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COUPLING OF FERROELECTRIC MODES IN CHIRAL SMECTIC LIQUID CRYSTALS

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Abstract We applied an optical method to detect the soft and Goldstone modes in ferroelectric liquid crystals. The method allowed us to discover an additional mode appearing in both smectic A and smectic C* phases. Its relaxation time is independent of temperature and equals to about 60 μs. The third mode couples to the Goldstone mode in the smectic C* phase and to the soft mode in the smectic A phase.

INTRODUCTION

Electric field applied in the direction parallel to the smectic layer plane of a ferroelectric liquid crystal causes a change in its molecular arrangement. The director motion can be resolved into two components:

i) changes in the tilt plane position (i.e. Goldstone mode) occurring in the smectic C^* phase,

ii) changes in the value of the tilt angle (i.e. soft mode) occurring in both smectic C^* and smectic A phases.

To detect the above modes one usually applies the dielectric method (e.g.[1-3]). However, this method has some disadvantages. First, while measuring the electric permittivity one detects the ferroelectric component on a high level background produced by other mechanisms of electric polarization

(dipolar, electronic etc.). Further, the dielectric method is insufficiently sensitive, in most cases the soft mode component can be detected only in the vicinity (≅ 1K) of the phase transition[1,3]. Besides, the electric conductivity makes the measurements at low frequencies hard to perform.

The above limitations inclined us to apply an optical method of detection[4]. It consists in the registration of changes in intensity of light passing through the sample placed between crossed polarizers. These changes are caused by the field-induced reorientation and deformation of the indicatrix.

In the experiment, we applied ac electric field of low strength (10 - 10^4 V/cm) and frequency from 3Hz to 100kHz. The light modulation was registered with a photodiode and a lock-in amplifier. The SmC*-SmA phase transition temperature of the investigated 4-octyloxy 4-[(2-methylbutyloxy)carbonyl] phenylbenzoate was 30.5°C.

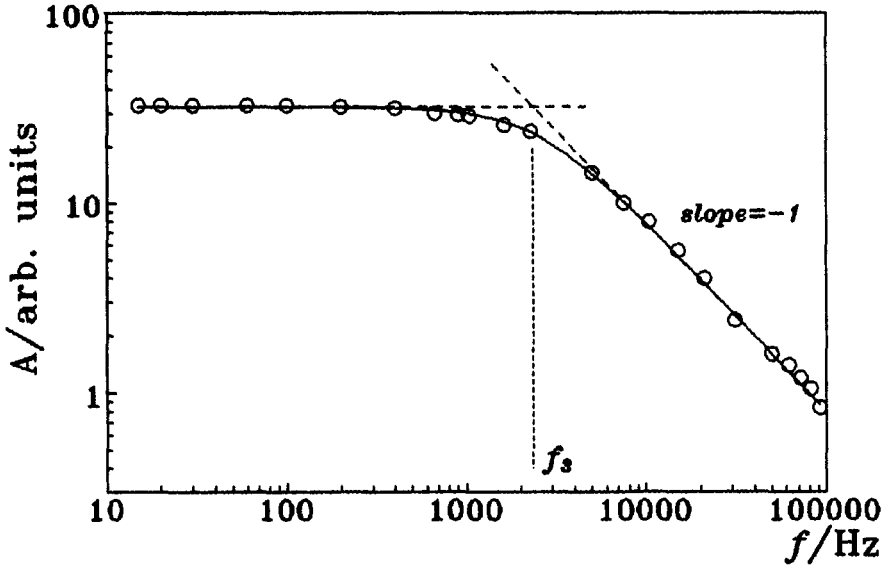

FIGURE 1. Typical dependence of the light modulation amplitude on frequency in the SmA phase. Circles represents the experimental points, line is calculated from equation (1) for τ = 60 μs.

RESULTS AND DISCUSSION

A typical dependence of the light modulation amplitude on frequency obtained in the SmA phase far away from the transition to SmC* is shown in Figure 1.

Dependence shown in Figure 1 is a curve typical for Debye relaxation with a single relaxation time[5]. The light modulation amplitude is described by:

$$A \sim (1 + \omega^2\tau^2)^{-1/2}, \qquad (1)$$

where ω is the angular frequency of the applied field, and τ is the relaxation time. The analogous dependence obtained in the SmC* phase has a different shape (Figure 2).

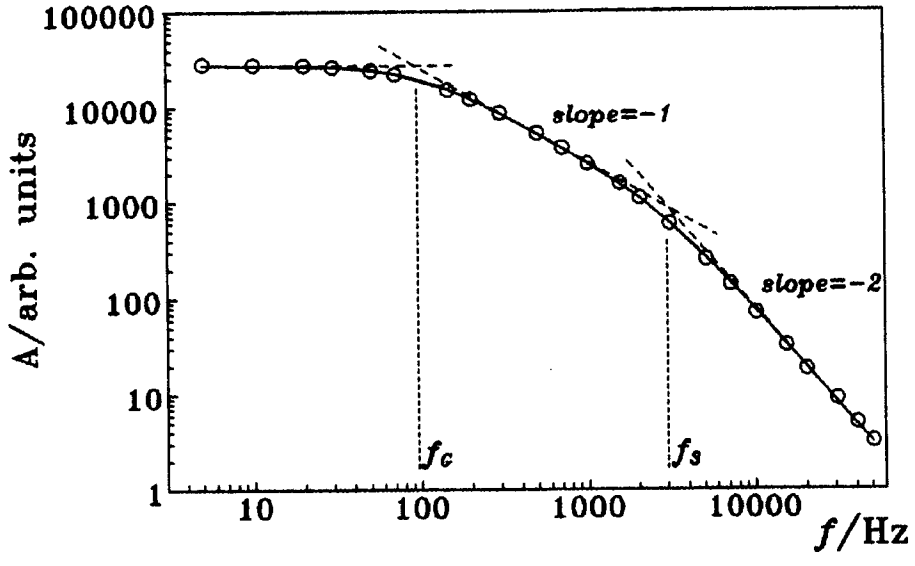

FIGURE 2. Typical dependence of the light modulation amplitude on frequency in the SmC* phase. Circles represent the experimental points, line is calculated from equation (2) for τ_1 = 1.6 ms and τ_2 = 63 μs.

Dependence in Figure 2 is composed of linear sections with distinctly various slopes. This shape cannot be explained by the existence of two or more independent vibrations with different relaxation times. The only possible explanation of such a behaviour is the presence of two coupled vibrations with relaxation times τ_1 and τ_2. In this case the modulation

amplitude is described by the following formula[4]:

$$A \sim (1 + \omega^2\tau_1^2)^{-1/2} \cdot (1 + \omega^2\tau_2^2)^{-1/2} \qquad (2)$$

Figure 2 shows excellent agreement of calculations performed according to formula (2) with the experimental data. The slow relaxation ($\tau_1 = 1/2\pi f_G \cong 1.6$ms) is assigned to the Goldstone mode. The faster relaxation ($\tau_2 = 1/2\pi f_3 \cong 60\mu$s) was not observed up to now with dielectric methods. It must be attributed to a new kind of motion. This third mode also couples to the soft mode in the SmA phase in the vicinity of transition to SmC* phase. Far away from the transition the relaxation frequency of the soft mode becomes very large exceeding the measurement range of our experiment (100 kHz). Then, only the third mode with the relaxation frequency f_3 of about 3 kHz is observed and the mode coupling effect is no longer visible (see Figure 1). The phase shift between the applied ac voltage and the optical response behaves as predicted by the coupled mode model (Figure 3).

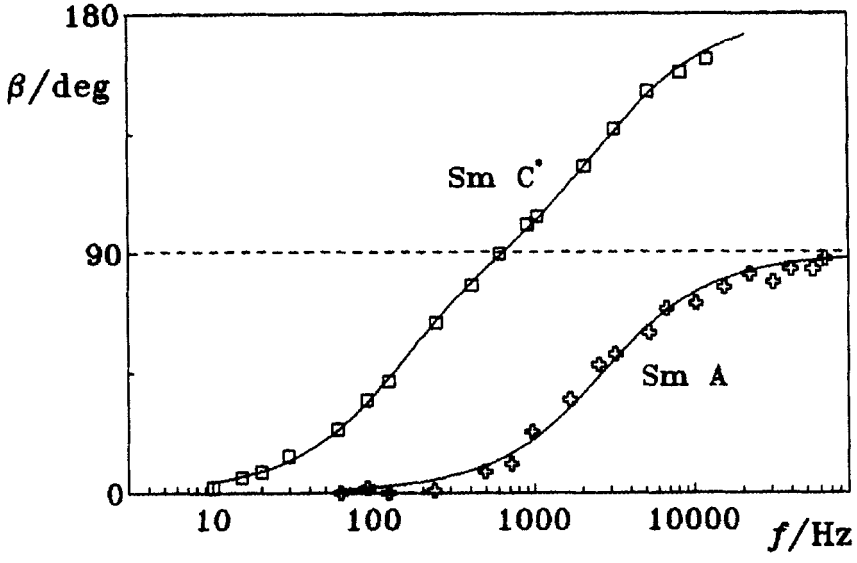

FIGURE 3. Phase lag of the optical response as a function of frequency. Squares and crosses represent the experimental data obtained in the SmC* and SmA phase, respectively. The solid lines are calculated from eqs. (4) and (3) for τ=60μs, τ_1=1.1ms and τ_2=57μs.

The phase shift β for a single mode changes in the range from 0 to 90^{o} and is described by the formula[5]:

$$tg\ \beta = -\omega\tau \tag{3}$$

For the two coupled modes β ranges from 0 to 180^{o} and is described by the formula[4]:

$$tg\ \beta = \frac{-\omega\ (\tau_1+\tau_2)}{1\ -\ \omega^2\tau_1\tau_2} \tag{4}$$

Figure 3 shows that the agreement of experimental data with the calculations performed according to formulae (3) and (4) is good. Thus, both dependences, of amplitude and phase shift, on frequency confirm the proposed idea of coupled modes.

A possible nature of the observed third mode is a separate question. In our opinion, the third mode is related to the field-induced changes in distribution of the azymuthal angles of molecules within a given smectic layer. In such a case the vibration would be controlled by elastic forces resulting from interactions inside the smectic layer, rather then by forces resulting from the interlayer interactions as in case of the Goldstone mode. To verify this hypothesis further experimental and theoretical studies are needed.

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