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Dielectric method for determining the rotational viscosity in thick samples of ferroelectric chiral smectic-C^* liquid crystals

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An alternative method, based on dielectric measurements on thick samples, for determining the Goldstone-mode rotational viscosity and the twist elastic constant of ferroelectric, chiral smectic-C^* liquid crystals is presented. By the use of this method the temperature dependence of these quantities for *p*-decyloxibenzilidine-*p*′-amino-1-methylpropyl-cinamate (DOBA-1-MPC) and *p*-*n*-decyloxybenzylidine-*p*-amino-2-methyl-butyl-cinamate (DOBAMBC) has been determined. An Arrhenius-type behavior of the Goldstone-mode rotational viscosity was obtained with activation energies $\mu=0.33$ eV for DOBA-1-MPC and $\mu=0.51$ eV for DOBAMBC.

I. INTRODUCTION

Ferroelectric, chiral smectic-C^* (Sm-C^*) liquid crystals[1] have attracted considerable attention during the past decade and much effort has been devoted to investigating these systems from a fundamental as well as from an applied point of view. In contrast to what is the case in thick cells, where the ferroelectric liquid crystals form a helicoidal structure, in thin cells they order homogeneously with possible bistability of two optically different states and with a fast, electric-field-induced transition between these two states.[2] The idea that this switching could be used to develop fast electro-optic devices is one reason that the interest in the subject of ferroelectric liquid crystals has increased considerably during the last few years. One important physical parameter that strongly influences the switching time between the two homogeneously ordered states is the so-called Goldstone-mode rotational viscosity. In this paper we will discuss the concept of rotational viscosities of the Sm-C^* phase and introduce a new method of measuring the Goldstone-mode rotational viscosity based on dielectric measurements.

Generally, for the Sm-C^* phase, we have to introduce[3] two rotational viscosities which we denote γ_G and γ_S. The first one, γ_G, is related to rotations along the smectic cone (the Goldstone mode), while γ_S is related to rotations in which the tilt of the molecules is changed (the soft mode). The rotational viscosity γ_G is the one of the two viscosities which controls the switching time in an electro-optic device. This viscosity is usually determined using thin samples by optical switching-time measurements[4,5] or by the polarization reversal current technique.[5-7] But the question remains whether the viscosity determined on thin, homogeneously ordered samples corresponds to the bulk viscosity of the system or if it is influenced by boundary effects. In the smectic-A (Sm-A) phase only soft-mode deformations are present and consequently we only expect one rotational viscosity $\gamma_{S,A}$ in this case. As rotations of the direct or along the smectic cone and rotations for which the tilt changes its magnitude are degenerate at T_c, we expect[3] that the three viscosities γ_G, γ_S, and $\gamma_{S,A}$ are degenerate at T_c.

In the absence of electric fields, in the Sm-C^* phase the molecules are tilted from the normal to the smectic layers and the direction of the tilt precesses as one goes from one smectic layer to another (z axis) forming a helicoidal structure. The in-plane spontaneous polarization is locally perpendicular to the tilt, thus also forming a helix. The macroscopic polarization of the system therefore equals zero. When an electric field E is applied in a direction perpendicular (we chose this direction to be along the y axis) to the helical axis, it deforms the helix in two ways, changing the magnitude as well as the direction of the tilt. The dielectric response of the Sm-C^* phase therefore consists of two contributions.[8,9] First, there is the soft-mode part, which has a large characteristic frequency and which corresponds to changes of the magnitude of the tilt. This part is small and is appreciable only close to the transition temperature T_c between the Sm-C^* and Sm-A phases. Secondly, there is the Goldstone-mode part, which has a low characteristic frequency and which corresponds to changes in the tilt direction. This response is much larger than the soft-mode response, except very close to T_c in the Sm-C^* phase. By measuring the frequency dependence of the complex dielectric susceptibility, these two contributions can be separated[8,9] from each other.

In this paper we will demonstrate how the rotational viscosity γ_G can be determined from measurements on bulk samples (i.e., samples which are thick enough to develop an unperturbed helicoidal structure) of the complex dielectric constant of the system, together with the measurements of the spontaneous polarization P and of the tilt θ. If the pitch of the system is also measured, we will show that we can also determine the twist elastic constant K_3 from the measurements. Further on, we will also discuss how the rotational viscosity $\gamma_{S,A}$ of the Sm-A phase can be determined from measurements of the complex dielectric constnat together with the measurements of the electroclinic[10-12] response of the system.

II. THE BALANCE OF TORQUE EQUATION: DEFINITION OF THE GOLDSTONE-MODE ROTATIONAL VISCOSITY

In the literature the electro-optical switching of the Sm-C^* phase is most often studied[4,5,13] by the use of an equation which has the structure

$$-\gamma_G(\sin^2\theta)\dot{\phi}+K_3(\sin^2\theta)\phi''-PE\sin\phi=0\ , \tag{1}$$

where ϕ is the phase of the order parameter and determines the tilt direction, while $\dot{\phi}$ denotes the time derivative and ϕ'' the second derivative with respect to the z coordinate. This is a balance of torque equation which simply expresses that at each moment the elastic, rotational, and electric torques acting on the molecules balance each other. In order to demonstrate in which way the final appearance of this balance of torque equation depends on how we are modeling the system, we will formulate our approach in terms of generalized forces and fluxes. In the case which we are studying we have a system with one degree of freedom (we assume the tilt of the molecules to be fixed) which we describe by a generalized coordinate $X(z)$. The elastic torque Γ^{el} is then a generalized force which is related[3] to the elastic free-energy density $g(X,X')$ as

$$\Gamma^{\text{el}}=-\left[\frac{\partial g}{\partial X}-\frac{d}{dz}\frac{\partial g}{\partial X'}\right]\ . \tag{2}$$

In Eq. (2) a prime denotes the derivative with respect to the z coordinate. Further on, the rotational torque Γ^r is related to the generalized flux $\dot{X}$ by a generalized viscosity coefficient γ as

$$\Gamma^r=-\gamma\dot{X}\ , \tag{3}$$

where a dot denotes the derivative with respect to time. Depending on how g and X are chosen, we will arrive at a dynamic equation with the structure of Eq. (1) but with different definitions of the rotational viscosity γ_G and of the elastic constant K_3.

To demonstrate how Eq. (1) is derived, we proceed as follows: As the generalized coordinate X we chose the physical displacement $\phi\sin\theta$ of the molecules under a rotation ϕ,

$$X=\phi\sin\theta\ . \tag{4}$$

For g, which here shall denote the sum of the elastic and electric free-energy density of the system, we chose the simplest form appropriate for the analysis of the present problem,[14,15]

$$g(\phi,\phi')=\tfrac{1}{2}K_3\left[\frac{d\phi}{dz}-\frac{\Lambda}{K_3}\right]^2\sin^2\theta-PE\cos\phi\ . \tag{5}$$

Here E is a time-dependent electric field applied at an angle ϕ to the polarization vector, while Λ is the coefficient of the Lifschitz term which exists because of the chiral symmetry of the molecules. From Eqs. (2)–(5) we now get the elastic (Γ^{el}), electric (Γ^{ε}), and rotational (Γ^r) torques as

$$\Gamma^{\text{el}}+\Gamma^{\varepsilon}=-\frac{1}{\sin\theta}\left[\frac{\partial g}{\partial\phi}-\frac{d}{dz}\frac{\partial g}{\partial\phi'}\right]$$

$$=K_3\sin\theta\phi''-\frac{PE}{\sin\theta}\sin\phi\ , \tag{6}$$

$$\Gamma^r=-\gamma_G\dot{\phi}\sin\theta\ . \tag{7}$$

The equation governing the rotation of the director under the influence of the electric field and of the twist elasticity is now given by the balance of torque equation $\Gamma^{\text{el}}+\Gamma^{\varepsilon}+\Gamma^r=0$, which after some rearrangement will give Eq. (1). We thus see that the detailed form of Eq. (1) depends on three things: (a) The choice of $\phi\sin\theta$ as the generalized coordinate, (b) the assumption that the tilt angle θ is not influenced by the field, and (c) the choice of the specific form (5) of the free-energy density of the system. Other forms of the free-energy density would, of course, give other dynamic equations. However, as long as the free-energy density depends only on $\cos\phi$ (the electric-field dependent part in the case when only the ferroelectric coupling to the field is considered) and quadratically on the space derivative ϕ' (the usual form of an elastic energy), the structure of Eq. (1) would be unchanged. By choosing more complicated (and more realistic) models for the free-energy density and also taking changes of the magnitude of the tilt into account, we would only introduce a renormalized elastic constant $\tilde{K}_3$ and a renormalized Lifschitz coefficient $\tilde{\Lambda}$ into the model. The explicit form of $\tilde{K}_3$ depends on the theoretical model which is used in describing the system. For the classical Landau model[14,16] the renormalized elastic constant is given by

$$\tilde{K}_3=K_3-\varepsilon\mu^2\ . \tag{8}$$

Here ε is the high-frequency value of the dielectric susceptibility and μ represents the flexoelectric coupling between tilt and polarization. Within this model $\tilde{K}_3$ is temperature independent. For the generalized Landau model one obtains[17]

$$\tilde{K}_3=K_3-\frac{\varepsilon\mu^2}{1-\varepsilon\Omega\theta^2+3\eta\varepsilon P^2}\ , \tag{9}$$

where Ω and η are material parameters of the higher-order coupling terms which are introduced[15] in this model. This form of $\tilde{K}_3$ contains a temperature dependence which, depending on the magnitude of the ingoing parameters, might or might not be negligible. We have intestigated the temperature dependence of $\tilde{K}_3$ according to Eq. (9) and found realistic values of the material parameters which give a rather weak temperature dependence of $\tilde{K}_3$ over most of the Sm-C^* phase with a rather pronounced drop occurring in the region a few degrees below T_c.

Equation (1) is often found written in a slightly different form[13,18]

$$-\gamma_G^{\text{eff}}\dot{\phi}+K_3^{\text{eff}}\phi''-PE\sin\phi=0 . \tag{10}$$

This equation is obtained if we do not define the rotational and elastic torques with respect to the true physical displacement $\phi\sin\theta$ but instead with respect to the angle ϕ, and if we include the $\sin^2\theta$ dependence into the definition of the twist elastic constant K_3. Thus, if Eq. (10) is used to analyze an electro-optical switching experiment in order to obtain the Goldstone-mode rotational viscosity or the twist elastic constant, we see by comparing Eqs. (1) and (10) that the quantities determined are what can be called an effective rotational viscosity γ_G^{eff} and an effective twist elastic constant K_3^{eff} defined as

$$\gamma_G^{\text{eff}}=\gamma_G\sin^2\theta , \tag{11}$$

$$K_3^{\text{eff}}=K_3\sin^2\theta . \tag{12}$$

We will not, however, promote this definition of effective material parameters because it is γ_G and K_3 which are related to the true physical torque which acts upon the molecules. Thus, when we are studying the temperature dependence of the effective material parameters, we must be aware of the fact that with this definition we have imposed an extra temperature dependence through the factor $\sin^2\theta$.

III. THEORY

A. The Goldstone-mode rotational viscosity of the smectic-C^* phase

We shall now calculate the Goldstone-mode part of the dielectric susceptibility with Eq. (1) as a basis. We assume that the electric field has a time dependence $E=E_0e^{j\omega t}$ where, as we are only interested in the linear response of the system, we consider E_0 to be a small quantity. Now expanding $\phi(z)$ to first order, $\phi(z)=qz+\delta\phi$, $q=\Lambda/K_3=2\pi/p$ being the wave vector of the unperturbed pitch and $\delta\phi$ being of the same order as E_0. By making the ansatz

$$\delta\phi=\delta\phi_0\sin(qz)e^{j\omega t}$$

and by linearizing Eq. (1), we arrive at an equation determining $\delta\phi_0$:

$$j\omega\gamma_G(\sin^2\theta)\delta\phi_0+K_3(\sin^2\theta)q^2\delta\phi_0+PE_0=0 . \tag{13}$$

Assuming $\sin\theta\sim\theta$, this equation implies

$$\delta\phi_0=-\frac{E_0P}{K_3q^2\theta^2}\frac{1}{1+j\omega\gamma_G/K_3q^2} . \tag{14}$$

The induced polarization per unit volume of the sample is easily calculated as

$$\langle P_y\rangle=P\langle\cos(qz+\delta\phi)\rangle=-\tfrac{1}{2}P\delta\phi_0e^{j\omega t} . \tag{15}$$

Substituting Eq. (14) into Eq. (15) now gives

$$\langle P_y\rangle=\frac{E_0P^2}{2K_3q^2\theta^2}\frac{e^{j\omega t}}{1+j\omega\gamma_G/K_3q^2} . \tag{16}$$

The dielectric susceptibility is defined as

$$\chi=\lim_{E\to0}(\langle P_y\rangle/E) .$$

Assuming that χ can be written as

$$\chi=\varepsilon_0\Delta\varepsilon_G/(1+j\omega\tau) ,$$

where $\Delta\varepsilon_G$ is the dielectric strength of the Goldstone-mode part of the dielectric constant and $\tau=1/2\pi f_G$ is the corresponding relaxation time, we thus get the final result (ε_0 being the permittivity of free space)

$$\varepsilon_0\Delta\varepsilon_G=\frac{P^2}{2K_3q^2\theta^2} , \tag{17}$$

$$f_G=\frac{K_3q^2}{2\pi\gamma_G} . \tag{18}$$

From these two equations[19,20] we get the Goldstone-mode rotational viscosity as

$$\gamma_G=\frac{1}{4\pi\varepsilon_0}\frac{P^2}{\theta^2\Delta\varepsilon_Gf_G} . \tag{19}$$

Thus we can evaluate the Goldstone-mode rotational viscosity of the Sm-C^* phase from the measured values of the polarization, tilt, dielectric strength of the Goldstone mode, and its corresponding relaxation frequency. If the pitch of the system is measured as well, we see from Eq. (17) that the elastic constant K_3 can also be evaluated from the experiment. We would also like to stress that Eqs. (17)–(19) are generally valid as long as the ϕ fluctuations represent an eigenmode of the dielectric susceptibility. Thus these equations are also applicable in the vicinity of T_c, where tilt fluctuations also give an appreciable contribution to the dielectric susceptibility of the system.

B. Estimation of nonlinear effects in the dielectric response close to T_c

Dielectric measurements assume that the testing field E_0 is so small that the induced change of the phase angle ϕ allows the linearization of Eq. (1) from which Eq. (14) is derived. The amplitude of $\delta\phi$ can be expressed from Eq. (14) and (17) for $\omega=0$ as

$$|\delta\phi_0(\omega=0)|\equiv\Delta\phi_0=\frac{PE_0}{K_3q^2\theta^2}=\frac{2\varepsilon_0\Delta\varepsilon_GE_0}{P} . \tag{20}$$

As both P and θ go to zero at T_c and do so in such a way that the ratio P/θ is finite,[21] we see that close to T_c, $\Delta\phi_0$ will be very large as long as a finite testing field E_0 is used in the experiment. Thus we must be aware of the fact that the linearization of Eq. (1) is not valid in a temperature interval just below T_c that the relation (17) in this region will suffer from a considerable error. In order to estimate an upper limit of $\Delta\phi_0$ beyond which nonlinear effects start to influence the response, we write down the expression[22,23] of the critical field E_c, for which the helix of the system will be completely unwound,

$$E_c=\frac{\pi^2}{16}\frac{K_3\theta^2q^2}{P} . \tag{21}$$

From Eqs. (20) and (21) we derive

$$\Delta\phi_0=\frac{\pi^2}{16}\frac{E_0}{E_c}\simeq 0.6\frac{E_0}{E_c} \ . \tag{22}$$

From Eq. (22) we can get a quantitative measure of how large $\Delta\phi_0$ is allowed to be if we want to remain in the regime of linear response. If the ratio E_0/E_c is too large, it is obvious that we are close to unwinding the helix completely and the response of the system is no longer linear. By solving Eq. (1) exactly in the stationary case, Urbanc *et al.*,[24] have shown that if $E_0/E_c\leq 0.5$, the response of the system is to a good degree of accuracy still in the linear regime. Thus we see from Eq. (22) that if $\Delta\phi_0$ is less than approximately 0.3 rad $\simeq 20°$, we can rely on the data we obtain in a dielectric measurement. In this way we can always check whether we are in the linear regime or not when performing a dielectric experiment by the use of Eq. (20).

C. The soft mode rotational viscosity of the smectic-*A* phase

In this section we will show how the soft-mode rotational viscosity $\gamma_{S,A}$ of the Sm-*A* phase can be determined from dielectric measurements if we also perform a study of the electroclinic effect[10–12] of the system. The expressions of the soft-mode dielectric strength $\Delta\varepsilon_A$ and the relaxation frequency f_A of the Sm-*A* phase have been derived previously in the literature:[9,16,17]

$$f_A=\frac{1}{2\pi\gamma_{S,A}}[\alpha(T-T_c)+(K_3-\varepsilon\mu^2)q_0^2] \ , \tag{23}$$

$$\varepsilon_0\Delta\varepsilon_A=\frac{\varepsilon^2C^2}{\alpha(T-T_c)+(K_3-\varepsilon\mu^2)q_0^2} \ . \tag{24}$$

In these equations α is the usual coefficient contained in the temperature-dependent term of the Landau expansion of the free-energy density, while μ and C are the coefficients of the flexoelectric and piezoelectric bilinear coupling, respectively. The constant ε represents the dielectric constant of the system in the high-frequency limit (the polarization-mode[17] dielectric strength), while q_0 is the wave vector of the pitch at T_c. From Eqs. (23) and (24) we can derive the following expression:

$$\gamma_{S,A}=\frac{\varepsilon^2C^2}{2\pi\varepsilon_0 f_A\Delta\varepsilon_A} \ . \tag{25}$$

We thus see that we cannot determine $\gamma_{S,A}$ from dielectric measurements unless we know the product of the Landau parameters ε and C. This product can, however, be eliminated from Eq. (25) by the use of the electroclinic effect. Applying an electric field of magnitude E_0 in the Sm-*A* phase will induce a tilt θ_0, the magnitude of which is given by[25]

$$\theta_0=\frac{\varepsilon C}{\alpha(T-T_c)+(K_3-\varepsilon\mu^2)q_0^2}E_0=kE_0 \ . \tag{26}$$

In Eq. (26) we have defined what we will denote as the electroclinic coefficient k, which is the slope of the straight line we obtain by plotting the induced tilt as a function of the applied field at a given temperature. From Eqs. (24) and (26) we obtain

$$\varepsilon C=\frac{\varepsilon_0\Delta\varepsilon_A}{k} \ . \tag{27}$$

Introducing Eq. (27) into Eq. (25), we thus get

$$\gamma_{S,A}=\frac{\varepsilon_0\Delta\varepsilon_A}{2\pi f_A k^2} \ . \tag{28}$$

From Eq. (28) we see that we can evaluate the rotational viscosity $\gamma_{S,A}$ by measuring the complex dielectric constant of the Sm-*A* phase and the electroclinic coefficient. Such an evaluation has recently been presented by Gouda *et al.*[26]

IV. EXPERIMENTAL RESULTS

The dielectric method for determining the rotational viscosity has been applied to two ferroelectric liquid crystals:[19,20] DOBA-1-MPC (*p*-decyloxibenzilidene-*p*′-amino-1-methylpropyl-cinamate) and DOBAMBC (*p*-*n*-decyloxybenzylidene-*p*-amino-2-methyl-butyl-cinamate). The temperature dependence of the spontaneous polarization, the tilt, the pitch, and the frequency dependence of the complex dielectric constant was obtained using a 50-μm- thick sample in the case of DOBA-1-MPC and a 20μm-thick sample in the case of DOBAMBC. The amplitude of the voltage applied over the cells in the dielectric measurements was in both cases 1.4 V.

The tilt angle was determined by the conventional crossed polarizer method at a constant field, while the

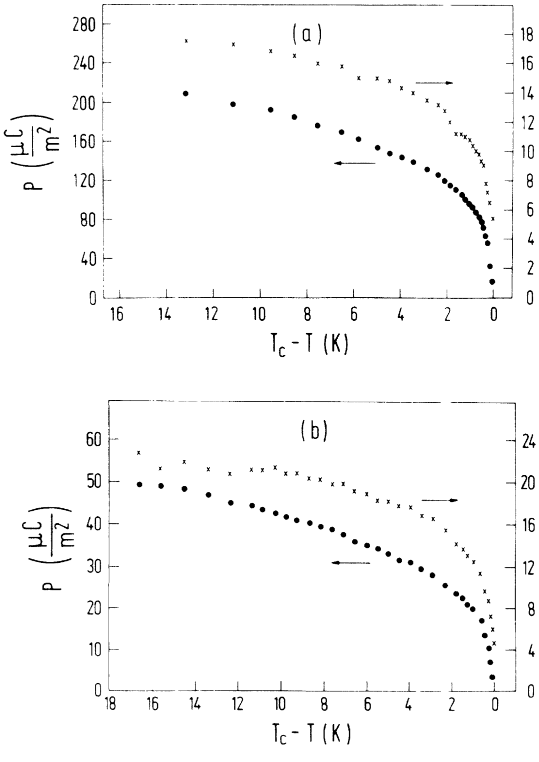

FIG. 1. Experimentally determined tilt (×) and polarization (●) for (a) DOBA-1-MPC and (b) DOBAMBC.

spontaneous polarization was measured simultaneously with a Sawyer-Tower bridge at 70 Hz. Both methods are dynamic, but allow the determination of the static quantities P and θ. The spontaneous polarization is determined by extrapolating the saturated part of the hysteresis loop to zero field. The tilt for DOBA-1-MPC and DOBAMBC was determined at constant electric field $E = 9.4$ kV/cm and $E = 23.7$ kV/cm, respectively, and is equal to the spontaneous tilt except in the region $T_c - T \leq 0.5$ K because of the electroclinic[10-12] effect. In Fig. 1 the temperature dependences of the obtained tilt and polarization are shown.

The pitch was determined by the use of a polarizing microscope. The resulting temperature dependence for the two samples is shown in Fig. 2.

The frequency and temperature dependences of the complex dielectric constant were measured in a frequency range between 30 Hz and 15 kHz with a Hewlett Packard 4192 LF impedance analyzer. The experimental details and the analysis of the data for this type of experiment have been discussed by us elsewhere.[8,9] From the data, the temperature dependences of the relaxation frequency and of the dielectric strength of the Goldstone mode were determined. In Fig. 3 the results for the Goldstone mode are shown. The shape of the maximum of the dielectric strength which appears approximately 1.0 K below T_c depends strongly on the quality of the sample and on the temperature gradient in the sample. For not very well aligned samples and for not small enough temperature gradients, the maximum disappears.

From the experimental data presented in Figs. 1–3 we have evaluated γ_G and K_3 by using Eqs. (19) and (17), respectively. The temperature dependences of these quantities thus obtained are shown in Figs. 4 and 5. As a comparison, in Fig. 4(a) we have also shown the values of γ_G for DOBA-1-MPC evaluated from experimental values of γ_{eff} obtained by Kimura *et al.*[6] by the polarization reversal current technique. In the evaluation [Eq. (11)] the experimental values for the tilt [Fig. 1(a)] were used. We notice that the results of our method and theirs are in good agreement with each other, except for the small bump which our data exhibit close to T_c. We believe that the existence of this bump is mainly related to the difficulties of determining the maximum of $\Delta\varepsilon_G$ accurately. The bump close to T_c in the determined temperature dependence of K_3 which we notice in Fig. 5(a) is of the

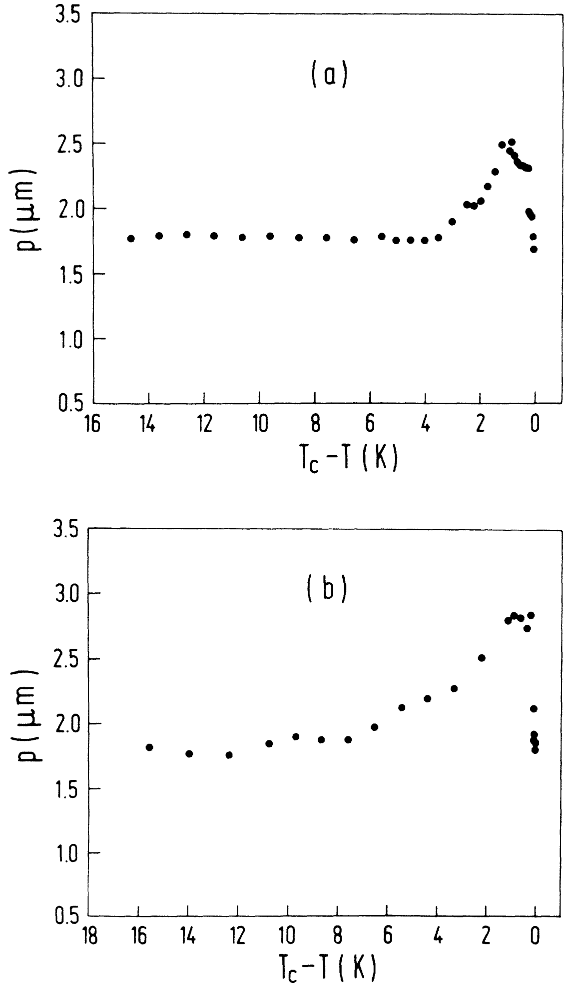

FIG. 2. Experimentally determined pitch for (a) DOBA-1-MPC and for (b) DOBAMBC.

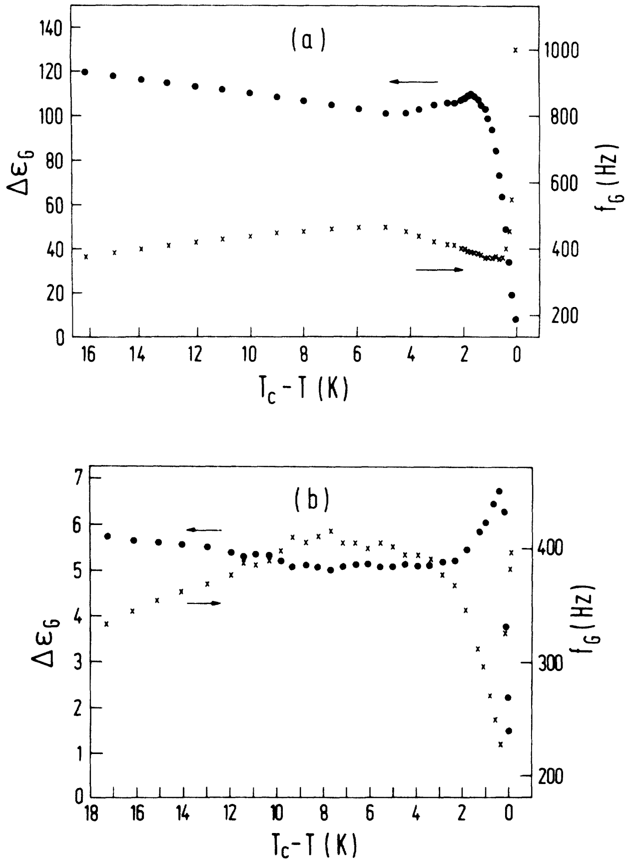

FIG. 3. Experimentally determined values of the Goldstone-mode dielectric strength (●) and of the corresponding relaxation frequency (×) for (a) DOBA-1-MPC and for (b) DOBAMBC.

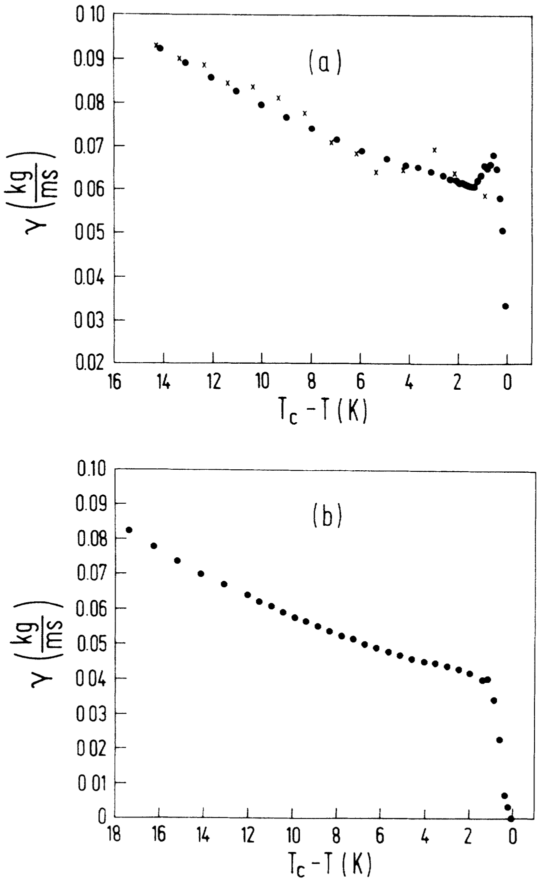

FIG. 4. Experimentally determined Goldstone-mode rotational viscosity for (a) DOBA-1-MPC and for (b) DOBAMBC. The crosses in (a) represents the measurements of γ_G performed by Kimura *et al.*[6]

same origin.

As was discussed in Sec. III B, it is important when performing a dielectric experiment to check that the applied electric field is small compared with the critical unwinding field of the system. By calculating $\Delta\phi_0$ by the use of Eq. (20), we can determine the ratio E_0/E_c from Eq. (22). In the case of DOBA-1-MPC, this ratio is everywhere small enough not to introduce substantial errors in the measured $\Delta\varepsilon_G$. For DOBAMBC, where a thinner cell was used (i.e., a larger electric field), we have probably overestimated $\Delta\varepsilon_G$ in the temperature interval $T_c - T \leq 0.5$ K.

V. DISCUSSION

The main aim of this paper is to show how it is possible to determine experimentally the Goldstone-mode rotational viscosity γ_G for a ferroelectric Sm-C^* liquid crystal from the measurements of the complex dielectric constant in combination with the measurements of the spontaneous polarization and the tilt. The obtained temperature dependence of the two systems is shown in Fig. 4, where available data from another experimental method are also shown for comparison.

In Fig. 6 we show the Arrhenius plots, i.e., $\ln\gamma_G$ versus $1/T$, of the obtained rotational viscosities. The tempera-

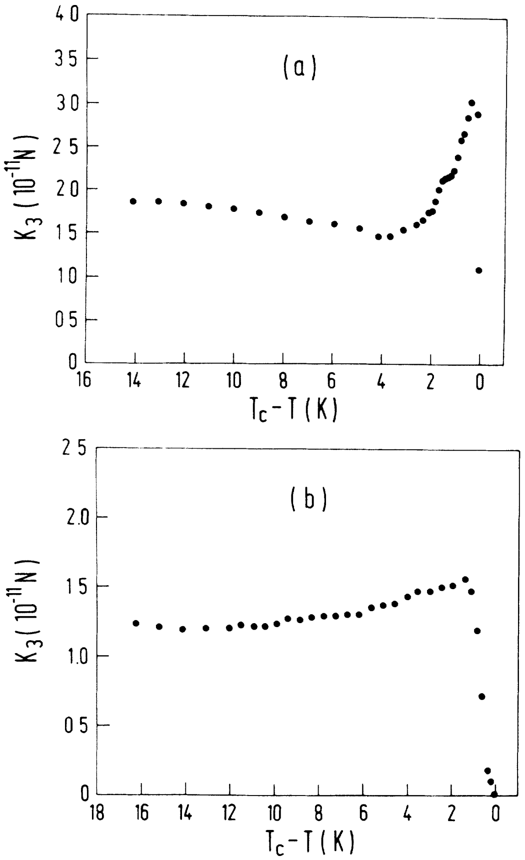

FIG. 5. Experimentally determined elastic constant for (a) DOBA-1-MPC and for (b) DOBAMBC.

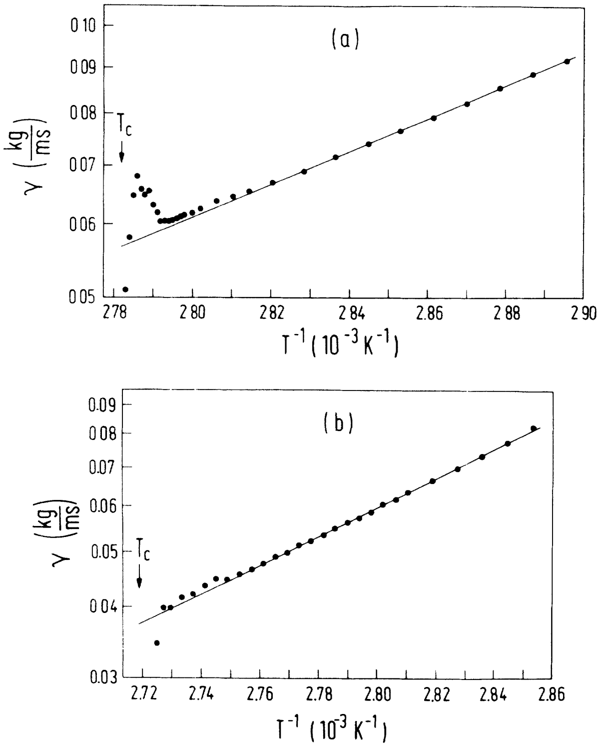

FIG. 6. Arrhenius plot of the experimentally determined Goldstone-mode rotational viscosity for (a) DOBA-1-MPC and for (b) DOBAMC.

ture dependence of γ_G obeys the Arrhenius law $\gamma_G \sim e^{\mu/kT}$ fairly well with activation energies $\mu=0.33$ eV for DOBA-1-MPC and $\mu=0.51$ eV for DOBAMBC, respectively. These evaluated values of the activation energies compare well with the values $\mu \sim 0.35-0.45$ eV obtained by Escher *et al.*[7] for some other compounds. The activation energy for DOBAMBC is comparable with the value $\mu=0.52$ eV obtained for DOBAMBC by Garoff and Meyer[10] for the activation energy of the soft-mode rotational viscosity of the Sm-A phase. Close to T_c we observe in both liquid crystals the deviations from the Arrhenius law in rotational viscosity. These deviations can be attributed to various effects, which influence the results of measurements close to T_c:

(i) All the measured quantities ($\Delta\varepsilon_G$, f_G, P, θ, p) are strongly temperature dependent and therefore less accurately determined close to T_c.

(ii) The dielectric measurements were performed with an ac measuring field which becomes comparable to the unwinding field close to T_c leading to large values for $\Delta\varepsilon_G$.

(iii) Quality of samples and finite temperature gradients influence the shape of the maximum in $\Delta\varepsilon_G$.

(iv) The constant field values were used in the tilt measurements.

The elastic constant K_3 for DOBA-1-MPC and DOBAMBC is approximately temperature independent except for $T_c - T \leq 1.5$ K. The measured temperature dependence in this temperature range is less accurate because of the effects mentioned above. The drop observed in the elastic constant of DOBAMBC close to T_c could be explained by the renormalization of the elastic constant following from the generalized model of ferroelectric liquid crystals [Eq. (9)].

In conclusion, we summarize that we have presented an alternative method of determining the Goldstone-mode rotational viscosity of the Sm-C^* phase based on dielectric measurements performed on thick samples. Such a method has the advantage that in a dielectric experiment only small oscillations of the director are excited. Thus we avoid problems such as disclination formation and backflow effects. Both the compounds for which we determined γ_G exhibit an Arrhenius type behavior for the rotational viscosity with activation energies which are in agreement with what other authors have obtained by other methods. As the soft-mode rotational viscosity in DOBAMBC was shown[10] to obey the Arrhenius law in the smectic-A phase also very close to T_c with the same activation energy as the one determined in this work for the Goldstone-mode rotational viscosity γ_G in the smectic-C^* phase, we believe that the Arrhenius law is valid for γ_G in the whole Sm-C^* phase and that the two viscosities match continuously at T_c.

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[1] R. B. Meyer, L. Liebert, L. Strzelecki, and P. Keller, J. Phys. (Paris) Lett. **36**, L69 (1975).

[2] N. A. Clark and S. T. Lagerwall, Appl. Phys. Lett. **36**, 899 (1980).

[3] T. Carlsson and B. Žekš, Liq. Cryst. **5**, 359 (1989).

[4] J. Z. Xue, M. A. Handschy, and N. A. Clark, Ferroelectrics **73**, 305 (1987).

[5] K. Sharp, K. Flatischler, and S. T. Lagerwall, Ferroelectrics **84**, 183 (1988).

[6] S. Kimura, S. Nishiyama, Y. Ouchi, H. Takezoe, and A. Fukuda, Jpn. J. Appl. Phys. **26**, L255 (1987).

[7] C. Escher, T. Geelhaar, and E. Böhm, Liq. Cryst. **3**, 469 (1988).

[8] A. Levstik, T. Carlsson, C. Filipič, I. Levstik, and B. Žekš, Phys. Rev. A **35**, 3527 (1987).

[9] C. Filipič, T. Carlsson, A. Levstik, B. Žekš, R. Blinc, F. Gouda, S. T. Lagerwall, and K. Skarp, Phys. Rev. A **38**, 5833 (1989).

[10] S. Garoff and R. B. Meyer, Phys. Rev. Lett. **38**, 848 (1977).

[11] C. H. Bahr and G. Heppke, Liq. Cryst. **2**, 285 (1987).

[12] G. Andersson, I. Dahl, P. Keller, W. Kuczynski, S. T. Lagerwall, K. Skarp, and B. Stebler, Appl. Phys. Lett. **51**, 640 (1987).

[13] M. Handschy and N. A. Clark, Ferroelectrics **59**, 69 (1984).

[14] S. A. Pikin and V. L. Indenbom, Usp. Fiz. Nauk. **125**, 251 (1978). [Sov. Phys.—Usp. **21**, 487 (1987)].

[15] T. Carlsson, B. Žekš, A. Levstik, C. Filipič, I. Levstik, and R. Blinc, Phys. Rev. A **36**, 1484 (1987).

[16] Ph. Martinot-Lagarde and G. Durand, J. Phys. (Paris) Lett. **41**, L43 (1980).

[17] T. Carlsson, B. Žekš, C. Filipič, and A. Levstik, Phys. Rev. A (to be published).

[18] W. Kuczynski, Ber. Bunsenges. Phys. Chem. **85**, 234 (1981).

[19] A. Levstik, Z. Kutnjak, C. Filipič, I. Levstik, B. Žekš, and T. Carlsson, *Second International Conference on Ferroelectric Liquid Crystals, Göteborg, 1989* [Ferroelectrics (to be published)].

[20] A. Levstik, Z. Kutnjak, I. Levstik, and B. Žekš, in *Proceedings of the Seventh International Meeting on Ferroelectricity, Saarbrücken, 1989* [Ferroelectrics (to be published)].

[21] S. Dumrongrattana and C. C. Huang, Phys. Rev. Lett. **56**, 464 (1986).

[22] S. Dumrongrattana and C. C. Huang, J. Phys. (Paris) **47**, 2117 (1986).

[23] B. Žekš and R. Blinc, in *Ferroelectric Liquid Crystals,* edited by J. W. Goodby (Gordon and Breach, New York, in press).

[24] B. Urbanc, B. Žekš, and T. Carlsson, in *Proceedings of the Second International Conference of Ferroelectric Liquid Crystals, Göteborg, 1989* [Ferroelectrics (to be published)].

[25] F. Gouda, G. Andersson, S. T. Lagerwall, K. Sharp, B. Stebler, T. Carlsson, B. Žekš, C. Filipič, and A. Levstik, Liq. Cryst. **6**, 219 (1989).

[26] F. Gouda K. Skarp, G. Andersson, I. Dahl, and S. T. Lagerwall, in *Proceedings of the Second International Conference on Ferroelectric Liquid Crystals, Göteborg, 1989* [Ferroelectrics (to be published)].