Pretransitional effects near blue phases of a cholesteric liquid crystal

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The rotation of the polarization plane of light (RPPL) in an isotropic liquid and in the blue and "foggy" phases of a short-pitch cholesteric liquid crystal (LC) is measured. Nonmonotonic behavior of the pretransitional RPPL is observed near the transition to the "foggy" phase. The coefficients in the Landau–de Gennes expansion are determined.

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In recent years,[1,3] phenomena occurring near the isotropic liquid (IL)-cholesteric with short pitch have been studied extensively. This transition is of interest in that in an interval of ~1 K between the temperature of the IL-LC transition and the temperature of absolute instability of the liquid, several stable phases appear (blue phases) with anomalous properties. The peculiarities of the pretransitional effects in the short-pitch cholesterics near the transition to the blue phases is indicated in the theoretical papers Refs. 4-5. The most characteristic pretransitional effect for a cholesteric LC is rotation of the polarization plane of light.

The temperature dependence of RPPL for cholesterol pelargonate in the blue phases *BP* I (363.25–363.65 K), *BP* II(363.65–363.75 K) and the "foggy" (363.75–363.8 K) phases and in the isotropic liquid ($T > T_c = 363.8$ K) is shown in Fig. 1. The temperature is regulated and measured to within 0.005 K and the angle to within 0.15′. The temperature of the transitions between the blue phases was determined according to the measurement of RPPL and the lattice parameters of the blue phases. The formation of the phases was monitored visually at the same time. From the jumps in RPPL at the phase boundaries, it is evident that all phase transitions are of first order. In the temperature interval presented, RPPL consists of a molecular contribution,[1)] as well as primarily a structural contribution ϕ_s in *BP* I and *BP* II ($\phi_s > \phi_0$) and of a fluctuation contribution in the isotropic liquid ($\phi_f \lesssim \phi_0$). A decrease of the structural RPPL in blue phases with increasing temperature and with the phase transition *BP* I→*BP* II correlates with the change in the parameters of the cubic lattice of the LC in this temperature range. The parameter of orientational ordering of molecules decreases by 10% with a transition between the cubic phases *BP* I→*BP* II.

In the isotropic phase, RPPL has a maximum near the transition to the LC state. This behavior of pretransitional RPPL can be explained in terms of the Landau–de Gennes theory of short-pitch cholesterics. According to Ref. 4, pretransitional RPPL is related to the fluctuational modes of the flat and conical spirals. At temperatures $T - T_c > 1$ K, RPPL is determined by the contribution of the modes of the conical spiral.[4–6]

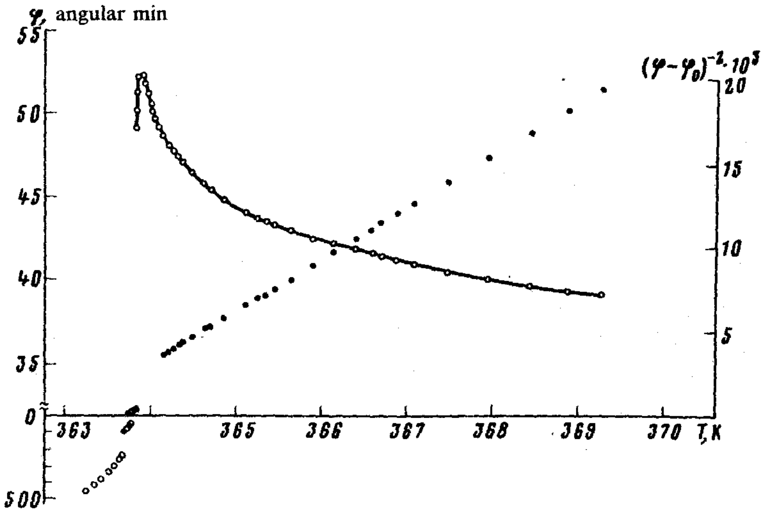

FIG. 1. ○ – RPPL(φ) in the blue "foggy" phases of LC and in the pretransitional region; ● – the dependence $(\phi - \phi_0)^{-2}$ of the liquid at $T - T_c > 0.3$ K, $d = 2$ mm and $\lambda = 633$ nm.

$$\phi - \phi_0 = \frac{k_0^2\, q_0\, k_B\, T}{48\, \epsilon_0^2\, (1 + c/2b)^{3/2} (ba_0)^{1/2}\, \tau_3^{1/2}},$$

where $\tau_3 = T - T_3^*$ and T_3^* is the temperature of the absolute instability of the IL relative to the transition to the conical spiral structure.

The experimental dependence of $(\phi - \phi_0)^{-2}$ at these values of T is described well by a linear function (see Fig. 1). Using the experimental value of the pitch of the spiral of the cholesteric near the transition to BP I, $p = 2350$ Å, $\varepsilon = 2.2$[7], and $c/b = 0.6$,[6] and applying the method of least squares to the experimental data, the product of the coefficients in front of the quadratic and gradient terms in the Landau–de Gennes expansion were determined: $a_0 b = 4 \times 10^2$ erg/cm^4·K, $T_3^* = 363$ K. The sign of the rotation in the plane of the fluctuation mode differs[4] from the sign of the rotation in the conical spiral mode. The relative magnitude of the contributions of these modes is determined by the ratio of their correlation lengths[4,5]

$$\xi_s = \xi_0\, (T^*/\tau_s)^{1/2}, \quad s = 1,\ 3,$$

where $\tau_1 = T - T^* - \dfrac{b}{a_0} q_0^2$,

$$\tau_3 = T - T^* - \frac{b}{a_0} q_0^2\, \frac{1}{4(1 + c/2b)}.$$

To estimate ξ_0, we shall use the Hornreich-Shtrikman phase diagram[3]: $\xi_0 = 7 \div 9$ Å, and at the same time $a_0 \sim 1.3 \times 10^7$ erg/cm^3·K and $b \sim 3 \times 10^{-5}$ erg/cm, which are an order of magnitude greater than the corresponding quantities for nematic MBBA.

Near a phase transition to the "foggy" phase the correlation lengths of the modes in our case differ appreciably: $\xi_1/\xi_3 = 1.3 \div 1.7$ ($T - T_c = 0.2$ K), since fluctuations of the plane of the spiral are energetically favorable. Softening of the flat mode leads to a considerable contribution of the mode $s = 1$ to RPPL and to nonmonotonic behavior of the fluctuational rotation. In the "foggy" phase, the RPPL is of the same order of magnitude as the fluctuational rotation in the isotropic phase near the transition to the LC state. This could indicate that fluctuations play an important role in the "foggy" phase as well, where the temperature of the transition is near the divergence of fluctuations of the flat spiral. In this case, renormalization effects are strong.[8] The development of a quantitative theory of pretransitional effects in the case of strongly developed fluctuations will permit explaining the role of interacting fluctuations in the formation of blue and "foggy" phases.

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[1)]The intrinsic molecular rotation, which was measured in a 1% benzene solution, is 32' and has the same sign as RPPL in an isotropic liquid.

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