Twist viscosity and splay elastic constant near the nematic–smectic-A–smectic-C multicritical point

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The twist viscosity has been measured in a binary liquid-crystal mixture above the nematic–smectic-C transition near the nematic–smectic-A–smectic-C multicritical point using the dynamic Fréedericksz deformation and found to exhibit a monotonic divergence. This study also shows that the splay elastic constant has a relatively mild temperature dependence compared with that of the bend elastic constant.

Extensive experimental[1–12] and theoretical[13–15] work has been devoted to the study of the nematic–smectic-A–smectic-C (N-A-C) multicritical point in binary liquid-crystal mixtures. Several key experimental findings, such as the form of the smectic susceptibility using x-ray scattering[4] and the behavior of the bend elastic constant K_3 above the nematic–smectic-C (N-C) transition far away from the N-A-C point using light scattering,[3] are found to be consistent with the predictions of the Chen-Lubensky model,[13] in which the N-A-C point is a Lifshitz point. On the other hand, several experimental quantities which are expected to diverge above the N-C transition near the N-A-C point, including the longitudinal ($\xi_\parallel$) and transverse ($\xi_\perp$) smectic correlation lengths[4,9] and the inverse of the bend-mode light-scattering intensity,[3,5,10] show anomalous nonmonotonic temperature dependences. The fact that K_3, which diverges montonically, is not proportional to $\xi_\parallel$ near the N-A-C point is not described by the existing models.[7] Another theoretical prediction, that the pretransitional behavior of the twist viscosity γ_1 should be proportional to $\xi_\parallel$ near the N-A-C point,[14] has not yet been tested. Recently, the light-scattering anomaly has been found to be related to tilt fluctuations that manifest themselves in the nonhydrodynamic regime near the N-A-C point.[12] In an attempt to find out whether similar anomalous behavior is also present in other physical properties, we have measured γ_1 and the splay elastic constant K_1 above the N-C transition near the N-A-C point using dynamic Fréedericksz deformation.

We studied mixtures of heptyloxy-p'-pentylphenylthiobenzoate ($\bar{7}$S5) containing various mole fractions x of octyloxy-cyanobiphenyl (8OCB), in which the multicritical concentration $x_{N\text{-}A\text{-}C}$ has been found to be[9] 0.0217. This Brief Report contains results obtained in the mixture with a near-multicritical concentration $x=0.0191$, which exhibits anomalous properties above the N-C transition in other measurements. The effect of Fréedericksz deformation on the birefringence was measured optically using a planar-aligned sample with the magnetic field normal to the director.[16,17] When the magnetic field is increased to a value beyond the critical field,

$$H_c=(\pi/d)(K_1/\Delta\chi)^{1/2} , \tag{1}$$

where d is the sample thickness and $\Delta\chi$ is the anisotropy in the diamagnetic susceptibility, and then turned off, the birefringence returns to the equilibrium value with an exponential decay time:

$$\tau=\gamma_1/(2\Delta\chi H_c^2) . \tag{2}$$

Equation (2) is expected to be valid because of the small backflow effect in our experimental geometry.[16]

The result for the temperture dependence of $\gamma_1/\Delta\chi$, as determined from Eq. (2), is shown in Fig. 1 for the mixture with $x=0.0191$. It can be seen that $\gamma_1/\Delta\chi$ diverges monotonically as the N-C transition temperature $T_{N\text{-}C}$ is approached from above. Thus the twist viscosity in this mixture shows a pretransitional behavior that would be expected in an ordinary nearly second-order phase transition, in contrast to the anomalous behavior in other phys-

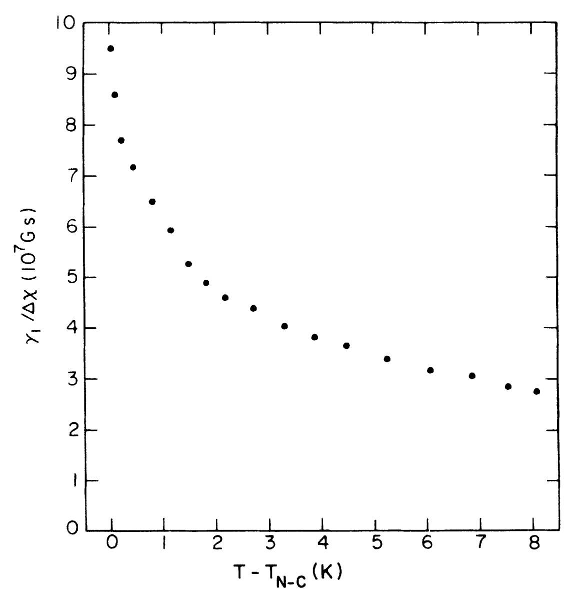

FIG. 1. Temperature dependence of $\gamma_1/\Delta\chi$ above $T_{N\text{-}C}$ in a $(\bar{7}S5)_{1-x}(8OCB)_x$ mixture, with $x=0.0191$.

ical quantities observed in x-ray and light-scattering measurements, and in contradiction to the theoretical expectation that it should be proportional to $\xi_{\parallel}$ near the N-A-C point.[14] In fact, the divergence in γ_1 is qualitatively similar to that found in many materials that exhibit a second-order nematic–smectic-A (N-A) transition.[16–21] This temperature dependence of γ_1 has also been found to account for the results of a recent splay-mode quasielastic–light-scattering experiment.[12] We have not attempted to extract any critical exponent from this measurement, since there is no theoretical prediction for its value and it is known that the presence of the large background contribution makes such an analysis extremely tricky.[22]

In the course of this study, we have also determined the splay elastic constant K_1 using the measured values of H_c in accordance with Eq. (1). The temperature dependence of K_1 is shown in Fig. 2, where we have also plotted for comparison the behavior of the bend elastic constant K_3 that was reported previously in another Fréedericksz deformation experiment using a different sample geometry.[7] Two observations can be made. The first is that K_1 shows a monotonic gradual increase as the temperature is lowered toward $T_{N\text{-}C}$. This result is somewhat reminiscent of the regular behavior of K_1 in most nematics near the N-A transition.[23–25] The second is that the relatively mild temperature dependence of K_1 is in sharp contrast to the strong divergence of K_3. In fact, the increase in K_3 is more than an order-of-magnitude higher than that in K_1. The nondivergent behavior of K_1 near the N-A-C point reported here is interesting, because it is generally expected theoretically that, *away* from the N-A-C point, all three elastic constants should diverge in the nematic phase near the N-C transition.[13,26,27] One should note, however, that our observed difference in the degree of divergence of K_1 and K_3 above the N-C transition is qualitatively consistent with that described in the Chen-Lubensky model *near* the N-A-C point,[13] even though the precise relationship between the elastic constants and the smectic correlation lengths predicted by that model is clearly not obeyed.

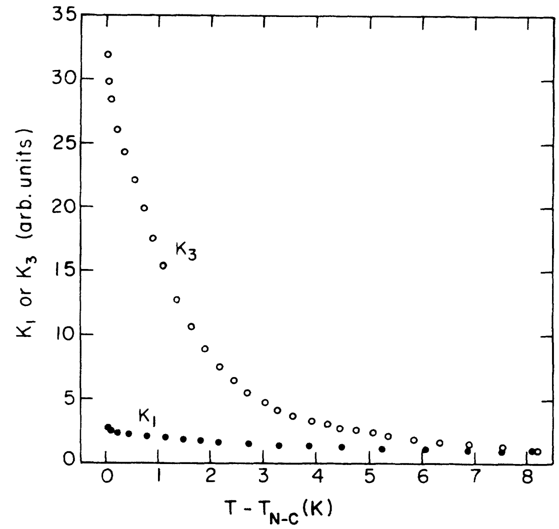

FIG. 2. Temperature dependence of K_1 (●) and K_3 [(○) from Ref. 7] above $T_{N\text{-}C}$ in a $(\bar{7}S5)_{1-x}(8OCB)_x$ mixture, with $x=0.0191$. Both quantities have been rescaled to be equal to unity (in arbitrary units) at the highest temperature.

In summary, a dynamic Fréedericksz deformation experiment has revealed no anomalous behavior in the twist viscosity and in the splay elastic constant in a $\bar{7}$S5-8OCB mixture above the N-C transition near the N-A-C multicritical point.

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