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THE CHIRALITY EFFECT ON THE OPTICAL NONLINEARITY OF SMECTIC C* LIQUID CRYSTALS

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Abstract We have studied the chirality effect on the optical nonlinearity of ferroelectric liquid crystals (FLCs) by means of the second-harmonic generation (SHG). A chiral agent of CB15 is added to a commercial FLC of CS1017 for controlling the optical activity in terms of chirality. From the electric-field dependence of the SHG profiles, it is found that the chirality contributes to both the second-order nonlinearity and third-order one which couples with a static electric field.

INTRODUCTION

Ferroelectric liquid crystals (FLCs) have been extensively studied from the fundamental as well as practical viewpoints. Based on an elegant physical argument, it was first shown by Meyer et al.[1] that the smectic C phase (Sm C*) of the chiral molecules possesses the ferroelectricity. Recently, FLCs have been explored for use in optical second-harmonic generation (SHG) devices. Since the first observation of SHG in FLC cells,[2] considerable progress has been made; for example, angle phase matching,[3] microcavity action,[4] and molecular design for FLC molecules with high optical nonlinearity.[5]

Until now, two approaches to the improvement of the nonlinear optical (NLO) properties of FLCs have been most commonly taken. One of them is to combine the NLO functionality with ferroelectricity and liquid crystallinity. The other is to introduce high NLO activity into FLCs by doping with proper NLO molecules. However, a description of the chirality effect on the optical nonlinearity of FLCs has not fully made so far. In general, the chirality is expected to change the structural symmetry of FLCs, which leads to the change in the helical pitch, the spontaneous polarization, and the SHG process. In the present work, we have studied the chirality effect on the NLO properties of FLCs, doped with the chiral agent, by performing both the SHG and spontaneous polarization measurements. It is found that the second-order NLO coefficient exhibits exactly the same behavior as the polarization and the optical tilt in the Sm C* state while the third-order one saturates with increasing the dopant concentration.

EXPERIMENTAL

The LC used in this study was a commercially available FLC mixture, CS1017, from Chisso Petrochemical Co. It has the Sm C* phase between −20°C and 55°C. The molecular tilt Θ and the spontaneous polarization P_s of CS1017 is 26° and $-9.3\mathrm{nC/cm^2}$ at 20°C, respectively. It has a left-handed helical pitch. For studying the chirality effect, the FLC was doped with a chiral agent of CB15, obtained from E. Merck, whose pitch is right-handed.

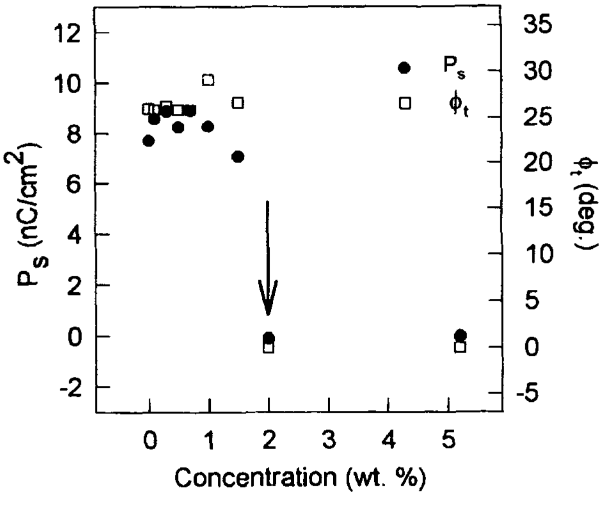

Figure 1: The spontaneous polarization and the optical tilt as a function of the dopant concentration. The arrow marks the concentration for discontinuity in P_s.

For each doped FLC cell aligned homogeneously, the SHG measurements were made at an oblique incidence[6] as a function of the azimuthal angle for rotation. The nominal thickness of the cell was 10μm. A Q-switched Nd:YAG laser (repetition rate: 10 Hz, wavelength: 1064 nm, pulse duration: 10 ns, and output power: 10 mJ/pulse) was used as the fundamental light source after passing through a visible cut filter, a polarizer, and a half-wave plate. The transmitted SHG intensity of 532 nm generated from the FLC was detected with a photomultiplier after passing through an analyzer. The detected signal was fed into a boxcar integrator. The triangular wave method was used for measuring the spontaneous polarization.[7] All the measurements were carried out at room temperature.

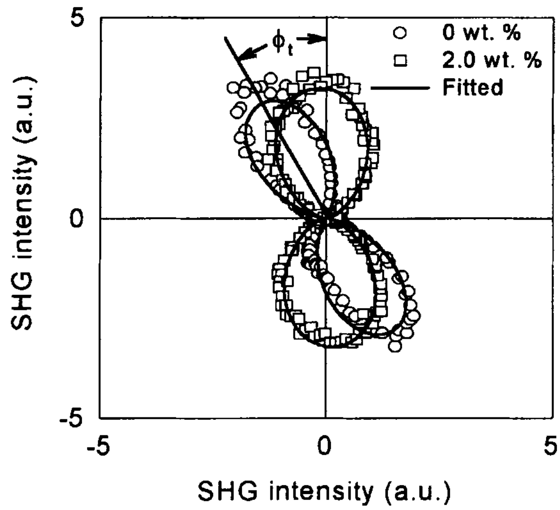

Figure 2: The azimuthal angle dependence of the SHG intensity. The solid lines are best theoretical fits using the expression for the SHG profiles in Ref. 8.

RESULTS AND DISCUSSION

Experimental results for the spontaneous polarization P_s and the optical tilt ϕ_t with respect to the rubbing direction are shown in Fig. 1. Note that the optical tilt ϕ_t, determined from the SHG profiles, does not mean the molecular tilt Θ which vanishes at the Sm A - Sm C* transition. As shown in Fig. 1, it is clear that both P_s and ϕ_t exhibit the same behavior of discontinuity at about 2.0 wt.% of the chiral dopant; both P_s and ϕ_t abruptly drop to zero at that concentration. This indicates that the nature of the chiral agent, different from the enantiomer, dictates the ferroelectricity in doped FLCs. Below the concentration for discontinuity, $P_s = -8.25\text{nC/cm}^2$ and $\phi_t = 26.0°$, which is consistent with the literature values.

Now, let us examine the azimuthal angle dependence of the SHG intensity in the presence of a dc electric field E. The unwound state of FLC has C_2 symmetry along the direction of the spontaneous polarization. When the Kleinman symmetry is properly taken and the biaxiality of FLC is ignored, the SHG intensity from the unwound state can be written in terms of the input and output polarizations[8]. Fig. 2 shows the SHG intensities from two FLC cells, one of which is a bare CS1017 and the other is a 2.0 wt.% doped CS1017, at $E = 10\mathrm{V}/\mu\mathrm{m}$. As shown in Fig. 2, no optical tilt ($\phi_t \approx 0$) with respect to the rubbing direction was observed for the doped CS1017. The variations of ϕ_t with the dopant concentration have been already shown in Fig. 1. Note that P_s vanishes above a certain concentration (2.0 wt.%) as ϕ_t does. FLCs will not be optically active when properly doped with a chiral agent of opposite handedness. This is exactly what was seen from our data. In fact, the SHG would come from the chiral nature of the FLC as well as the intrinsic optical activity of the dopant itself.

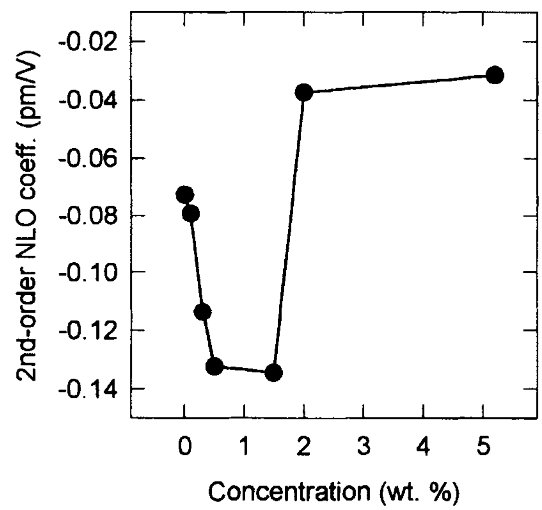

Figure 3: The second-order NLO coefficient $\chi^{(2)}_{222}$ as a function of the dopant concentration.

The optical tilt observed in the SHG profiles can be understood as follows. In the Sm C* phase, a linear coupling of P_s with a dc electric field E makes the molecules tilted away from the rubbing direction (the reference direction). Above a certain concentration of the dopant, P_s becomes to zero and no direct coupling between P_s and E is expected for the nonchiral version. In this case, the dielectric anisotropy is solely responsible for the reorientation of the molecules. Consequently, the average optic axis of the system will coincide with the rubbing direction since the tilt direction is independent of the polarity of the field. This makes the SHG profiles not tilted away from the rubbing axis, meaning that $\phi_t = 0$.

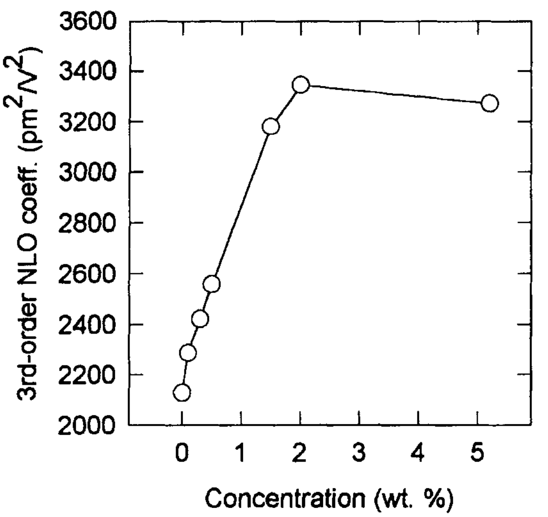

Figure 4: The third-order NLO coefficient $\chi^{(3)}_{2222}$ as a function of the dopant concentration.

From the linear dependence of the measured SHG profiles on the electric field, the values of $\chi^{(2)}_{222}$ and $\chi^{(3)}_{2222}$ were determined. Here, the subscript 2 denotes the direction of the spontaneous polarization. Let us describe these two coefficients in view of the variations of P_s with the dopant concentration in Fig. 1. Figs. 3 and 4 show $\chi^{(2)}_{222}$ and $\chi^{(3)}_{2222}$ as a function of the dopant concentration, respectively. Clearly, these NLO coefficients exhibit two distinct regions separated at about 2.0 wt.%. It is noted that this concentration is exactly the same as the one for discontinuity observed in P_s. At low concentrations ($\leq$ 2.0 wt.%), both $\chi^{(2)}_{222}$ and $\chi^{(3)}_{2222}$ increase monotonically with increasing the dopant concentration. At high concentrations ($\geq$ 2.0 wt.%), however, $\chi^{(2)}_{222}$ approaches zero while $\chi^{(3)}_{2222}$ saturates. This behavior can be described in terms of the optical nonlinearity inherent to the chiral dopant, CB15, and the structural symmetry of the FLC.

Suppose that CB15 possesses the optical nonlinearity in itself. The NLO response of CS1017, doped with CB15, to a dc field will then be effectively as $\chi^{(2)}_{eff} = \chi^{(2)}_{ijk} + \chi^{(3)}_{ijkl}E_l(0)$. In this case, both the second- and third-order NLO coefficients along the polarization direction (denoted by 2) increase with increasing the concentration of CB15. It is contrasted to a mixture of enantiomers. At about 2.0 wt.% of CB15, $\chi^{(2)}_{222}$ drops sharply since it reflects the non-centrosymmetric nature of the system as P_s does. However, $\chi^{(3)}_{2222}$ continuously increases and remains constant in a nonchiral version of the doped CS1017.

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