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PRELIMINARY COMMUNICATION

Degeneracy of rotational viscosities at the chiral smectic C-smectic A transition in DOBAMBC

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From measurements of the electroclinic effect and the dielectric strength of the soft mode close to the chiral smectic C-smectic A transition we show that the rotational viscosities associated with the Goldstone and soft modes are degenerate at T_c, in agreement with the predictions of the theoretical model.

It has been predicted theoretically [1] that the relaxation frequencies of the amplitude and phase fluctuations of the order parameter in ferroelectric liquid crystals are degenerate at the phase transition from S_C^* to S_A phases

$$f_G(T_c) = f_S(T_c) = f_A(T_c) = \frac{\Gamma K_3}{2\pi} q_0^2. \tag{1}$$

Here f_G and f_S represent the relaxation frequencies of the Goldstone and soft mode fluctuations in the S_C^* phase, and f_A represents the soft mode relaxation frequency in the S_A phase. The constant K_3 is the twist elastic constant while q_0 is the wavevector of the pitch of the helix at the transition temperature T_c. A necessary condition for this prediction is that the kinetic coefficients, which are inverse rotational viscosities [2] associated with each relaxation mode, are also degenerate at T_c

$$\Gamma = \Gamma_G = \Gamma_S = \Gamma_A.$$

Equation (1) has been confirmed experimentally [3], but independent confirmation of the rotational viscosity degeneracy at T_c would give additional support to the validity of the model. Here we present experimental evidence that the rotational viscosities Γ_G^{-1} and Γ_A^{-1} are degenerate at T_c within the experimental error.

The rotational viscosity Γ_A^{-1} can be determined from [4, 5]

$$f_A = \frac{\Gamma_A}{2\pi}[\alpha(T - T_c) + (K_3 - \varepsilon\mu^2)q_0^2], \tag{2}$$

$$\varepsilon_0 \Delta\varepsilon_A = \frac{\varepsilon^2 C^2}{\alpha(T - T_c) + (K_3 - \varepsilon\mu^2)q_0^2},$$

$$\frac{d\theta}{dE} = \frac{\varepsilon C}{\alpha(T - T_c) + (K_3 - \varepsilon\mu^2)q_0^2}, \tag{3}$$

where α, ε, C and μ are coefficients appearing in the Landau free energy expansion [6]. From a measurement of the frequency dependence of the complex dielectric susceptibility we can determine the relaxation frequency f_A and dielectric strength $\Delta\varepsilon_A$ of the soft mode. If we measure the induced tilt angle as a function of the applied field the electroclinic coefficient $d\theta/dE$ can be determined. Equations (2) and (3) then allow us to determine εC, α and Γ_A separately. If we are studying a ferroelectric liquid crystal with a small dielectric strength for the soft mode we must use some other measurement. Such an experiment was done by Garoff and Meyer [7, 8] when they studied the change in the birefringence of DOBAMBC. The tilt angle induced by a sinusoidal voltage E with an angular frequency ω is described by an amplitude θ_0 and a phase δ relative to the applied field given by

$$\theta_0 = \frac{\varepsilon C E}{(A^2 + \omega^2/\Gamma_A^2)^{1/2}}, \tag{4a}$$

$$\tan\delta = -\omega/A\Gamma_A. \tag{4b}$$

Taking

$$A = a\left(\frac{T - T_c}{T_c}\right)^{\gamma}$$

and

$$\Gamma_A^{-1} = \Gamma_0 \exp\left(\frac{U}{kT}\right)$$

they wrote equation (4 *b*) as

$$\ln\tan(\delta_b - \delta) = \frac{U}{kT} + \ln\frac{\omega\Gamma_0}{a} - \gamma\ln\left(\frac{T - T_c}{T_c}\right), \tag{5}$$

where δ_b is the constant background phase shift of the electronics. Using measurements of the phase of the electroclinic effect they were able to show that Γ_A^{-1} obeys an Arrhenius law with an exponent U/k of 6000 K. However, they did not determine Γ_A^{-1} in absolute units, that is, they did not determine Γ_0. Furthermore, they found $\gamma = 1{\cdot}13 \pm 0{\cdot}06$. This result can be explained if an error of 10–20 mK in the determination of T_c has been made. We decided therefore to replace equation (5) with

$$\ln\tan(\delta_b - \delta) = \frac{U}{kT} + \ln\frac{\omega\Gamma_0}{\alpha T_c} - \ln\left(\frac{T - T_c}{T_c}\right), \tag{6}$$

where we took $A = \alpha(T - T_c)$ [6] and the term $(K_3 - \varepsilon\mu^2)q_0^2$ has been neglected. In order to obtain Γ_0 from equation (6) we first determined α from equations (3). The ferroelectric liquid crystal sample was oriented in a magnetic field of 6·3 T and the dielectric constant was measured parallel to the smectic layers. Figure 1 shows the critical behaviour of the dielectric strength of the soft mode in the S_A phase. The dielectric strength $\Delta\varepsilon$ ($= \varepsilon_0 - \varepsilon_\infty$) was determined from the measurement of the dielectric susceptibility at 30 Hz. The high frequency dielectric constant ε_∞ of 4·46 was taken [9, 10] from the measurement of the complex dielectric susceptibility as a function of frequency in the S_C^* phase.

Figure 2 shows the critical behaviour of the electroclinic coefficient which was determined from the measurement of the tilt angle versus the electric field. The tilt angle was measured by the conventional crossed polarizer method. From the slopes

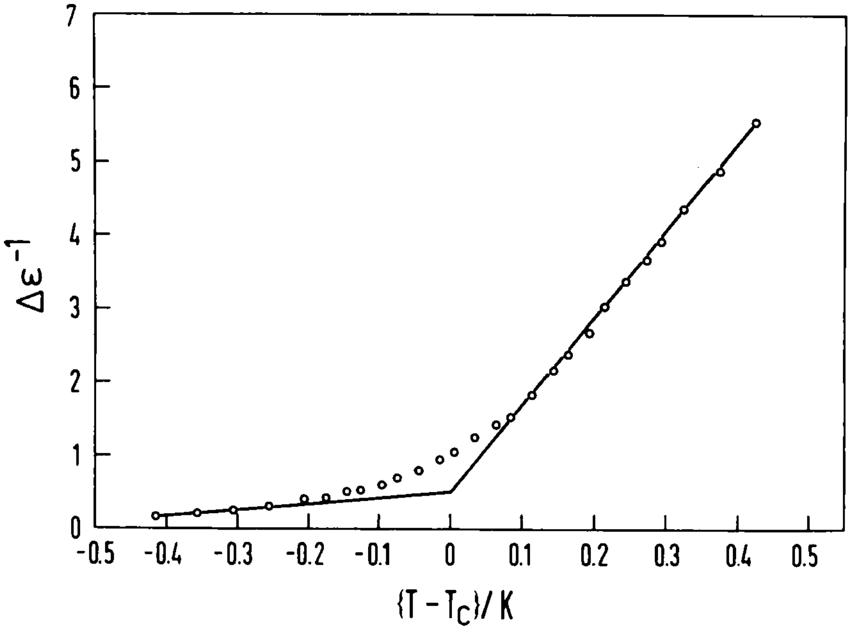

Figure 1. Reciprocal dielectric strength as a function of temperature in DOBAMBC.

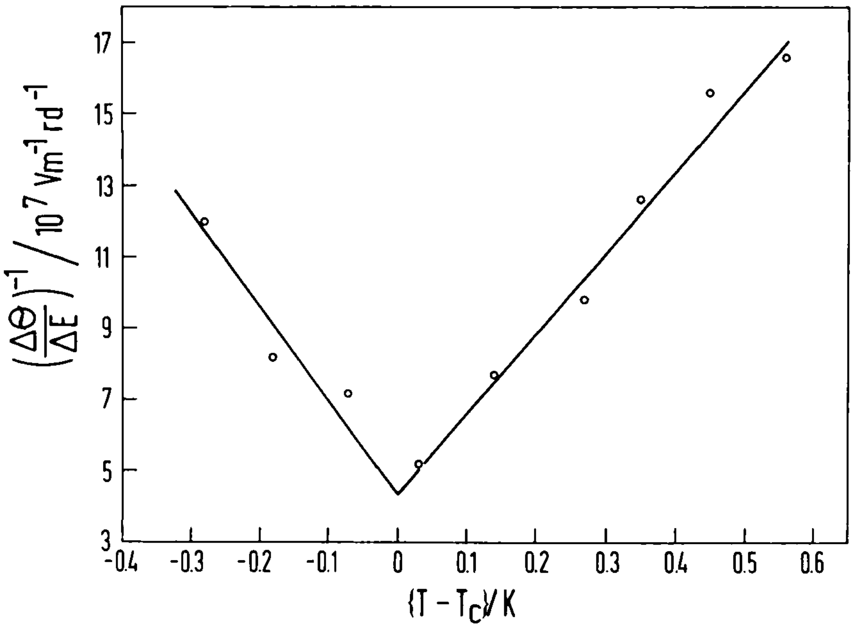

Figure 2. Electroclinic effect as a function of temperature close to the S_C^*–S_A phase transition.

of the straight lines in figures 1 and 2 we find that εC is $1{\cdot}66 \times 10^{-4}$ As/m^2 (1 ± 0·18) and that α is $3{\cdot}7 \times 10^4$ N/K m^2 (1 ± 0·19). The value of α is very close to the value obtained before [11]. Given α equation (6) allows us to evaluate Γ_0 using the experimental results of Garoff *et al.* [7, 8]. Using the values U/k of 6000 K and $T_c \sim 95°$C [7, 8] we extrapolate Γ_A^{-1} to T_c to find the value 0·041 kg/ms (1 ± 0·24). We compare this with the value for Γ_G^{-1} obtained for DOBAMBC in the S_C^* phase by a dielectric method [9], where we found the activation energy U/k of 5900 K. Extrapolation of Γ_G^{-1} to T_c gives Γ_G^{-1} as 0·038 kg/ms, in agreement with Γ_A^{-1} to within the experimental error. As the activation energy is the same for both rotational viscosities they can be expressed in the same form $\Gamma_0 \exp(U/kT)$. From equation (2) the previously neglected term $(K_3 - \varepsilon\mu^2)q_0^2$ has also been determined. Since $f_A = f_G$ at T_c we substitute the known value for f_G at T_c [9]. The value obtained for $(K_3 - \varepsilon\mu^2)q_0^2$ of 100 N/m^2 is close to the value for $K_3 q_0^2$ of 150 N/m^2 obtained from previous results [9]. Thus with the analysis of two experiments we have shown that as well as the relaxation frequencies of the soft mode and Goldstone mode the rotational viscosities at the S_C^* to S_A phase transition are degenerate. This is in agreement with the predictions of the Žekš [6] model.

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