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Twist instability of a flexoelectric nematic domain in an external field

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We report the observation of an instability in a flexoelectric nematic domain due to a linear coupling with an external dc electric field. We use a hybrid aligned nematic cell with two antiparallel flexopolarized domains. Above a certain threshold, the field induces a twist instability in domains with the flexopolarization opposed to the field. The threshold field in methoxybenzylidenebutylaniline is in reasonable agreement with a calculation which also includes the effect of the dielectric anisotropy.

Field-induced textural transitions are well known in nematic liquid crystals. The Freedericksz transition and the electrohydrodynamic instability (EHD) are two examples which occur at specific threshold voltages. These effects arise from apolar couplings of the dielectric and conductivity anisotropies with an external electric field: the phenomena occur for either polarity of the field, or for (low frequencies in the case of EHD) ac fields, a consequence of the well-known apolar character of the nematic director $\vec{n}$.[1] As was first argued by Meyer,[2] a nonuniformly oriented nematic can have a net electric polarization. The "flexoelectric" effect was originally envisaged by him as arising from a coupling either between the longitudinal dipole moments of "pear-shaped" molecules with a splay deformation (characterized by the coefficient e_1) or between the transverse dipole moments of "banana-shaped" molecules with a bend deformation (e_3). It is now known[3] that e_1 and e_3 arise more generally, even in the absence of shape asymmetries, due to the quadrupolar densities of the medium coupling with the deformations of the director field. However, if the deformation is weak, or if the two flexoelectric coefficients are equal ($e_1 = e_3$), the coupling between $\vec{P}$ and $\vec{E}$ results only in surface effects. If the surface anchoring is strong, these effects are not visible and one needs weakly anchored surfaces to see them.[4] On the other hand, if $e_1 \neq e_3$ and the deformation of the director is substantial, a volume flexoelectric polarization is obtained, as was recently demonstrated.[5] In this Rapid Communication, we report the first observation of an electric instability in a nematic flexoelectric domain produced by a linear coupling of the flexopolarization with an external dc electric field opposite to the polarization.

Consider a hybrid cell as shown in Fig. 1 (inset) in which the lower and upper plates are treated to give homeotropic and homogeneous alignments, respectively. This leads to a splay-bend distortion in the orientation pattern of the director with two possible opposite curvatures; which in turn gives rise to two types of domains with opposite volume flexoelectric polarization $\vec{P}$ parallel to x and localized close to the planar oriented plate

$$\vec{P} = e^* \vec{n}\, \mathrm{div}\vec{n} \simeq e^* \vec{n}/d.$$

$e^* = e_1 - e_3$ is the anisotropic volume flexo coefficient which is the only relevant part[5] for coupling with an external uniform electric field, and d the sample thickness. If a dc field is applied along the x axis in the domains with $\vec{P}$ opposite to $\vec{E}$, an instability of the director orientation can be expected to occur beyond a threshold field E_{th} while in the domains with $\vec{P} \| \vec{E}$, the director orientation is stabilized. In order to appreciate the physical origin of the instability, we first write the free energy density of the sample under the field taking into account only the flexoelectric polarization and the elastic energy, using the one curvature elastic constant ($=K$) approximation:

$$F = e^* \sin^2\theta \cos\phi \frac{d\theta}{dz} E + \frac{K}{2}\left[\left(\frac{d\theta}{dz}\right)^2 + \sin^2\theta\left(\frac{d\phi}{dz}\right)^2\right],$$

where the angles θ and ϕ are defined in the inset of Fig. 1. For $E < E_{th}$, $\phi = 0$ and just above E_{th}, ϕ is very small and we can assume that $d\theta/dz = \pi/2d$ as in the field free case. We can easily evaluate E_{th} by expanding $\cos\phi$ in ϕ and then ϕ in a Fourier series: $\phi = \sum_i \phi_i \cos q_i z$ where $q_i = i\pi/2d$. The lowest threshold mode occurs for the first Fourier component. Retaining only this component and integrating F over the sample thickness, we can expand for very small ϕ, the free energy per unit area in the form

$$\mathcal{F} = \mathcal{F}_0 + a\left[\frac{1}{2}(E_{th} - E)\phi^2 + \frac{1}{4}\left(\frac{E}{12}\right)\phi^4 + \ldots\right], \quad (1)$$

where $a = \pi e^*/16$, and

$$E_{th} = 3\pi K/(2de^*). \quad (2)$$

Obviously for $E > E_{th} > 0$ an instability of the director field occurs, with nonzero values of ϕ. Above E_{th}, $\phi = [12(E - E_{th})/E]^{0.5}$. For $E < 0$, F remains minimum for $\phi = 0$.

The experiments were done at room temperature on

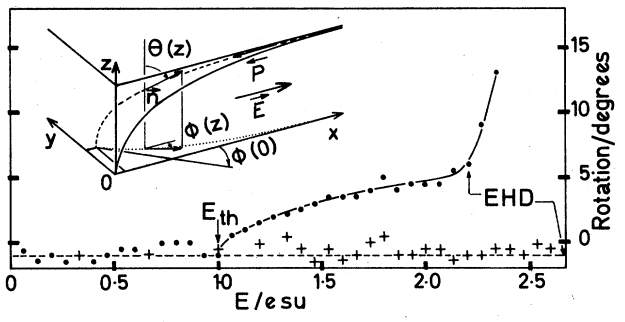

FIG. 1. Rotation $\phi(0)$ of the plane of polarization plotted as a function of the applied field. ●, $\vec{E}$ antiparallel to $\vec{P}$. E_{th} is the threshold field for the twist instability of the flexodomain + $\vec{E}$ parallel to $\vec{P}$. Inset shows the experimental geometry.

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methoxybenzylidenebutylaniline (MBBA) for which both K and e^* have been independently measured.[1,5] The planar orientation was obtained by coating glass plates with silicon monoxide at an oblique angle and the homeotropic orientation by coating with *N,N*-dimethyl-*N*-octadecyl-3-aminopropyltrimethoxysilyl chloride (DMOAP) silane. Two stainless-steel wires of 49-μm diameter were used as spacers with electrodes. The instability should result in a nonzero $\phi(z)$ which can be seen as a rotation of the plane of polarization of light beam traveling from the homogeneously treated plate to the homeotropic one, as we are in the wave-guide regime[5] [$\Delta n(d\phi/dz) >> \lambda$, where Δn is the effective birefringence and λ the wavelength of light]. The experimental results are shown in Fig. 1, in which $\phi(0)$ is plotted as a function of E. The instability occurs at a well-defined threshold field ($E_{\rm th} \simeq 1$ esu) when $\vec{E}$ is in a direction opposite to that of $\vec{P}$. From conoscopic observations we could determine the curvature of the director in the given domain, and hence, the direction of $\vec{P}$, as the sign of e^* is known to be positive.[5] When $\vec{E}$ and $\vec{P}$ are in the same direction, no instability occurs, proving that the origin of the instability is the linear coupling between $\vec{P}$ and $\vec{E}$. Equivalently, when there are two neighboring domains with opposite curvatures, the instability occurs in one or the other domain depending on the sense of the applied field. When the field is increased beyond about 2 esu, the medium is set into electrohydrodynamic (EHD) motion. The EHD instability threshold is obviously found to be larger, when $\vec{E}$ is parallel to $\vec{P}$ (stabilizing geometry), than for the destabilizing antiparallel geometry. Using the experimental data of $K \simeq 5\times10^{-7}$ dyne,[1] and $e^* = 1\times10^{-4}$ esu,[5] $E_{\rm th}$ calculated from Eq. (2) is about five times too large. We improve the calculation by including the destabilizing effect of the negative dielectric anisotropy $\Delta\epsilon$ and of the anisotropic elastic constants, which change the $\theta(z)$ profile below $E_{\rm th}$. The free energy density is given by

$$F = e^* \sin^2\theta \cos\phi E \frac{d\theta}{dz} - \Delta\epsilon \sin^2\theta \cos^2\phi E^2/8\pi + \tfrac{1}{2}K_{33}\left[(1-K_1\sin^2\theta)\left(\frac{d\theta}{dz}\right)^2 + \sin^2\theta(1-K_2\sin^2\theta)\left(\frac{d\phi}{dz}\right)^2\right],$$

where $K_1 = (K_{33}-K_{11})/K_{33}$, $K_2 = (K_{33}-K_{22})/K_{33}$, K_{11}, K_{22}, and K_{33} being the splay, twist, and bend elastic constants, respectively. As we are interested in the calculation of $E_{\rm th}$ only, we first find the new $\theta(z)$ profile under the action of E with $\phi = 0$. The corresponding Euler-Lagrange equation is

$$\frac{-\Delta\epsilon E^2 \sin 2\theta}{(8\pi K_{33})} + 0.5K_1 \sin 2\theta\left(\frac{d\theta}{dz}\right)^2 - (1-K_1\sin^2\theta)\frac{d^2\theta}{dz^2} = 0.$$

It can be integrated to yield

$$z = (A-C)^{-1/2}\int_0^{\theta}[(1-K_1\sin^2\alpha)(1+P\sin^2\alpha)^{-1}]^{1/2}d\alpha \ , \tag{3}$$

where $A = \Delta\epsilon E^2/(8\pi k_{33})$ and $P = -2A/(A-C)$, and C a field-dependent integration constant which can be determined from Eq. (3) using the boundary condition that $\theta = \pi/2$ for $z = d$. In order to determine $E_{\rm th}$ we proceed as before, writing $\phi(z) = \phi(0)\cos(\pi z/2d)$. After integration over z, we again write $(\mathcal{F} - \mathcal{F}_0)$ as a power series of $\phi(0)$. The coefficient of $\phi(0)^4$ is again positive and the coefficient of $\phi(0)^2$ is now a quadratic function of E and hence there are two threshold values. We can evaluate the smaller $|E_{\rm th}|$ only by a numerical calculation. The larger one corresponds to the balance of the destabilizing dielectric effect and the stabilizing flexo contribution, for $\vec{E}\|\vec{P}$. The smaller one corresponds to the superposed destabilizing effects from both contributions, for $\vec{E}$ opposed to $\vec{P}$. With $\Delta\epsilon = -0.7$[1], $K_{33} = 8\times10^{-7}$ dyne, $K_{11} = 5\times10^{-7}$ dyne, and $K_{22} = 3\times10^{-7}$ dyne,[1] we obtain $E_{\rm th} = 1.3$ esu for $d = 49\times10^{-4}$ cm, which is in reasonable agreement with the experimental value of 1 esu.

Hence, the dielectric torque effectively reduces the threshold field not only by a direct effect on the planar oriented molecules but by changing the $\theta(z)$ profile such that the flexopolarization is enhanced. There is another physical process which can also be expected to influence the $\theta(z)$ profile: the flexopolarization results in a charge density $q = -\mathrm{div}(P)$ which is of opposite signs near the two plates. For low enough conductivities of the sample, these charges are not completely screened by ionic impurities in the medium. Under the action of an external field, the material could flow in opposite directions near the two plates. The resulting hydrodynamic torque linear in E can be estimated to be negligibly small. Anyway, in our sample, the conductivity is large enough to screen these polarization charges. The conductivity anisotropy also leads to a similar effect quadratic in E. We expect that the change in $\theta(z)$ will not be important near $E_{\rm th}$.

In conclusion, we have for the first time experimentally demonstrated an electrically produced instability in the orientation of a nematic liquid crystal due to linear coupling with an electric field, in a flexopolarized hybrid aligned cell. The threshold field for the instability can be almost quantitatively accounted for by the known flexoelectric, dielectric and elastic constants of the material used.

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