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Smectic-*A* Fréedericksz transition

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In the smectic *A* phase at a temperature near the smectic-*A* to smectic-*C* point a field induced Freedericksz transition is predicted to exist by this work. This transition can occur due to the possibility of induced molecular tilt accompanied by smectic layer distortion. The existence of the transition is confirmed experimentally. Temperature dependence of the transition voltage is predicted, and also confirmed experimentally. Further, the influence of finite surface tilt on the transition is considered. [S1063-651X(98)51308-6]

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The smectic-*A* phase of liquid crystal material has both orientational and positional ordering [1]. Orientational order is similar to that in the nematic phase, and allows the definition of a director **n** as the average molecular long axis. Positional order consists of layering of the molecules, with the layer normal in the **n** direction and the pitch of the layering being approximately equal to the molecular length.

It is well known that in the aligned nematic phase a field induced orientational transition can take place. If a nematic liquid crystal sample is placed between bounding plates that induce parallel alignment of the liquid crystal director, then a so called homogeneously aligned liquid crystal layer will be formed. Provided the dielectric anisotropy of the liquid crystal material is positive then application of a field (electric) normal to the bounding plates will then tend to reorientate the director to be parallel with the field, and thus perpendicular to the bounding plates, if the director is anchored at the surfaces then a distorted state will be formed. This orientational transition is termed the Freedericksz transition [2], and if the field is provided by a voltage applied across the bounding plates it takes place above a critical threshold given by

$$V_{\mathrm{th}} = \pi \sqrt{(K/\varepsilon_0 \Delta\varepsilon)},$$

where K is the elastic constant and $\Delta\varepsilon$ is the relative dielectric anisotropy of the nematic liquid crystal material.

In the smectic-*A* phase it is normally accepted that such a transition cannot be observed [3]. Again starting with a homogeneously aligned sample with the director parallel to bounding plates there will now be smectic layering present, with the layer normal parallel to the surfaces also. If a field which tends to reorientate the director is applied across such a sample no elastic distortion should take place. This is because there is no reorientation profile that can retain the interlayer spacing and packing structure of the smectic layering. Thus, no reorientation is seen until a threshold for plastic distortion is reached [4]. This results in permanent reorientation of the layering through defect formation which can only be recovered by thermal annealing of the sample in the nematic phase. It appears therefore that no smectic-*A* Freedericksz transition with an elastic distortion analogous to that in the nematic phase is observable.

Near the smectic-*A* to smectic-*C* phase transition point however tilt can occur in the smectic-*A* phase, accompanied by smectic layer shrinkage. For example, physical compression of the smectic layering can induce a tilt [5]. A similar process also takes place across the smectic-*A* to smectic-*C* phase transition, and in homogeneously aligned samples generally results in the formation of a kinked (or chevron) structure in the smectic layering. The occurrence of this allows retention of the layer packing formed in the smectic-*A* phase together with molecular tilt relative to the smectic layer normal. Generally this tilt is in the opposite direction from the layer tilt direction, resulting in approximate retention of the homogeneous alignment. However, under field application in the smectic-*C* phase, for materials with positive dielectric anisotropy, the director can reorientate so that the tilts are in the same direction [6], and the resulting director tilt is the sum of the layer tilt angle and the tilt of the director relative to the layer normal. Given that near the smectic-*A* to smectic-*C* transition point layer compression can induce tilt, it should also be possible to induce this state through field application to a homogeneously aligned sample in the smectic-*A* phase just above the transition to the smectic-*C* phase. An applied field would then tend to induce director tilt through the same mechanisms of layer shrinkage and tilt. Thus, we would expect a smectic-*A* Freedericksz transition to exist near the smectic-*A* to smectic-*C* transition point.

In order to predict the voltage threshold of this transition we assume that the internal energy density of the sample near the smectic-*A* to smectic-*C* transition can be expressed as

$$f = \frac{a}{2}\,\theta^2 + \frac{b}{4}\,\theta^4 + \frac{K}{2}\left(\frac{d\chi}{dz}\right)^2 - \frac{1}{2}\,\varepsilon_0 \Delta\varepsilon E^2 \chi^2,$$

where θ is the induced molecular tilt relative to the layer normal, a and b are Landau coefficients, χ is the tilt of the director relative to the sample boundaries, K is an elastic constant, z is the direction normal to the surfaces, $\Delta\varepsilon$ is the (positive) relative dielectric anisotropy, and E is the internal electric field. It is assumed that near the transition the director tilt is small and therefore small angle approximations have been used in the above equation. In order to solve this it is necessary to relate the director tilt relative to the layer normal to the director tilt relative to the sample boundaries (i.e., to relate θ and χ). This is done through the approximation $\delta \approx c\,\theta$, where δ is the layer tilt angle and c is a constant of proportionality, typically around 0.9 [7]. As the layer tilt and director tilt are expected to be in the same direction (i.e., to add together) the total director tilt can be written as

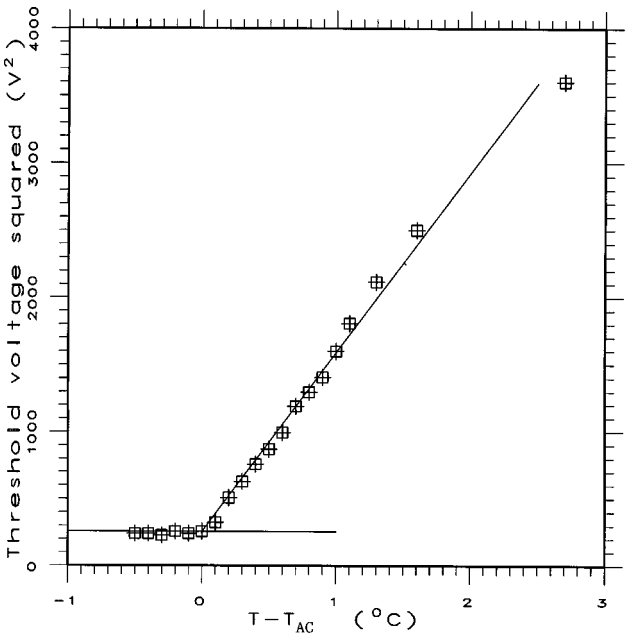

FIG. 1. Threshold voltage squared as a function of temperature (measured from the smectic-*A* to smectic-*C* transition point). Data are shown as points, together with two straight line fits with the break point at $T-T_{AC}=0$ °C.

$\chi \approx (1+c)\theta$. Taking this together with the above equation allows an Euler-Lagrange equation in χ to be derived as

$$\frac{\alpha(T-T_{AC})}{(1+c)^2}\chi+\frac{b}{(1+c)^4}\chi^3+K\frac{d^2\chi}{dz^2}-\varepsilon_0\Delta\varepsilon E^2\chi=0,$$

where the coefficient a in the Landau terms has been written in its temperature dependent form. On field application it is expected that the lowest mode of distortion will have nodes at the surfaces and a node in the center of the sample. This is in order to be consistent with the retention of smectic layer packing and formation of a kinked structure, analogous to the chevron structure [7], as discussed above. Thus, looking for solutions of the above equation in the form $\chi=\chi_0 \sin(2\pi z/d)$, where d is the device thickness, allows the determination of a critical, or threshold, voltage for the smectic-*A* Freedericksz transition (SAFT) as

$$V_{\mathrm{SAFT}}=\frac{1}{\sqrt{\varepsilon_0\Delta\varepsilon}}\left\{4\pi^2K+\alpha(T-T_{AC})\frac{d^2}{(1+c)^2}\right\}^{1/2}.$$

There are a couple of interesting points to notice in this expression:

(i) At the smectic-*A* to smectic-*C* phase transition temperature the expression simplifies to one very similar to that for the nematic case, but with a threshold voltage of twice that for the nematic Freedericksz transition. This is also the same as the value obtained by Kedney and Stewart for the lowest order sin mode of layer deformation in a smectic-*C* phase [8].

(ii) The threshold voltage squared is linear in temperature.

In order to test the above theory an experiment is performed to study the smectic-*A* Freedericksz transition. A parallel aligned low pretilt homogeneous test cell of around 2 μm thickness is filled with the liquid crystal material $\overline{8}$S5. This material has nematic, smectic-*A*, and smectic-*C* phases, and also has positive dielectric anisotropy. The test cell is placed between crossed polarizers, in a temperature controlled environment, and the transmission observed during field application.

First, the Freedericksz transition in the nematic phase was investigated, at very low frequency. Recording the threshold voltage for this at a number of temperatures, and extrapolating to the smectic-*A* to smectic-*C* phase transition temperature gives a value of 8.5 ± 0.5 V at $T=T_{AC}$. It was noted above that the smectic *A* Freedericksz transition threshold at the smectic-*A* to smectic-*C* transition point is expected to be twice the nematic value, i.e., this predicts a threshold voltage of 17.0 ± 1.0 V at $T=T_{AC}$.

The smectic-*A* Freedericksz transition threshold is determined by applying a low frequency triangular wave to the device, increasing the amplitude of this until a response is observed at the peaks, and then extrapolating this response back to the background level to define the threshold. This is analogous to the technique commonly used for measuring the threshold in nematic devices.

Results are plotted in Fig. 1 in the form of $V_{\mathrm{SAFT}}{}^2$ against $T-T_{AC}$. Two regions are observed:

(i) Below T_{AC} (for a range of 0.5° at least) the threshold is approximately constant, remaining at the value at the phase transition of 16 ± 1 V. This gives a parameter value of $4\pi^2K/(\varepsilon_0\Delta\varepsilon)=256\pm33$ V^2. The threshold at the transition temperature is very close to the value predicted by extrapolation from the nematic phase Freedericksz transition, and confirms the existence of a node in the center of the device due to the smectic layer packing constraints. It is believed that the threshold remains approximately at this value in the

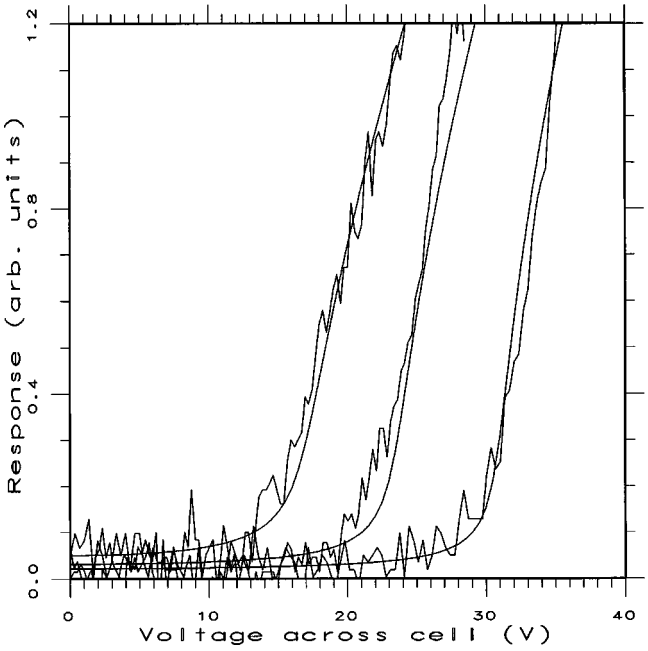

FIG. 2. Voltage-dependent behavior of the response (average tilt for theory and transmission change in arbitrary units for the experiment) around the smectic-*A* Freedericksz transition. The smooth lines are the theory and the noisy lines are the data. Results are shown (from left to right) at $T-T_{AC}=0°$, $T-T_{AC}=0.2°$, and $T-T_{AC}=0.5$ °C.

smectic-*C* phase because a mode of reorientation is now available through the Goldstone mode only. This involves elastic distortion and no layer perturbation. At the phase transition point the dominant terms (i.e., those which determine the threshold) in the above Euler-Lagrange equation also contain only the terms for director distortion. The consequence of this is that the threshold remains constant in the smectic-*C* phase close to T_{AC}.

(ii) Above the phase transition temperature the threshold voltage squared increases linearly with temperature. This is as predicted above, and the slope gives $\alpha d^2/[\varepsilon_0 \Delta\varepsilon(1+c)^2] = 1333 \pm 133\ \mathrm{V}^2/°$. Dividing this by the parameter determined above, and putting in typical values for K and c leads to $\alpha \approx 1850\ \mathrm{Nm}^{-2}/°$.

In practice the threshold is not sharp, especially near the smectic-*A* to smectic-*C* transition temperature. This is most likely to be due to finite surface pretilt in the device, which leads to a similar effect in the nematic Freedericksz transition. In order to investigate this, the Euler-Lagrange equation given above is solved numerically in the regime of the smectic-*A* Freedericksz transition, including a surface pretilt of $\approx 1°$ in the calculations. Experimentally, for small perturbations in tilt the expected optical response is proportional to the average tilt of the director in the device. These are compared in Fig. 2, where a modified coefficient of $4\pi^2 K/(\varepsilon_0 \Delta\varepsilon) = 320 \pm 36\ \mathrm{V}^2$, and an arbitrary scaling on the vertical axis are used. Clearly the comparison is very good, and confirms that the lack of sharpness (or rounding) at the threshold is due to a finite surface pre-tilt. Additionally there is a change in the parameter value due to surface tilt modification of the threshold voltage.

In summary, a smectic-*A* Freedericksz transition has been predicted to exist near the smectic-*A* to smectic-*C* phase transition point. A simple theory based on the need to retain smectic layer packing has allowed the variation with temperature of the threshold voltage for this transition to be determined. Comparison of these predictions with experiment has shown the correct threshold voltage at the smectic-*A* to smectic-*C* transition point, and the correct behavior with temperature. Additionally softening or rounding of the threshold has been shown to be due to surface pretilt, which also shifts the threshold voltage at the smectic-*A* to smectic-*C* transition point by around 10%.

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