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POLARIZATION OF THE SCATTERED LIGHT IN THE ISOTROPIC PHASE OF A CHOLESTERIC LIQUID CRYSTAL

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ABSTRACT: In the only measurement to date of the ratio R_d of depolarized to polarized light scattering intensities in the isotropic phase of a cholesteric liquid crystal, Harada and Crooker reported that the value of 3/4 predicted by the order parameter fluctuation theory of de Gennes was not observed. We present here measurements of R_d in cholesteryl oleyl carbonate (COC) using a more precise rotating polarizer technique. R_d is observed to have a strong temperature dependence which we can explain by the de Gennes theory plus an additional fluctuating component as described by Courtens.

Measurements of pretransitional phenomena near the cholesteric-isotropic phase transition of a cholesteric liquid crystal have shown that, contrary to previous expectations, this transition has features quite different from the nematic-isotropic transition. In a paper describing light scattering measurements in the isotropic phase of cholesteryl oleyl carbonate (COC), Harada and Crooker[1] have shown that: (1) the Rayleigh intensity and linewidth change continuously through the extrapolated second-order phase transition temperature T*, thereby making it impossible to detect any first-order phase transition temperature T_c optically; (2) below T* the autocorrelation function of the scattered light becomes a sum of exponentials; (3) the ratio of the depolarized to polarized scattered light intensities is .691 ± .007 instead of 3/4 as predicted by the order parameter fluctuation theory of de Gennes[2]. In addition, Mahler, et al.[3] have shown that the high temperature extrapolations of the Rayleigh linewidth and intensity lead to different values of T*, contrary to the de Gennes[2] prediction. Finally, Ajgaonkar and Keyes[4] have observed a pretransitional increase in the shear viscosity. The discovery of the so-called "blue phase"[5], a feature found at many cholesteric-isotropic transitions but not at nematic-isotropic transitions has further complicated the picture.

In this letter we report, using a more precise rotating polarizer technique, the ratio R_d of the depolarized to polarized components of the scattered light intensity in the isotropic phase of COC. The details of the temperature behavior of this ratio are now observable, and we find that it decreases linearly with increasing temperature from a value of .75 at the extrapolated second-order phase transition temperature T*. We are able to explain our data by assuming that an additional fluctuating component contributes to the scattering, and we show that our parameters also explain the data of Harada and Crooker.

The COC used in our experiments (Aldrich Chemical Co., lot number 111107) was first filtered through a .1 micron filter, degassed for 18 hours in a vacuum system, then sealed under dry nitrogen in a 10 mm square scattering cell. Before and after the experiment the quality of the sample was monitored by measuring the depolarized component of the scattered intensity as a function of temperature and determining T*. This value remained constant at (37.07 ± .07) $^{\circ}$C. During the experiment the temperature of the sample was controlled to within .02 $^{\circ}$C by a circulating water bath.

Light scattering was performed at 90° using a 5 mw He-Ne laser and incident light polarized perpendicular to the scattering plane (V polarization). The scattered light was passed through a rotating polarizer (ω = 4 rev/s), and detected using conventional photon counting techniques; the resulting signal pulses were then accumulated by a PDP-8/E minicomputer operating as a multichannel scaler. The computer also received photoelectric timing pulses from the rotating polarizer so that signal pulses corresponding to the polarized scattering (VV) and depolarized scattering (VH) could be stored in separate memory locations. At the end of a run the accumulated VV and VH counts were read out, corrected for background, and the ratio of the depolarized to polarized intensities $R_d = I_{VH}/I_{VV}$ computed.

In Fig. 1 we present the ratio R_d as a function of the reduced temperature T/T*. The scatter of the data is caused by uncertainties in the timing pulses and not by photon counting statistics. Clearly the predicted ratio of .75 is not achieved for T>T*. We ascribe this deviation of R_d from 3/4 to the presence of density fluctuations as has been discussed by Courtens[6]. Let the VV scattering intensity I_{VV} consist of a contribution J^Q from order parameter fluctuations and a small contribution J^ρ from density fluctuations; thus $I_{VV} = J^Q + J^\rho$. According to the de Gennes theory $I_{VH} = KT/(T-T^*)$, where K is a constant of proportionality, and $J^Q = (4/3)I_{VH}$. Thus, for J^ρ small,

$$R_d = \frac{I_{VH}}{I_{VV}} \simeq \frac{3}{4}\left[1 - \frac{3}{4}\left(\frac{J^\rho}{KT}\right)\left(T-T^*\right)\right] \qquad (1)$$

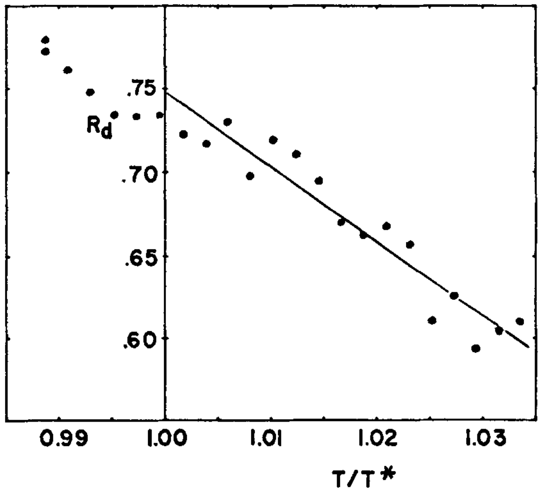

FIGURE 1. Depolarization ratio R_d versus reduced temperature.

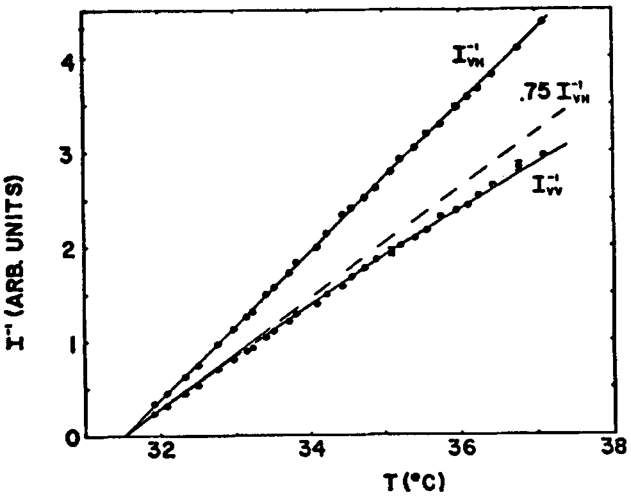

FIGURE 2. Data from Ref. 1. The lines are a fit to our theory as explained in the text.

The solid line in Fig. 1 is a least squares fit to Eq. 1 in the form $R_d = a + b(T-T^*)$, using the data for $T/T^*>1$ and $T^* = 37.07\ ^{\circ}C$ determined previously from the depolarized intensity measurements. We obtain $a = .748 \pm .008$ in good agreement with de Gennes value of 3/4. From the slope b we obtain $J^{\rho}/KT = (.025 \pm .002)\ ^{\circ}C^{-1}$. This value is 30 times that for MBBA and is by far the largest measured value of J^{ρ}/KT to date[6]. Nevertheless, our value of J^{ρ}/KT is not unexpected if one realizes that J^{ρ}/KT is a strong function of molecular volume v. Indeed, the expression $J^{\rho}/KT = Av^B$ (where A and B are constants) yields Courtens' values for MBBA and four of its homologues to within 15%, and using $v = 1113\ \mathring{A}^3$ for COC, the expression extrapolates to within 15% of our value.

A novel feature of Fig. 1 is the continuation of the data below T*. This is associated with the inability to optically detect the first-order phase transition temperature T_c and with the presence of the blue phase. Such behavior is outside the scope of the de Gennes model[2].

We now show that the data of Harada and Crooker[1] (Fig. 2) are consistent with Eq. 1 by refitting their data (solid lines in Fig. 2) to $I_{VH}^{-1} = (\text{const})(T-T^*)$ and $I_{VV}^{-1} = I_{VH}^{-1} R_d(T)$. We use $R_d(T) = a + b(T-T^*)$, where a and b have the values determined in this paper and we use Harada and Crooker's value of $T^* = 31.5\ ^{\circ}C$. The fit is quite good ($\chi^2 = 0.8$) and we are able to explain their data within the framework of the de Gennes theory. Notice also that their value of T* is 5.6 °C less than ours; thus it is apparent that shifts in the transition temperature only move the curves on the temperature axis without changing the essential behavior.

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