

# Optical method of determining the critical exponents of the phase transition from an isotropic liquid to a nematic crystal

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A method is proposed for studying the pre-transition phenomena in a nematic liquid crystal on the basis of the pre-transition behavior of a cholesteric liquid crystal with a large helix pitch.

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The phase transition from an isotropic liquid to a nematic liquid crystal has recently become the subject of intensive experimental study.<sup>(1-7)</sup> However, the question of the character of the anomalies in the pre-transition region remains open. The following three assumptions are possible: the Ginzburg parameter is small and the exponents are obtained by the self-consistent-field method; by accident, the phase transition takes place near the tricritical point and it is necessary to take into account the logarithmic corrections to the self-consistent field<sup>(8)</sup>; the exponents have the scaling values.<sup>(9)</sup> The results of calorimeter measurements<sup>(4)</sup> indicate that self-consistent-field methods are not applicable. However, the accuracy of the experiment in<sup>(4)</sup> is insufficient to

make a choice between the critical and tricritical behavior. In<sup>(1-4)</sup> they measured the heat-capacity exponent. To determine more precisely the character of the phase transition, it is proposed to consider the rotation of the plane of polarization of the electromagnetic wave in a mixture of the given substance with a small amount of optically active molecules. The known optical experiments<sup>(5-7)</sup> on Rayleigh scattering of light in the transition region suffer from a shortcoming connected with light scattering by particle-like impurities with diameters on the order of  $10^3 \text{ \AA}$ ,<sup>(7)</sup> which are present in the sample. Since these impurities have no optical activity, the proposed method serves both as a refinement of the experimental results on the anomaly of the heat capacity<sup>(1-4)</sup> and as an explanation of the discrepancy between the results of the optical measurements<sup>(6,7)</sup> discussed in<sup>(7)</sup>. A weak solution of optically active molecules in the original liquid crystal below the phase-transition point has cholesteric order with a large helix pitch.

In the pre-transition region, the correlation length  $\xi$  of the molecule-orientations, which characterizes the short-range order which is inherent in nematic liquid crystals, does not exceed  $200 \text{ \AA}$ ,<sup>(5-7)</sup> which is much less than the large pitch  $p \approx 10^4 \text{ \AA}$  of the cholesteric helix. The thermodynamic behavior of this mixture is therefore the same as that of the initial liquid crystal. In an optically active liquid crystal, however, it is possible to measure the pre-transition polarization-plane rotation due to the appearance in the permittivity tensor  $\epsilon_{\alpha\beta}(\mathbf{k})$  of a term that is invariant to the substitution  $\mathbf{k} \rightarrow -\mathbf{k}$ <sup>(10)</sup>:

$$\epsilon_{\alpha\gamma}(\mathbf{k}) - \epsilon_{\alpha\gamma}(-\mathbf{k}) = \frac{k_\alpha^2}{4\pi\epsilon_0} \int \frac{d\mathbf{q}}{(2\pi)^3} D_{\beta\delta}(\mathbf{q} + \mathbf{k}) [G_{\alpha\beta}^{\gamma\delta}(\mathbf{q}) - G_{\alpha\beta}^{\gamma\delta}(-\mathbf{q})], \quad (1)$$

where  $k_\alpha = \epsilon_0^{1/2}\omega/c$ ,  $|\mathbf{k}| = k_0$ , and the photon Green's function in a gauge  $\phi=0$  is

$$D_{\beta\delta}(\mathbf{k}) = \frac{4\pi}{k_\alpha^2 - k^2} \left( \delta_{\beta\delta} - \frac{k_\beta k_\delta}{k_\alpha^2} \right).$$

In (1),  $G_{\alpha\beta}^{\gamma\delta}(\mathbf{q})$  is the correlation function of the order parameter  $Q_{\alpha\beta}(\mathbf{q})$  and is determined by the expansion of the free energy near the phase transition of the isotropic liquid into a cholesteric liquid crystal, as proposed by de Gennes<sup>(11)</sup>

$$\begin{aligned} \frac{F - F_0}{T} = \frac{1}{2!} \int \frac{d\mathbf{q}}{(2\pi)^3} [ a Q_{\alpha\beta}^2 + b (q_\alpha Q_{\beta\gamma})^2 + c q_\alpha Q_{\alpha\beta} q_\gamma Q_{\gamma\beta} \\ + 2bq_\alpha q L_{\alpha\beta} Q_{\alpha\gamma} Q_{\gamma\beta} + \mu Q_{\alpha\beta} Q_{\beta\gamma} Q_{\gamma\alpha} + \lambda (Q_{\alpha\beta}^2)^2 ], \end{aligned} \quad (2)$$

where  $L_{\alpha\beta} = e_{\alpha\beta\gamma} q_\gamma / q$  and  $q_0 = 2\pi/p$ . From experiments on the scattering of light<sup>(6)</sup> it is known that  $c \ll b$ . We shall henceforth assume  $c=0$ . By virtue of the inequality  $\xi \ll q_0^{-1}$ , the optically active part of the Hamiltonian (2) can be regarded as a perturbation. The fluctuations of the order parameter  $Q_{\alpha\beta}$  lead to the appearance of an anomalous dimensionality of the operator  $L_{\alpha\beta}$ , a fact that must be taken into account in the calculation of the integral (1). The value of the anomalous dimensionality is determined by the  $\epsilon$ -expansion method. For a nematic liquid crystal ( $q_0=0$ ) there are two

days of calculating the exponents from the  $\epsilon$  expansion.  $Q_{\alpha\beta}$  is a tensor in a space with  $d=4$  in the first case and in three-dimensional space in the second. At  $q_0 \neq 0$  only the second possibility exists, since an absolutely antisymmetrical object  $e_{\alpha\beta\gamma}$  exists only in three-dimensional space. Calculation of the anomalous dimensionality of the operator  $L_{\alpha\beta}$  is similar to the calculation of the anomalous dimensionality  $\eta$  of the isotropic part of a Green's function.<sup>(9)</sup> If  $\eta_L < \eta$ , then isotropization of the Green's function takes place at the second-order phase-transition point ( $L_{\alpha\beta} \rightarrow 0$ ). This situation arose for the anomalous dimensionality of the anisotropic parts of Green's functions determined by the coefficient  $c$ .<sup>(9)</sup> In our case  $\eta_L = \eta$ :

$$\begin{aligned} (G_{\alpha\beta}^{\gamma\delta})^{-1} &\sim q^2 - \eta [\Delta_{\alpha\beta}^{\gamma\delta} - (q_0/2q)(L_{\alpha\gamma}\delta_{\beta\delta} \\ &+ L_{\alpha\delta}\delta_{\beta\gamma} + L_{\beta\gamma}\delta_{\alpha\delta} + L_{\beta\delta}\delta_{\alpha\gamma})], \\ \Delta_{\alpha\beta}^{\gamma\delta} &= (1/2)(\delta_{\alpha\gamma}\delta_{\beta\delta} + \delta_{\alpha\delta}\delta_{\beta\gamma}) - (1/3)\delta_{\alpha\beta}\delta_{\gamma\delta}. \end{aligned}$$

Thus, the fluctuations of the order parameter  $Q_{\alpha\beta}$  do not influence the pitch of the cholesteric helix and the integral (1) is determined by the purely nematic indices. Assuming  $k_0 \approx q_0$ ,  $\xi \ll q_0^{-1}$  we expand the integrand of (1) up to first order in  $q_0$  and  $k_0$ :

$$\epsilon_{\alpha\gamma}(\mathbf{k}) - \epsilon_{\alpha\gamma}(-\mathbf{k}) = \frac{k_0 q_0 \xi L_{\alpha\gamma}}{6\pi \epsilon_0 b}.$$

The quantity  $\eta$  is numerically small ( $\eta \approx 0.02$ ).

If  $1 \ll \ln(\xi/\xi_0) \ll \eta^{-1}$ ,<sup>(9)</sup> then the quantity  $\eta$  can be neglected. In this case

$$\epsilon_{\alpha\gamma}(\mathbf{k}) - \epsilon_{\alpha\gamma}(-\mathbf{k}) = \frac{k_0 q_0 \xi^{1-\eta} \xi_0^\eta L_{\alpha\gamma}}{6\pi \epsilon_0 b}.$$

The rotation  $\psi/d$  of the polarization plane is equal to

$$\frac{\psi}{d} = \frac{k_0^2 q_0 \xi}{24\pi \epsilon_0^2 b} \sim r^{-\gamma/2}, \quad \gamma = 1.26. \quad (2)$$

At the point of the phase transition into a cholesteric liquid crystal  $\psi/d \approx 10^\circ/\text{cm}$ .

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