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## Estimation of Surface and Flexoelectric Polarization in Smectic Liquid Crystal using a Pyroelectric Technique

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Nematic and smectic A liquid crystals are uniaxial media with a preferable direction of molecular axes  $\mathbf{n}$  called the director. The director has no polar axis and its states  $\mathbf{n}$  and  $-\mathbf{n}$  are indistinguishable. Consequently, the nematic and smectic A phases have no spontaneous polarization. At the interface with a glass or at the free surface exposed to the air, the mirror symmetry is broken and the surface polarization  $\mathbf{m}_{surf}$  arises[1]. It is one of the order-electric polarizations[2] which are derived from the spatial dependence of order parameter  $S(z)$  in a thin bulk layer close to the interface,

$$\mathbf{P}_s = r_1(\mathbf{n} \cdot \text{grad}S)\mathbf{n} + r_2\text{grad}S. \quad (1)$$

In the quadrupolar approximation, with the electric polarization given by the gradient of the quadrupolar density[3]  $\mathbf{P} = q\nabla Q$  the order-electric polarization takes the form:

$$\mathbf{P}_s = \frac{3}{2}e_0(\nabla S) \left( \mathbf{n} \cdot \mathbf{n} - \frac{I}{3} \right). \quad (2)$$

A macroscopic polarization may also be induced in the bulk of a nematic and smectic A liquid crystals by a bend or splay distortion of the director field. A commonly used term for that polarization is flexoelectric one[4] and its general form satisfying symmetry requirements

$$\mathbf{P}_f = e_1\mathbf{n}(\nabla \cdot \mathbf{n}) - e_3(\mathbf{n} \times (\nabla \times \mathbf{n})) \quad (3)$$

consists of two terms with correspondent flexoelectric coefficients  $e_1$  and  $e_3$  related to the splay and bend distortions. From the microscopic point of view, dense packing of dipolar banana- or pear shape molecules in a bent or splayed structure inevitably creates a dipole moment in a unit space.

Despite the fact that the concept of the surface and flexoelectric polarizations is discussed for many years, the quantitative data are very scarce. Only rough estimations of  $\mathbf{m}_{surf}$  at room temperature have been done from observations of surface instabilities. The situation with the sign of  $\mathbf{m}_{surf}$  is also controversial. The temperature dependence of the surface polarization has never been measured. Therefore it is difficult to estimate the contribution of the

nematic order into the magnitude of the polarization. In our previous work, the method of polarization estimation based on a pyroelectric effect has been established and the temperature dependence of the "nematic part" of the surface and flexoelectric polarizations have been measured for conventional nematic liquid crystal 5CB[5].

In this study, for the first time, the temperature dependence of the "nematic and smectic A phases" of both the surface and flexoelectric polarizations have been measured. Pure and bisazo-dye doped 8OCBs were used in cells with controlled asymmetry for light absorption. The flexoelectric polarization has been measured in a hybrid aligned cell, and the surface polarization in a planar and homeotropic aligned cells by a pyroelectric technique using short pulses of a YAG laser at  $\lambda = 532 \text{ nm}$  to create a temperature increment  $\Delta T$ , see Fig. 1. In turn, the temperature increment  $\Delta T$  was measured independently by a novel time-resolved "optical thermometer" technique monitoring temperature dependent birefringence by a He-Ne laser beam. The polarization is derived from  $\Delta T$  and pyroelectric response  $V_p$  using the following

$$P(T) = \int_{T_i}^T \gamma(T) dT, \quad V_p = -\frac{A\Delta T\gamma}{C} \exp\left(-\frac{t}{RC}\right) \quad (4)$$

here,  $C$  is sum of the input and cell capacitances and  $A$  is an area of electrode.

The flexoelectric polarization estimated from the pyroelectric response of hybrid cell is shown in Fig. 2. The sum of flexoelectric coefficients ( $e_1 + e_3$ ) is positive in both the nematic and smectic A phases. In the nematic phase, the discrepancy exists between the two curves for the flexoelectric coefficients. This discrepancy is explained by the nonuniformity of the director curvature along the  $z$ -axis due to the difference in the elastic moduli  $K_{11} \neq K_{33}$ . On transition to the smectic A phase, the discrepancy between the two curves increases markedly. In the smectic A phase focal-conic texture has been observed. This indicates the distribution of the flexoelectric polarization along  $z$ -

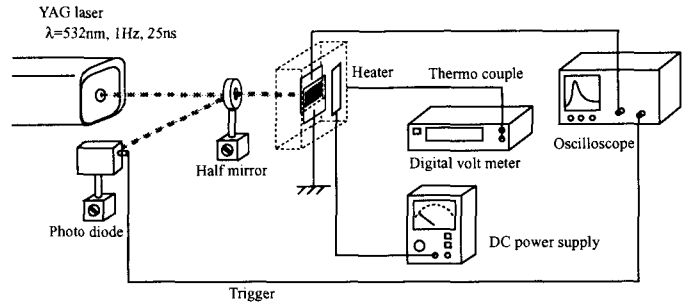


Fig. 1. Experimental setup

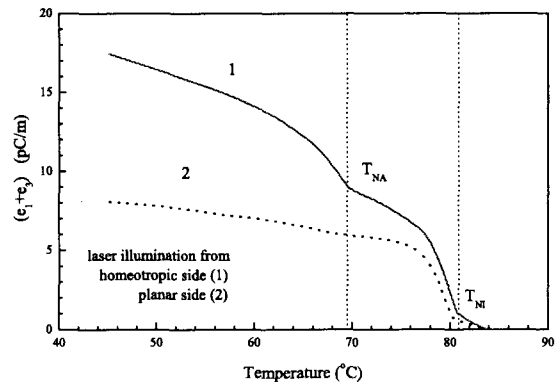


Fig. 2. Temperature dependence of the sum of flexoelectric coefficients. The hybrid cell was illuminated from either the planar interface or the homeotropic interface.

axis(perpendicular to the interface) becomes strongly inhomogeneous. In a hybrid cell, the flexoelectric polarization is directed from the planar interface to the homeotropic one. The shape of the relevant "molecular" quadrupole immediately follows from the sign of the flexoelectric polarization, as shown in Fig. 3. Figure 3 shows the shape of molecular quadrupoles consistent with the direction of the flexoelectric polarization in the hybrid cell filled with 8OCB ( $\epsilon_0 > 0$ ).

Both in the homeotropic and planar cell, the surface polarizations are directed from the substrate into the liquid crystal. From eq. (2), the following equations for surface polarization in the planar and homeotropic orientations are obtained

$$m_p = -\frac{1}{2}e_0\Delta S_p \quad \text{and} \quad m_h = e_0\Delta S_h, \quad (5)$$

here  $\Delta S_{p,h} = S_{p,h} - S$  is the difference between the surface and bulk order parameters. In the case of 8OCB both  $m_h$  and  $m_p$  are positive, as shown in Fig. 4, therefore,  $\Delta S$  should have opposite signs for the two interfaces.

At the planar interface of 8OCB in the case of polyimide, the magnitude of the orientational order parameter at the surface is higher than that in the bulk. At the homeotropic interface of 8OCB, due to the surfactant used, the magnitude of the orientational order parameter at the surface is lower than the bulk value. On transition to the smectic A phase, in both cases the surface order increases strongly than that of the bulk.

In conclusion, the temperature dependence of the electric polarization has been measured for 8OCB in homeotropic, planar and hybrid aligned cells in nematic and smectic A phases. The polarization was determined using the pyroelectric response of the cells to a short laser pulse; the light at  $\lambda = 532$  nm was absorbed by a dye. As a result the temperature dependence of both surface and flexoelectric polarizations was determined separately in the nematic and smectic A phases. The flexoelectric coefficient was shown to be positive and polarization was directed from planar interface to

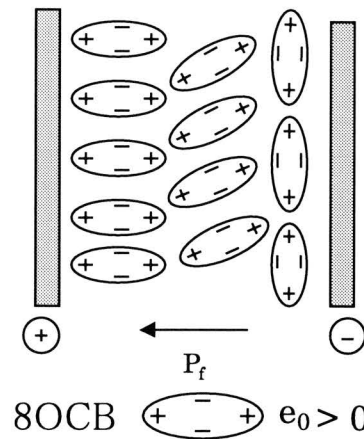


Fig. 3. Temperature dependence of the surface polarization  $m_h$  and  $m_p$  for homeotropic and planar interfaces.

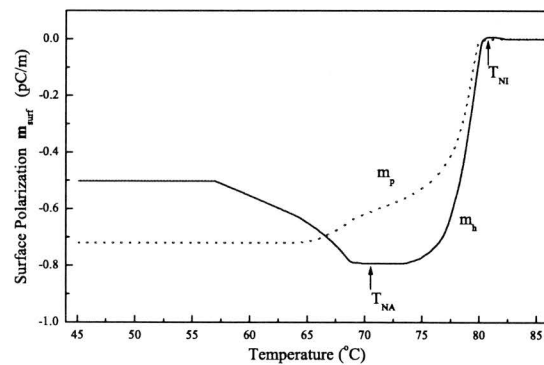


Fig. 4. Temperature dependence of the surface polarization  $m_h$  and  $m_p$  for homeotropic and planar interfaces.

homeotropic one, which is opposite to the case of 5CB[5]. On the molecular scale, the difference is accounted for by the difference of electric charge distribution in molecular quadrupoles formed in each liquid crystal. The surface polarization was directed from substrate into the liquid crystal in both homeotropic and planar orientations.

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