Phase-synchronous optical second-harmonic generation in a ferroelectric liquid crystal

M. I. Barnik, L. M. Blinov, and N. M. Shtykov

Research Institute for Organic Semifinished Products and Dyes
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"Temperature" phase synchronism is obtained in optical second-harmonic generation in a ferroelectric smectic C^* whose helicoid is untwisted by a constant electric field. The nonlinear second-harmonic susceptibility due to spontaneous polar ordering of the molecules is estimated.

The ferroelectric smectics C^* and H^* are the only liquid-crystal phases with a polar structure that should make possible in them second-harmonic generation (SHG) via the quadratic nonlinearity $\chi^{(2)}$. Up to now, only nonsynchronous SHG was reported in a ferroelectric liquid crystal (LC).[1,2] We present here the results of an investigation of phase-synchronous SHG in the ferroelectric phase C^* with untwisted helicoid. The investigated substance was *p*-decyloxybenzylidene-*p*-amino-2-methylbutylcinnamate (DOMAMBC).

In the absence of external forces in an LC, the director and the spontaneous-polarization vector of the smectic layer form in space a helicoidal structure similar to the cholesteric, the helicoid axis being perpendicular to the plane of the smectic layers. The spontaneous polarization vector of the smectic layer lies in the plane of the layer and is perpendicular to the molecule inclination plane. Application of a dc electric field perpendicular to the helicoid axis untwists the helicoid, but this raises the problem of separating the contribution made to the SHG by the spontaneous polarization from that of the polarization induced by the electric field. This is one of the problems solved in the present paper.

The usual method of obtaining phase-synchronous SHG in a nematic phase and in a smectic-*A* is to change the angle between the pump wave vector and the LC optical axis by rotating the cell around the direction of the dc electric field [3]. To obtain phase synchronism in this case we varied the temperature to change the inclination of the molecules in layers of the smectic C^* phase. A calculation of the expected synchronism direction θ_c from the refractive indices of DOBAMBC yields about 23.5° and 33.8° for interactions of the *ee–o* and *oe–o* type, respectively. Thus, in DOBAMBC, in which the angle between the molecule inclination and the normal to the smectic layer ranges from 0 to 29°,[4] it is possible to obtain temperature-controlled phase synchronism for *ee–o* interaction.

The SHG was investigated with a previously described[3] setup. We used cells with uniform LC layer thicknesses 18.4 and 90 μm, similar in configuration to the planar cell of Ref. 3, the helicoid axis being perpendicular to the walls. The dc electric field was perpendicular to the helicoid axis.

The temperature dependence of the second-harmonic intensity $I_{2\omega}$ is shown in Fig. 1. Synchronism is observed at the temperature $T_{C^*-A} - T \approx 8$ °C, corresponding[4] to an average angle $\sim 23°$ between molecule inclination and the normal to the smectic layers, and in good agreement with the calculations. The half-width of the synchronism curve and the degree of polarization $\rho = 0.95$ of the second-harmonic radiation attest to good homogeneity of the LC layer orientation.

The dependence of $I_{2\omega}$ on the electric field intensity E_0 is shown in Fig. 2. The curve has three distinct sections: *a*—the helicoid is not yet untwisted and the SHG signal is weak; *b*—the helicoid becomes untwisted and the second-harmon-

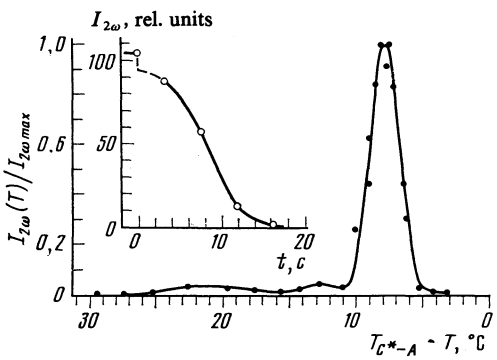

FIG. 1. Temperature dependence of the second harmonic of light. LC layer thickness μm, dc electric field intensity $E_0 = 3$ kV/cm. The inset shows the dependence of the second-harmonic intensity on the time elapsed from the instant of turning off the helicoid-untwisting field. LC layer thickness 18.4 μm, $E_0 = 4.1$ kV/cm, $T_{C^*-A} - T = 16.1$ °C.

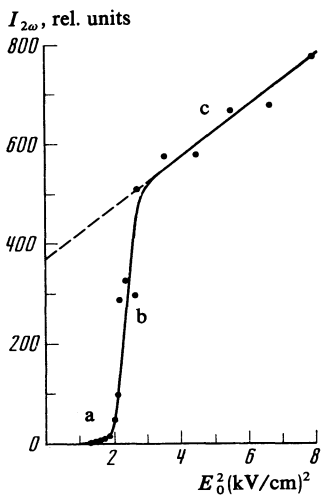

FIG. 2. Second-harmonic intensity at the synchronism maximum vs dc electric field intensity. LC layer thickness 90 μm.

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ic intensity grows rapidly because the synchronization condition sets in; *c*—the helicoid is completely untwisted. The growth of $I_{2\omega}$ on the last section of the curve is obviously governed by the known mechanism of optical SHG induced by the electric field. Extrapolation of this section to zero field yields a nonzero second-harmonic intensity, thus confirming the presence of spontaneous polar ordering of the molecules in the smectic C^* phase. The appreciable slope of the plot of $I_{2\omega}$ vs E_0 on section *c* of Fig. 2 is evidence that the contributions made to the SHG by the spontaneous and field-induced polarizations are comparable at the given temperature, and that it is impossible to estimate their individual shares in this experiment. Such an estimate, however can be obtained by investigating the kinetics of the falloff of $I_{2\omega}$ if the helicoid relaxation time after turning off the untwisting field is long enough. This condition is realized in the low-temperature region of the C^* phase. The decrease of $I_{2\omega}$ with time after turning off the electric field at the temperature $T_{C^*-A} - T = 16.1$ °C is shown in the inset of Fig. 1; the synchronism was achieved in this case by rotating the cell. At the instant $t = 0$ the electric field is turned off, and since the field-induced ordering of the dipole relaxes within a time on the order of 10^{-7} sec, the SHG after a time $t = 3.3$ sec is certainly due only to the spontaneous polar ordering of the molecules. Comparing the values of $I_{2\omega}$ at these two instants of time we see that at the given lowered temperature, in an untwisting field of the order of 4 kV/cm, the electric-field-induced SHG mechanism makes no significant contribution to the resultant value of $I_{2\omega}$. This may possibly be due to an increased contribution to $I_{2\omega}$ from the spontaneous polar ordering of the molecules with decreasing temperature, as manifest in particular by the increase of the spontaneous polarization P_s.[4] The nonlinear second-order susceptibility $\chi^{(2)}$ calculated from the value of $I_{2\omega}$ at $t = 3.3$ sec is 3.5×10^{-12} cgs esu. An estimate of $\chi^{(2)}$ from the value of P_s for DOBAMBC, according to the known relation between them,[5] agrees with the experimentally obtained value.

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