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Corrigendum

Theoretical investigation of helix distortion and dielectric spectrum of antiferroelectric liquid crystals

Deblal Das, Tanmoy Lahiri, Tapas Pal Majumder*

Department of Physics, University of Kalyani, Kalyani, Nadia 741235, West Bengal, India

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ABSTRACT

Landau–Ginzburg equation is utilized to investigate the dielectric spectrum of an antiferroelectric liquid crystal (AFLC). Two collective modes, one associated with the in-phase motion at the low frequency side and the other related with the anti-phase motion at the high frequency side, are discussed in detail utilizing the concept of bulk free energy of the material in terms of the pair interactions of two adjacent Smectic layers. The phenomenon of helix distortion related to the in-phase motion is observed theoretically based on the interlayer interactions of the AFLC system. The anti-phase mode related to the degree of the antiferroelectric ordering is also an important phenomenon depending on the layer-to-layer interactions. Lastly we have theoretically obtained the critical field for helix unwinding with the model proposed by de Gennes.

1. Introduction

Antiferroelectric liquid crystals (AFLC) [1–5] are a class of Smectic tilted phase that can exhibit a strong variation in the azimuthal component of the tilt angle from layer to layer forming a non-synclinic structure. The projection of the average layer molecular director on the layer is termed as the C director for an AFLC system. The anticlinic nature of the adjacent Smectic layers in an AFLC phase makes the C directors anti-parallel for successive layers as the molecules of each layer induce some slow precession motion of the tilt plane orientations by forming a macroscopic helical structure with a typical pitch of the order of some hundreds of nanometers in dimension. Moreover the chiral nature of the elongated molecules produces spontaneous polarization in each of the Smectic layers with polarization vectors being perpendicular to the C directors. Application of electric field with sufficiently large amplitude rotates the interlayer molecular arrangements with a fixed tilt angle and brings the successive layer-to-layer polarization vectors in a preferred orientation with each other and also with the applied field. This is known as the field induced ferroelectric state. Such "pretransitional effect" of induced ferroelectricity arises due to the field-induced distortion of the antiferroelectric helix and also due to the field-induced reorientations of the tilt planes. Dielectric spectroscopy of antiferroelectric liquid crystals is capable to analyze those pretransitional helix distortions of the tilted structures, utilizing the frequency dependent variation in dielectric permittivities (both real and imaginary components) in AFLC phase. The behavior of the real part of the dielectric permitivity at a particular frequency depends on the contribution from the different modes existed or on the change of polarization of adjacent Smectic layers with the applied electric field at that particular frequency. The imaginary part that represents the loss of the system shows peaks related to the corresponding mode relaxation. Therefore the investigation of the dielectric spectra for antiferroelectric liquid crystals reflects a strong light on the fundamental physical functionalities of those phases. Over the years researchers have endeavored [6–17] to clarify several modes with different mechanisms. However most of those reports suggest two definite relaxation modes observed in the frequency range of 100 Hz–10 MHz for an AFLC system. Early works [6–7] predicted such relaxation phenomena associated with the change in the inclination angle of the layer director with the layer normal and thus attributed the observed modes as similar to the soft modes related to the rotation of the molecules around the short molecular axis. More recent works [8–13] ascertained those modes associated to the collective behavior of C directors. They considered the in-phase mode at the low frequency region due to the rotation of the successive

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* Corresponding author. Tel.: +91 33 2582 0184; fax: +91 33 2582 8282.
E-mail addresses: tpm@klyuniv.ac.in, tpmajumder1966@gmail.com (T. Pal Majumder).

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adjacent Smectic layers and the anti-phase one at the high frequency region due to the rotation in the anti-parallel directions. Some results [10,11,13] also suggested that the in-phase motion arose due to the “residual polarization” present in the adjacent Smectic layers of an antiferroelectric phase as a result of the helicity of the structure. When the residual polarization couples with the applied electric field it distorts the helix. Others [13–18] attributed such helix distortions associated with the interaction between the applied electric field and the dielectric anisotropy. The anti-phase motion is appeared due to the net change in the polarization of the Smectic layers in the direction of the applied field brought by a change to the antiferroelectric ordering. Since the direction of the polarization in case of an AFLC system depends on the way in which the layers are counted [19], the origination of the in-phase motion gets changed as the coupling between the applied field and the residual polarization is held responsible for it. On the other hand the coupling between the applied field and the dielectric anisotropy is absolutely responsible for the helix distortion as the dielectric anisotropy for most of the ferroelectric and antiferroelectric phases was found to be negative [20]. We therefore propose the origination of the low frequency mode of an antiferroelectric phase, i.e. the in-phase motion of the C directors in the adjacent Smectic layers are associated not only with the coupling between applied field and induced polarization (rather than residual polarization) but also with the coupling between applied field and the interlayer interaction strength for an AFLC system. This is achieved theoretically utilizing the dependence of the dielectric strength on the externally applied bias for the in-phase motion of the AFLC system in consideration of both the couplings behind the mechanism of the helix distortion.

2. Theoretical approach and discussion

2.1. In-phase mode

The bulk free energy for a pair of Smectic layers of an antiferroelectric liquid crystal (AFLC) can be written as

$$\mathrm{F}=-\mathrm{EP}\cos\phi_{\mathrm{a}}\cos\phi_{\mathrm{b}}+\gamma\cos^2\phi_{\mathrm{b}}+2\mathrm{EV}_0\cos(2\phi_{\mathrm{a}})+\frac{1}{2}\,\mathrm{K}\left(\frac{\partial\phi_{\mathrm{a}}}{\partial x}-\frac{2\pi}{p}\right)^2 \tag{1}$$

where $\phi_a=(\phi_e+\phi_o)/2$ and $\phi_b=(\phi_e-\phi_o)/2$. ϕ_e and ϕ_o describe the azimuthal angles associated with C directors in even and odd layers of the AFLC system, respectively. Therefore we have considered here the variation of the in-phase and anti-phase fluctuations connected with ϕ_a and ϕ_b. The first term in Eq. (1) is the coupling between applied field E and spontaneous polarization P, the second term is the dipolar term determining the appearance of antiferroelectric ordering when the dipolar ordering co-efficient γ is positive, the third term is the coupling between the electric field E and the interlayer interaction for a pair of Smectic layers (V_0 being the interaction strength) and the last one is the elastic energy term with elastic constant K associated with the helical structure of the phase.

Since the in-phase fluctuation does not contain any appreciable variation with respect to the antiferroelectric ordering, minimization of Eq. (1) with respect to ϕ_b is a necessary condition to obtain the stability of the in-phase mode and therefore we obtain the following relation:

$$\sin\phi_b\,(EP\cos\phi_a-2\gamma\cos\phi_b)=0 \tag{2}$$

Since $\sin\phi_b\neq 0$ and hence ϕ_b is governed by the following expression:

$$\cos\phi_b=\frac{EP}{2\gamma}\cos\phi_{\mathrm{a}} \tag{3}$$

In view of the above equation we have the following expression for F, i.e.:

$$\mathrm{F}=-\frac{E^2P^2}{4\gamma}\cos^2\phi_{\mathrm{a}}+2EV_0\cos(2\phi_{\mathrm{a}})+\frac{1}{2}\mathrm{K}\left(\frac{\partial\phi_{\mathrm{a}}}{\partial x}-\frac{2\pi}{p}\right)^2 \tag{4}$$

The viscosity (η_a) connected with the in-phase motion can be expressed using the Landau–Ginzburg (L–G) equation for ϕ_a as

$$-\frac{\eta_{\mathrm{a}}\,p^2}{K}\,\frac{\partial\phi_{\mathrm{a}}}{\partial t}=\frac{p^2}{K}\left(\frac{E^2\,P^2}{4\gamma}-4EV_0\right)\sin(2\phi_{\mathrm{a}})-\frac{\partial^2\phi_{\mathrm{a}}}{\partial T^2} \tag{5}$$

Since the total applied electric field is the summation of bias field (E_b) and oscillatory field ($E_0\exp(i\omega t)$), i.e. $E=E_{\mathrm{b}}+E_0\exp(i\omega t)$, we can consider a trail solution [21] for Eq. (5) as

$$\phi_a=2\pi\mathrm{T}+(a+\mathrm{b}\exp(\mathrm{i}\omega\mathrm{t})+\mathrm{c}\exp(2\mathrm{i}\omega\mathrm{t}))\sin(4\pi\mathrm{T}) \tag{6}$$

where the first term of the trail solution is the ground state solution for an unperturbed helical structure and the second term is a perturbation of the ground state with the Fourier component sin $(4\pi T)$.

Substituting the above trial solution (Eq. (6)) into L–G (Eq. (5)) we obtain the solution for ϕ_{a} as

$$\phi_a=2\pi T-\delta(\omega)\sin(4\pi T) \tag{7}$$

where $T=(x/p)$ is a non-dimensional parameter with p being the pitch of the helix introduced in the L–G equation.

The $\delta(\omega)$ term in the above solution, which describes the motion, can be written as

$$\delta(\omega)=\left[\left(E_b^2+\frac{E_0^2}{2}-\frac{16V_0\gamma}{P^2}E_b\right)+\left(2E_{\mathrm{b}}E_0-\frac{16V_0\gamma}{P^2}E_0\right)\times\frac{\exp(i\omega t)}{1+i\omega\tau_{\mathrm{a}}}+\frac{E_0^2}{2}\,\frac{\exp(2i\omega t)}{1+2i\omega\tau_{\mathrm{a}}}\right]\frac{P^2p^2}{64\pi^2K\gamma} \tag{8}$$

with $\tau_a=(\eta_aP^2/16\pi^2K)$, the time constant for ϕ_a relaxation.

For those frequencies (low frequency) well below the ϕ_a relaxation ($\omega\ll(1/\tau_a)$), we have

$$\phi_{\mathrm{a}}=2\pi T-\frac{E^2P^2p^2}{64\pi^2K\gamma}\sin(4\pi T)+\frac{p^2V_0}{4\pi^2K}\,E\sin(4\pi T) \tag{9}$$

For those frequencies (high frequency) well above the ϕ_a relaxation ($\omega\gg(1/\tau_a)$), we have

$$\phi_{\mathrm{a}}=2\pi T-\left(E_{\mathrm{b}}^2+\frac{E_0^2}{2}\right)\frac{P^2p^2}{64\pi^2K\gamma}\sin(4\pi T)+\frac{p^2V_0}{4\pi^2K}\,E_{\mathrm{b}}\sin(4\pi T) \tag{10}$$

here $(E_b^2+(E_0^2/2))$ is the mean squared electric field. The difference between the amplitudes of ϕ_a for low and high frequency relaxations is equivalent to the difference between that for low frequency, ϕ_a as given in Eq. (9) containing total field E, and for high frequency, ϕ_a, as given in Eq. (10).

The net polarization for a two-layer AFLC system is $P_Z=\mathrm{P}\cos\phi_{\mathrm{a}}\cos\phi_{\mathrm{b}}$.

In view of Eq. (3), we have

$$P_Z=\frac{EP^2}{2\gamma}\cos^2\phi_{\mathrm{a}} \tag{11}$$

When the applied field is sufficiently small, we can assume $\delta(\omega)\ll 1$, then we can consider the following relation as given below:

$$\cos^2\phi_{\mathrm{a}}\approx\frac{1}{2}\,[1+\cos(4\pi T)]+\frac{\delta(\omega)}{2}[1-\cos(8\pi T)] \tag{12}$$