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Field-driven helix unwinding in antiferroelectric liquid crystal cells

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The interaction between the antiferroelectric helix and an electric field applied perpendicular to its axis is investigated, both experimentally and theoretically. A two-stage switching process is observed: (i) in the pretransitional regime the helix distorts and then unwinds to form a nonhelical antiferroelectric state, with the plane of the molecules parallel to the applied field; (ii) at higher fields switching to the ferroelectric state occurs. The mechanism for unwinding is the interaction of the applied field with a polarization that is induced by a change to the anticlinic ordering.

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Smectic liquid crystals have positional order such that the molecules are arranged approximately into layers forming two-dimensional liquids. In tilted smectics, the orientational order within each layer is such that the average direction of the molecules (the director) is at an angle θ to the layer normal. The *c*-director is the projection of this director onto the layer, and is described by an azimuthal angle ϕ. Antiferroelectric liquid crystals (AFLCs) [1–3] have an anticlinic coupling between layers, which causes the *c*-directors of adjacent layers to be almost antiparallel. The discrepancy is due to the chirality of the molecules, which causes a small precession of the *c*-directors from layer to layer, and hence a macroscopic helical structure. Each smectic layer has a spontaneous polarization that is perpendicular to both its normal and *c*-director. The coupling of these polarizations to an electric field applied along the smectic layers causes the polarizations to align with the field at sufficiently high fields: this is known as the ferroelectric state.

In this Rapid Communication, we concentrate on the ‘‘pretransitional’’ switching regime, that is, where the applied field is too low to cause switching to the ferroelectric state. Our microscopic observations of thick, homogeneously aligned AFLC cells show that the dechiralization lines (characteristic of the helical superstructure), disappear at fields within the pretransitional regime, suggesting that the helical structure is destroyed before the transition to the ferroelectric state takes place. Additionally, light that is Bragg scattered from the helical structure in the ground state disappears as the applied voltage increases, again before switching to the ferroelectric state occurs.

Although there have been a number of studies of the field driven transition of the antiferroelectric state to the ferroelectric state [4,5], the role of the macroscopic helix in the pretransitional regime is not well understood. Gorecka *et al.* [6] undertook studies in homeotropic cells and reported that under applied fields within the pretransitional regime, the helix distorts so that the *c*-directors rotate to align with the field while maintaining approximate anticlinic ordering between layers. Eventually the helix is unwound to form a non-helical antiferroelectric state in which the directors are in the plane containing the electric field. The authors attribute this process to the interaction of the applied field with the dielectric anisotropy of the molecules. Moritake *et al.* [7], Hiraoka *et al.* [8], and Panarin *et al.* [9] agree that the coupling of the electric field to the dielectric anisotropy could be responsible for the helix distortions observed in their dielectric studies. Panarin *et al.* also report a second mechanism for helix distortion in homogeneous cells: the interaction of the applied field with the ‘‘antiferroelectric polarization,’’ which they claim arises from the incomplete cancellation of the polarizations of adjacent layers of the AFLC helix. This interaction is also recognized by Buivydas *et al.* [10,11], who refer instead to the interaction of the electric field with the ‘‘residual polarization,’’ and do not consider coupling to the dielectric anisotropy.

There therefore seems to be no unified understanding of both the mechanism of electric field induced helix distortion and the nature of the unwound state. It is the aim of this work to give a complete description of the pretransitional regime of AFLC switching in homogeneously aligned cells. We will show that it is possible to explain a number of observed effects with a simple model of the interaction between the applied field and the polarizations of the AFLC layers, without any interaction with either the dielectric anisotropy or the ‘‘residual polarization.’’

Since the macroscopic helix is present in the ground state, we assume that the bulk of the liquid crystal is unaffected by the surfaces, and hence it is possible to model the structure in one dimension only: along the helical axis. We also assume that the helical axis does not tilt (because the smectic layers are very rigid) and the angle that the director makes with the layer normal, θ, is constant (because the electroclinic effect is negligible so far from the nearest phase transition). We can therefore consider only the changes in the azimuthal angles ϕ in each layer by solving the following torque equation for the *i*th smectic layer:

$$\eta\frac{\partial\phi_i}{\partial t}=-EP_s\sin\phi_i+\frac{\gamma}{2}\sin(\phi_i-\phi_{i+1}+\delta)+\frac{\gamma}{2}\sin(\phi_i-\phi_{i-1}-\delta).$$

The left-hand side (LHS) is the product of the viscosity coefficient η for changes in the azimuthal angle of the *c*-director, ϕ, and the time derivative of ϕ of the *i*th layer. This is equated to the total torque on the director, given by the terms on the RHS. The first is the interaction of the layer

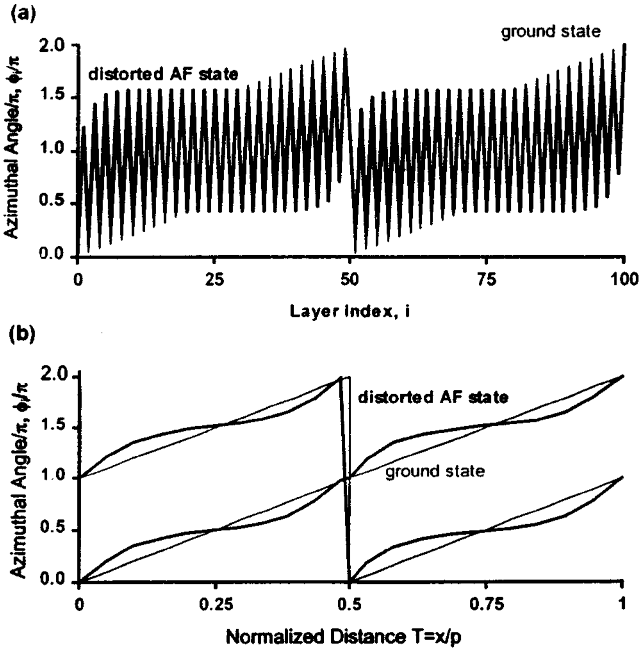

FIG. 1. Theoretical prediction of antiferroelectric helix distortion by an applied field using (a) discrete and (b) continuous order parmameters. The ground state is shown by thin lines, and the distorted state by thick lines. The director oscillates approximately from one side of the smectic one to the other, so the discrete model (a) shows an oscillatory behavior of the azimuthal angle (plotted between 0 and 2π). For the continuous model (b), however, the azimuthal angles in both odd and even layers are plotted simultaneously. Both models show that the *c*-directors rotate to align with the field, together with a small change in the anticlinic ordering.

spontaneous polarization (of magnitude P_s) with the applied electric field (of magnitude E). The other two terms represent the antiferroelectric interaction, of strength γ, of the ith layer with its neighboring layers $i-1$ and $i+1$. Both terms are zero in the ground state ($E=0$), when the directors are almost on opposite sides of the smectic cone (the discrepancy being an amount δ that is due the helical superstructure).

The steady state solution of the equation is determined numerically, and shown in Fig. 1(a) for a 100 layer pitch (i.e., $\delta=2\pi/100$), for both the ground state ($E=0$), and an applied field (in this case such that $EP_s=\gamma/2$). *The helix distorts so that those regions with the c-directors parallel to the field grow, while maintaining approximate anticlinic ordering.* This distorted helical structure will at higher fields unwind fully to form an uniform antiferroelectric (AF) structure, with the liquid crystal directors in the plane perpendicular to the bounding plates and containing the electric field. This will be referred to as a ''vertical'' AF (VAF) state, as opposed to a ''horizontal'' AF (HAF) state, in which the directors lie in the plane parallel to the cell plates. The model therefore gives results that are consistent with the experimental results of Gorecka *et al.* in homeotropic cells [6].

Figure 1(a) also shows the perturbation to the ground state helix occurs with a wavelength of half the pitch. Since the physical properties of an antiferroelectric liquid crystal repeat every half pitch (as seen from selective reflection [1]), it is clear that the results of the model are consistent with the symmetry of the antiferroelectric state. This disagrees with the hypothesis that the mechanism of helix distortion is the interaction of the electric field with the ''residual polarization'' [9–11], since in that case the distortions would occur with a wavelength equal to that of the residual polarization, i.e., one pitch. Examination of the results reveals that the *mechanism* for helix distortion is the interaction between the electric field and an *induced* polarization (with a wavelength of half a pitch). This is caused by a perturbation to the anticlinic ordering, and lies approximately along the local direction of the *c*-directors. The induced polarization interacts with the applied field to rotate the *c*-directors to align with the field, and eventually causes helix unwinding. The concept of an induced polarization was suggested by Hiraoka *et al.* [8], but was considered to arise initially from the interaction of the electric field with the dielectric anisotropy: we have, however, shown that this is not necessary.

In order to support these results, the antiferroelectric helix is also modeled using continuous order parameters ϕ_a and ϕ_b, defined as follows:

$$\phi_a=\frac{\phi_e+\phi_o}{2}, \quad \phi_b=\frac{\phi_e-\phi_o}{2},$$

where ϕ_e and ϕ_o are the azimuthal angles of the director in even and odd layers, respectively. ϕ_a describes the local average azimuthal angle of the director, and ϕ_b the type of ordering, where $\phi_b=0$ is ferroelectric ordering and $\phi_b=\pi/2$ is antiferroelectric ordering. The free energy of a pair of layers can be written as

$$F=-EP_s\cos\phi_a\cos\phi_b+\frac{K}{2}\left(\frac{\partial\phi_a}{\partial x}-\frac{2\pi}{p}\right)^2+\Gamma\cos^2\phi_b.$$

The first term is the interaction between the electric field and the spontaneous polarizations of both the odd and even layers in the pair [it is $-EP_s(\cos\phi_o+\cos\phi_e)/2$]. The second term is the elastic energy stored as a result of distortions to the helix, and hence only involves ϕ_a: K is an elastic constant; x is the distance along the helical axis, and p is the pitch of the antiferroelectric helix. The final term represents the antiferroelectric ordering, of strength Γ, and depends on ϕ_b only.

The numerical continuation package AUTO97 [12,13] is used to find the minimum energy states as a function of applied electric field. At $E=0$, the helical AF state is lowest in energy, as expected, but above a threshold field, the VAF state is lowest in energy (the HAF state is always highest in energy). Defects or thermal fluctuations in the cell will seed domain switching to the state of lowest energy and hence the helical AF state is expected to unwind into this VAF state. Figure 1(b) shows that the distortion of the helix for electric fields lower than the threshold for unwinding is such that the *c*-directors tend to align with the electric field, accompanied by a distortion of the anticlinic ordering. The relative amounts of helix distortion and changes to the anticlinic ordering are controlled by the dimensionless parameter $\Gamma p^2/K$: the larger it is, the smaller the changes to the anticlinic ordering are at a given field. The results shown in Fig. 1(b) are

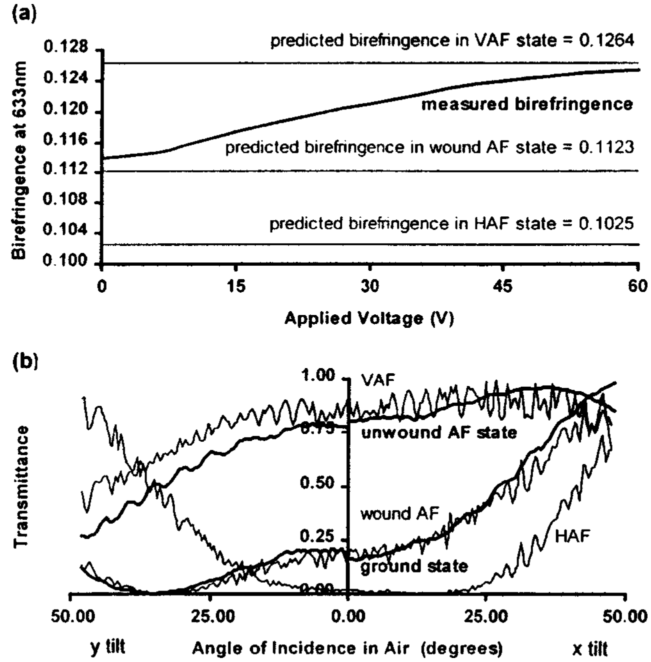

FIG. 2. Experimental confirmation of results of theoretical model as shown by (a) measurements of the birefringence as a function of applied field, and (b) laser conoscopy for the ground and unwound AF states (tilts about y and x axes in the left- and right-hand parts of the graph, respectively). The thick and thin lines represent experimental and theoretical data, respectively. The birefringence (a) increases in the pretransitional regime, towards a value that is predicted for the VAF state. In (b), the experimental data for the ground and unwound AF states are seen to agree with the Berreman predictions for the helical AF and VAF states, respectively. Both experiments confirm that the unwound state is not the HAF state, but the VAF state.

for $\Gamma p^2/K = 1\times 10^4$, and are clearly qualitatively the same as those of the discrete model. We therefore have a clear and unified prediction for the nature of helix distortion and unwinding in the pre-transitional regime from these two theoretical models.

In order to test the results of the modeling experimentally, it is necessary to consider the macroscopic consequences of the helix distortions shown in Fig. 1, which is followed eventually by complete helix unwinding. As the helix distorts, the refractive index in the direction along the smectic layers, but perpendicular to the applied field, decreases, whereas that perpendicular to the field and the smectic layers remains the same, i.e., the birefringence increases. A test of the model is therefore to measure the birefringence of a suitable sample as a function of applied field.

This was done by illuminating a thick (12 μm), homogeneously aligned cell of the commerical AFLC mixture from Chisso, CS4001, (between crossed polarizers) with a 632.8 nm HeNe laser at normal incidence, and measuring the transmittance as a function of the azimuthal orientation of the cell with respect to the polarizers. The result was used to determine the birefringence as a function of voltage. Figure 2(a) shows that in the pretransitional regime, the birefringence of the material increases (as the helix distorts) from a value of 0.1140 in the ground state, to 0.1255 at a voltage of 60 V (at which the helix is known to be unwound, from microscopic observations and laser scattering). At voltages greater than the threshold for switching to the ferroelectric state, the birefringence was measured to be 0.1524. This value was used to predict the birefringences of the helical AF, VAF, and HAF states: 0.1123, 0.1264, and 0.1025, respectively. It is clear that the predicted values for the helical AF and VAF states agree very well with the measured values for the birefringence at 0 and 60 V, respectively. The measurement of the birefringence of the liquid crystal therefore supports the theory that the antiferroelectric helix is unwound to the VAF state.

Further configuration of the theory comes from laser conoscopy experiments, in which the transmittance of the cell between crossed polarizers is measured as a function of the angle of incidence, i.e., the cell is tilted between the polarizers, both about the helical axis (x), and about the axis perpendicular to this and the electric field (y). For each axis of rotation, experimental results are shown in Fig. 2(b) both for the ground state, and for 60 V. Also plotted in Fig. 2(b) is the output from a Berreman 4×4 matrix optical model [16], for the helical AF, VAF, and HAF states, which all use the same values of the refractive indices for a single smectic layer. It is clear that, for tilts about both x and y axes, the predicted output for the uniaxial helical AF and biaxial VAF states agree well with the experimental data at 0 and 60 V, respectively. The laser conoscopy experiment therefore confirms the theory that the antiferroelectric helix is unwound to the VAF state.

A further consequence of the helix distortion is a change in the low frequency dielectric constant of the cell. From the continuous model, the component of the polarization along the direction of the applied field is

$$P_z = P_s \cos\phi_a \cos\phi_b \equiv \frac{EP_s^2}{2\Gamma}\cos^2\phi_a,$$

and therefore the relative dielectric constant is

$$\epsilon = \epsilon_\infty + \frac{P_s^2}{2\epsilon_0 p\Gamma}\int_0^p \cos^2\phi_a dx + \frac{EP_s^2}{2\epsilon_0 p\Gamma}\int_0^p \frac{\partial}{\partial E}(\cos^2\phi_a)dx.$$

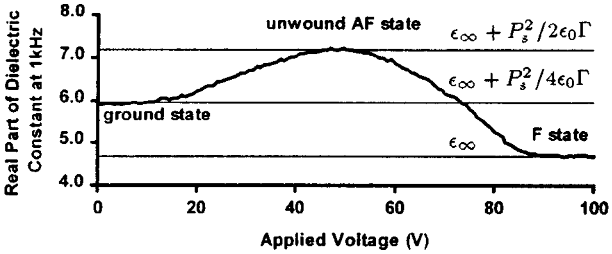

FIG. 3. Measurement of the low frequency dielectric constant as a function of bias field. Relative to the ferroelectric state, the dielectric constants of the unwound AF and ground states are in a ratio of 2:1, in quantitative agreement with the predictions of the continuous model. This supports the conclusion that the antiferroelectric helix unwinds to the VAF state in the pretransitional regime.

This has a value of $\epsilon_\infty + P_s^2/4\epsilon_0\Gamma$ for $E=0$, a value of $\epsilon_\infty + P_s^2/2\epsilon_0\Gamma$ for the VAF state, and a value of ϵ_∞ in both the HAF and fully switched F states. The dielectric constant at a frequency of 1 kHz (the lowest frequency at which ionic behavior was not present) was measured as a function of voltage, and the results are shown in Fig. 3. In the pretransitional regime, the dielectric constant increases, in agreement with the prediction of the continuous model (i.e., that the helix unwinds to the VAF state, which has double the dielectric constant of the ground state, relative to ϵ_∞). Clearly the helix cannot be unwinding to form the HAF state, as in that case we would observe no change in dielectric constant at the transition to the ferroelectric state. Comparison between experimental and theoretical results gives a value for the antiferroelectric ordering constant Γ of 1.6×10^4 J/m^3, given that $P_s = -80$ nC/cm^2 for CS4001.

In conclusion, our work has shown clearly for the first time that the mechanism of helix distortion in an AFLC is the interaction between the applied field and a polarization that is induced by an unequal rotation of the directors in odd and even layers (a perturbation to the anticlinic ordering). This is in contrast with the mechanisms suggested by others, in which the electric field interacts with either the dielectric anisotropy [6–8], the residual polarization [10,11], or both [9]. Our ‘‘induced polarization’’ repeats every half pitch through the helical structure, in agreement with the symmetry of the AFLC phase. The interaction eventually causes the helix to be completely unwound to a uniform antiferroelectric state with the *c*-directors aligned with the applied field (the VAF state), at fields lower than that required to switch the material into a ferroelectric state. Our models successfully explain in a unified way the following phenomena observed in the pretransitional regime of thick homogeneously aligned AFLC cells: two stage switching (as seen by microscopic observation and laser scattering), the increase in birefringence and the changes in the conoscopic image and low frequency dielectric constant.

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