  value<sup>4</sup> are used in the literature and the refractive index are used measured by the refractometer. Points indicated by symbols are obtained from the  $\delta_{+1}$  and  $\Delta\chi_{+1}$  data using the method describe in the previous paragraph. The agreement of the points and the calculated curves are good. The method indicated the previous paragraph is effective to evaluate the frequency dependence of the birefringence induced the ultrasound.

### References

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