Comments and Addenda

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Pretransitional viscosity in the isotropic phase of cholesteric liquid crystals

P. P. Crooker and W. G. Laidlaw*

Department of Physics and Astronomy, University of Hawaii, Honolulu, Hawaii 96822

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The measurements of the anomalous effective viscosity η obtained by Keyes and coworkers are analyzed in terms of the two separate viscosity coefficients ν and μ.

In the past several years a number of papers have appeared in which the pretransitional behavior near the isotropic to cholesteric transition has been probed.[1] Of particular interest have been those cases such as cholesteryl oleyl carbonate (COC) where an intermediate "blue phase" can be detected.[2]

Recently, Keyes and coworkers[3,4] reported light scattering and shear viscosity measurements near the transition temperature which show, to quote Keyes and Yang,[3] "an anomaly in the viscosity coefficient ν similar to the anomaly in η." These authors go on to comment that the anomalous decrease in ν goes[3] "hand-in-hand with the increase in η."

A careful analysis of the pretransitional theory of liquid crystals leads one to the position that the shear viscosity η measured in a capillary-flow experiment is an effective viscosity which depends explicitly on "uncoupled or ordinary shear viscosity η_0." For example, Stephen and Straley[5] as long ago as 1974 pointed out that the shear viscosity is $\eta_0 = \eta' + \nu^2/\xi$ which, in terms of the notation employed by DeGennes[6] and subsequently, by Keyes *et al.*, is just

$$\eta = \eta_0 + 2\mu^2/\nu. \quad (1)$$

We argue that the ordinary viscosity does not have anomalous behavior in the region of a liquid-crystal transition so that the anomalous behavior in η can then be attributed to the anomalous behavior in ν and/or the anomalous behavior of μ.

We now apply these ideas to the interpretation of the viscosity data of Keyes *et al.*[3,4] (see Fig. 1). In the temperature region of COC where the viscosities show only Arrhenius-type temperature dependence, Harada and Crooker[1(a)] have shown

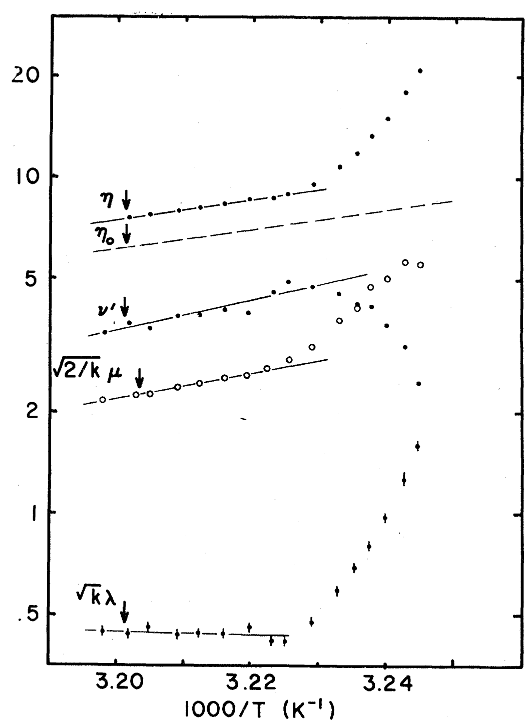

FIG. 1. Coefficients of the hydrodynamical equations. η (poise) and ν' (10^{-5} s K) are from Ref. 3. η_0 (poise), $\sqrt{2/k}\ \mu$(10^{-5} s K poise)$^{1/2}$ and $-\sqrt{k}\ \lambda$(poise/10^{-5} s K)$^{1/2}$ are discussed in text.

that $\mu^2/\eta\nu = 0.093$, a value similar to that found in nematics.[7] Using Eq. (1) we then get $\eta_{0A} = 0.81\eta_A$ where the subscript A refers to values in the Arrhenius region. We now postulate that η_0 remains Arrhenius-type well into the anomalous region; this is reasonable since η_0 is the ordinary or uncoupled viscosity and we associate the anomalous behavior with order-parameter coupling. Hence,

$$\eta = 0.81\eta_A + 2\mu^2/\nu$$

and the anomaly in η will be due to the anomalous behavior of μ and ν.

Given the data reported by Keyes and Yang for η and $\nu' = \nu/k$ (k is a constant), it is now an easy matter to extract the temperature dependence of μ as shown in Fig. 1. We find that μ shows a very significant pretransitional increase which, together with the decrease in ν, results in the significantly increased η. Also, we calculate the temperature dependence of the flow-order-parameter-coupling coefficient λ which, along with η_0 and $1/\nu$, would appear naturally in the expression for the order-parameter flux $R_{\alpha\beta}$ in terms of the conjugate forces $e_{\alpha\beta}$ and $\phi_{\alpha\beta}$ as in

$$(\partial/\partial t)Q_{\alpha\beta} = R_{\alpha\beta}, \quad R_{\alpha\beta} = \lambda e_{\alpha\beta} + (1/\nu)\phi_{\alpha\beta}$$

and in the momentum equation

$$(\partial/\partial t)(\rho v_\alpha) = \partial/\partial_\beta \sigma_{\alpha\beta}, \quad \tfrac{1}{2}\sigma_{\alpha\beta} = \tfrac{1}{2}\eta_0 e_{\alpha\beta} - \lambda\phi_{\alpha\beta}\,.$$

Figure 1 shows that λ exhibits an anomalous increase at the phase transition which parallels that of $1/\nu$. This is not unexpected since both λ and $1/\nu$ are coefficients associated with the order-parameter coupling.

Experiments to independently probe the pretransitional behavior of μ (which our analysis predicts) would seem to be in order.

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*Permanent address: Department of Chemistry, University of Calgary, Calgary, Alberta, Canada T2N 1N4.

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