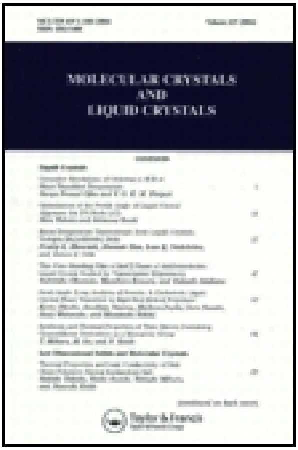

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ORIGIN OF THE ENHANCED OPTICAL NONLINEARITY OF CHROMOPHORES IN A CHIRAL SMECTIC LIQUID CRYSTAL ENVIRONMENT

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Abstract The enhancement mechanism for the second harmonic generation (SHG) of chromophores in chiral smectic liquid crystal environment has been studied. The liquid crystals (LCs) studied are SCE13 and its racemic version, SCE13R (British Drug House), the former of which has the chiral and the latter nonchiral smectic C phases at room temperature, respectively. The LCs are doped with 4-dimethylamino-4'-nitro stilbenes (DANS) molecules in weight. It is found that the SHG intensity is enhanced by the doping of the DANS molecules.

INTRODUCTION

In the last few years, much progress has been made in the development of nonlinear optical (NLO) organic materials for fast and efficient electro-optic modulators.[1] Because of the intrinsic noncentrosymmetry, ferroelectric liquid crystals (FLCs) are one of the promising classes of novel NLO materials. Based on an elegant physical argument, it was first shown by Meyer et al.[2] 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.

Two approaches to the improvement of the 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. In the present work, we take the second approach to the enhancement of the optical nonlinearity in FLC composites. Moreover, the physical origin of the enhanced nonlinearity of the chromophores in a chiral LC medium is studied by the SHG measurements. The second-order NLO coefficients of both Sm C and Sm C* LC composites exhibit a linear dependence on the concentration of doped NLO molecules.

EXPERIMENTAL

The liquid crystals used in this study were commercially available FLC mixture, SCE13, and its racemic version, SCE13R, obtained from British Drug House. The chiral and racemic mixtures exhibit the Sm C* and Sm C phases at room temperature, respectively. The materials were doped with 4-dimethylamino-4'-nitro stilbenes (DANS) molecules. The sample cells were made up of conductive indium-tin-oxide glass substrates. The cell gap was maintained with glass spacers of 10 μm thick. The inner surfaces of the substrates were coated with polyimides and rubbed, so that the planar, homogeneous alignment of the molecules was promoted.

The SHG measurements were made at an oblique incidence as a function of the azimuthal angle for rotation ϕ.[3] All measurements were carried out at room temperature. The value of $d_{11} = 0.49$ pm/V for a quartz plate[4] was used as a reference for evaluating the absolute magnitudes of resultant NLO coefficients.

RESULTS AND DISCUSSION

For chiral Sm C* materials, the molecular director makes an angle, ϕ_t, tilted with respect to the layer normal and rotates continuously on a cone, moving from one layer to the next, to form a helical structure. In fact, this helical structure is not optically active to the SHG process. In the presence of an applied electric field, it becomes unwound and the spontaneous polarization aligns along the field direction. This unwound state has C_2 symmetry with respect to the polarization direction (say axis 2), and the second-order NLO process will be then involved. For nonchiral Sm C materials, however, no SHG process is expected since no spontaneous polarization exists. In the presence of an electric field, only the third-order process due to a coupling with the field would occur.

Let us first describe the ϕ dependence of the SHG intensity in the presence of a dc electric field. The SHG intensities from SCE13, SCE13/DANS, SCE13R, and SCE13R/DANS were measured as a function of the azimuthal angle for rotation ϕ at $E = 10$ V/μm. The measured SHG profiles for SCE13, SCE13/DANS(1% in weight), SCE13R, and SCE13R/DANS(1% in weight) were shown in Figures 1(a), (b), (c), and (d), respectively. The solid lines are the least-square fits to theoretical expressions for the SHG intensities which have been reported previously.[5] Clearly, the SHG intensity was enhanced for doped LCs with the DANS molecules as shown in Figures 1 (b) and(d). Note that an optical tilt exists for SCE13 but no tilt for SCE13R because of the intrinsic molecular chirality.[6]

From the theoretical fits, the second-order NLO coefficients, d_{ij}'s, were determined self-consistently. The values of d_{ij}'s at $E = 10$ V/μm were collected in Table 1. As expected, for undoped Sm C* material, d_{22} makes a dominant contribution to the SHG process.[3] The direction of d_{22} lies along the spontaneous polarization direction (axis 2). It is evident from Table 1 that for both SCE13 and SCE13R, the enhancement of d_{22} by doping is comparable to each other. One interesting point is that all the coefficients are linearly dependent on the doping concentration.

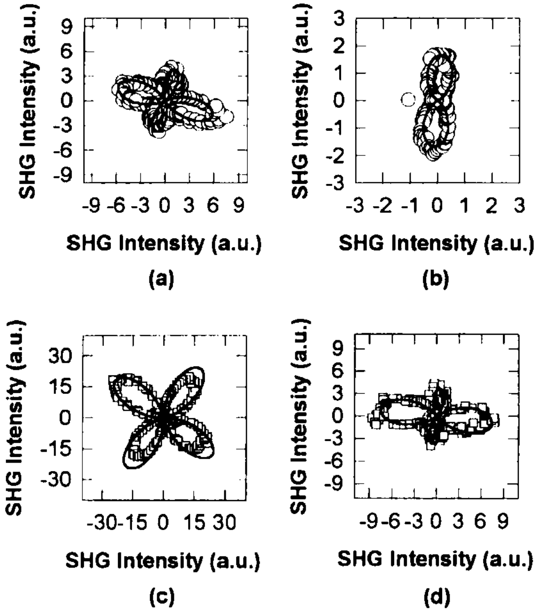

Figure 1: The SHG profiles of (a) SCE13, (b) SCE13R, (c) SCE13/DANS(1%), and (d) SCE13R/DANS(1%). The solid lines are the best theoretical fits. The measurements were made at $E = 10$ V/μm.

Based on the coupling argument,[5] the physical origin of the enhanced optical nonlinearity comes from a delicate interplay between the polar order of the doped NLO molecules and the ferroelectricity in the liquid crystal environment. For SCE13 composites, the coupling terms of the electric field with both the dipoles of the DANS molecules and the spontaneous polarization in the Sm C* phase will contribute to total interaction energy. For SCE13R, however, only the dipole interactions of the DANS molecules will be involved. Basically, the liquid crystalline potential acts as an orientational field for the DANS molecules along the molecular director, and a dc electric field gives a polar field for them in addition to the spontaneous polarization.

As a consequence, the delicate interplay between these two coupling terms results in the enhancement of the second-order NLO process in the doped FLC composites.

Table 1: The measured values of d_{ij}'s at $E = 10$ V/μm (in unit of pm/V)

Host	Guest	d_{22}	d_{23}	d_{16}	d_{14}
SCE13	- - -	0.34	0.00	0.00	-0.06
	DANS(1 wt%)	0.74	2.45	-3.21	1.35
	DANS(2 wt%)	1.35	7.11	-5.11	2.57
	DANS(5 wt%)	2.50	16.10	-11.50	6.50
SCE13R	- - -	0.27	0.00	0.00	0.00
	DANS(1 wt%)	0.84	0.06	0.31	0.00
	DANS(2 wt%)	1.41	0.11	0.65	0.00
	DANS(5 wt%)	2.55	0.14	1.00	0.00

CONCLUSION

We have studied the enhancement mechanism for the second-order NLO process of the DANS molecules in chiral liquid crystal environment. It was found that the molecular chirality provides a physical origin of the coupling between the polar order of the DANS and the ferroelectricity of the LC.

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