"Soliton switch" in chiral smectic liquid crystals

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Solitary waves are observed to mediate the transition between the two states with opposite polarization of chiral smectics. For samples thicker than the pitch, electric fields as high as 10^6 V/cm do not completely unwind the helix of ferroelectric liquid crystals because the vanishing of domain walls in these layered phases, unlike in the cholesteric phase, requires the nucleation of an array of dislocations with a large Burger's vector. In addition, samples thinner than the pitch but still $\gtrsim 10$ μm are not bistable as we have experimentally verified.

In recent years, interest in liquid crystals has focused on the layered phases called smectics.[1] In smectic C, for example, the director $\vec{n}$, characterizing the preferred direction of the molecules, is tilted at an angle with respect to the layer normal. In chiral smectic C, the tilt direction rotates about an axis perpendicular to the layers giving rise to a constant macroscopic pitch (cf. Fig. 1) which is usually of the order 1–10 μm. If there is additional ordering in the layer planes,
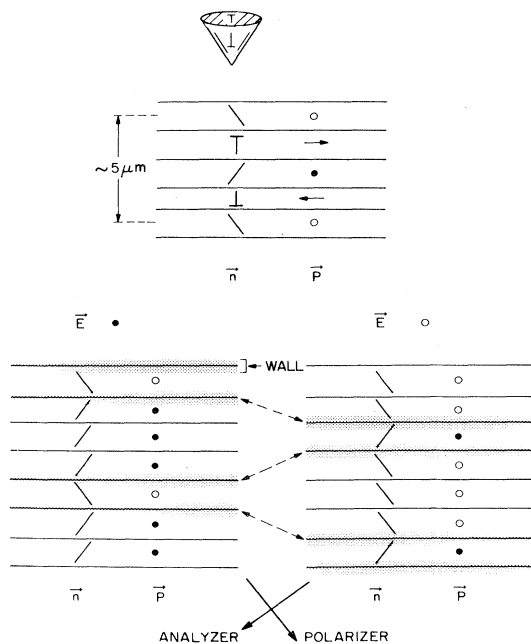

FIG. 1. Top: Schematic plot of a chiral smectic. The set of possible director orientations lies on a cone. There are typically $\approx 10^3$ smectic layers in a pitch. The director orientation ($\vec{n}$) is on the left, the short bar indicates the part of the director which is behind the plane of the cross section shown. The polarization is sketched on the right. Bottom: The two states shown by a chiral smectic viewed between crossed polarizers in an electric field. The walls are indicated as shaded lines.

one can find with decreasing temperatures C^* (Ref. 2), I^* (Ref. 3), F^* (Ref. 4), and G'^* (Ref. 5), phases. Meyer[1,6] pointed out that this kind of a structure (a chiral, tilted liquid-crystalline phase) implies the existence of a permanent polarization $\vec{P}$ perpendicular to both $\vec{n}$ and the layer normal. Thus $\vec{P}$ rotates also in a helical way.

When a dc electric field $\vec{E}$ is applied to a sample of a chiral smectic with a thickness larger than the pitch, regions where $\vec{P}$ is parallel to $\vec{E}$ grow continuously with increasing field[7] (see Fig. 1). At higher fields, regions where $\vec{P}$ is antiparallel to $\vec{E}$ become very small and are separated by walls from the regions where $\vec{P}$ is parallel to $\vec{E}$. Since the polarization is rigidly coupled to the director, there is an optical contrast between the two states when viewed between crossed polarizers. The tilt angle in many chiral smectics is close to 22.5° and thus there is no problem in obtaining high contrast.

In the following, we propose that although regions of unfavorable polarization become smaller as the field strength increases they never vanish completely even at high fields, i.e., the helix is never completely unwound.

Reversing the field, we observed that the walls moved in such a way as to maximize regions of favorable polarization. When a rectangular voltage pulse, connecting two states, is applied, the walls move as solitary waves. In this case, the switching time between the two states is found to be proportional to $1/\sqrt{V}$ in contrast to the $1/V$ dependence[6] expected for chiral smectics in the absence of soliton-type motion. When the field is turned off, the walls relax in the bulk in a time[6] $\tau \sim \gamma/K_2 q^2 \approx 5 \times 10^{-3}$ sec taking $\gamma = 1$ P, $K_2 = 2 \times 10^{-7}$ dynes for the Frank constant, and 3×10^4 cm^{-1} for q which is 2π times the inverse pitch.

Evidence for the motion of walls is most easily obtained by microscopic observations of a chiral smectic switching between the two states. In low fields (of order 1 V) pairs of surface lines which run parallel to the smectic planes are observed to separate, to move perpendicular to the layers, and, finally, to coalesce with their nearest neighbors (Fig. 1). The wall thickness ξ is given by $2(K_2/EP)^{1/2}$, which gives ~ 1.1 μm for $P \sim 10$ esu and $E \approx 2$ V/10 μm. At larger fields, the walls become too thin to be observable in the light microscope.

The continued existence of the walls is revealed when the

field is turned off. Surface disclinations, which form at regular intervals along the direction of twist[8] and which are no longer visible in high fields, reappear in the same place as before the field was turned on. This process takes place on a time scale of order 1 sec, being much larger than the relaxation time of the bulk and thus setting the time scale for the loss of the optical contrast.

These observations are in striking contrast to those made for the more fluid cholesteric liquid crystals which can be completely untwisted by an applied field. After being untwisted and upon removal of the field, the untwisted state can persist for times up to several seconds. Then the twisted state nucleates very slowly and gradually ($\approx 1-10$ min) fills up the sample. In addition, it is important to note that there is no memory effect. The new twisted texture is not a copy of the previous one as is the case for chiral smectics.

To understand the difference between making unfavorable regions vanishingly small in cholesterics as opposed to chiral smectics, one must consider how walls separating these regions from the favorable ones can end. In cholesterics, the wall ends with a simple disclination whereas the underlying layer structure of chiral smectics implies, in addition to disclinations, the nucleation of an array of dislocations with large Burger's vector since each wall is about 100 molecular lengths thick. The generation of dislocations of large Burger's vector requires a large amount of energy and provides strong support for our hypothesis that the helix in a chiral smectic does not unwind completely.

From the observed mechanism it is intuitively clear that a state is established for which the dissipative losses are compensated by the external driving force. If we denote the tilt angle by θ and the azimuthal phase of the director by ϕ and ignore the elasticity of the smectic layers, we obtain, upon minimizing the Frank energy, the following dynamic equation:

$$\theta^2 K \frac{\partial^2 \phi}{\partial z^2} - \theta EP \sin\phi - \gamma \frac{\partial \phi}{\partial t} \theta^2 + W = I \frac{\partial^2 \phi}{\partial t^2} \theta^2 \ , \qquad (1)$$

where W is the driving force, $I = \rho \xi^2$ the moment of inertia, ρ the density, and ξ the wave width. Equation (1) is a driven sine-Gordon equation with a damping term. The solutions of this equation have been discussed, e.g., by McLaughlin and Scott.[9] They find that (1) has solitary wave solutions which travel at a fixed speed u and with a constant pulse width ξ. The solitary wave motion occurs parallel to the layer normals. If there is a strong coupling between the layers, however, the director can no longer rotate freely and the above analysis would have to be modified.

The maximum soliton speed[9] C is obtained from the linearized form of Eq. (1), $C = (PE\theta/\rho)^{1/2}$, which yields $C \approx 30$ cm/sec for a sample of 12-μm thickness, $V \cong 100$ V, $P \cong 10$ esu, $\theta = 0.4$ rad, $\rho = 1$ g/cm^3. Damping γ and forcing W lead to a reduction of the velocity by an amount we estimate from the perturbation analysis of Ref. 9,

$$u = \frac{\pi}{2} C \frac{\sqrt{\rho K_2}}{\gamma} \ , \qquad (2)$$

which gives $u = 0.023 \sqrt{V}$ cm/sec with the use of the values cited above and $K_2 = 2 \times 10^{-7}$ dynes, $\gamma = 0.1$ P, and V is in volts.

For the switching time τ, we then take the ratio of u and the pitch p_0. Inserting $p_0 \cong 2 \times 10^{-4}$ cm and $V \sim 100$ V, $\tau \cong 1$ msec. In particular, we expect from Eq. (2)

$$\tau \propto \gamma/\sqrt{V} \ . \qquad (3)$$

Figure 2 shows a log-log plot of the switching time we measured in a 12-μm-thick sample of DOBAMBC (p-decyloxybenzylidene p'-amino-2-methylbutylcinnamate) by switching between the two states shown in Fig. 1 for various applied voltages. The open circles represent the raw data. A linear plot of τ vs $1/\sqrt{V}$ determined an intercept τ_0. ($\tau - \tau_0$) is shown as closed circles and its slope is $\approx 26°$ or $\tau - \tau_0 \propto 1/\sqrt{V}$.

There is no physical significance to τ_0 except to indicate that the regime of solitary wave propagation is sandwiched between two voltage regimes. At low voltages, solitons are not well formed and at high voltages other factors, such as, e.g., electrohydrodynamic instabilities in DOBAMBC, affect the switching times. We have also performed analogous measurements in I^* and G'^* phases and have found the $1/\sqrt{V}$ law to hold in these phases as well. The slope of τ vs $1/\sqrt{V}$ is about 100 times steeper in the G'^* phase than in the C^* and I^* phases, qualitatively verifying the role of viscosity in Eq. (3).

In smectic X, a probably solidlike phase which has very recently[10] been discovered to occur at lower temperatures than G'^*, director reorientation proceeds via a motion parallel to the layers implying a strong correlation between layers. In contrast to the chiral smectic phases described above, X is bistable (and achiral).

Recently, Clark and Lagerwall[11] have put forward the hypothesis that a sample of a chiral smectic material which has a thickness smaller than the pitch would be bistable. Since the pitch of chiral smectics in pure compounds is of the order $2-5$ μm (Ref. 7) they studied samples of a thickness of 1 μm to find support for their hypothesis. To test this hypothesis, however, it is possible to proceed more simply by doping an achiral smectic with a cholesteric material.[2] As a result, one easily obtains materials with a permanent polarization and with pitches ranging from $10-100$ μm which can be studied in the usual 10-μm cells. We prepared a number of samples, e.g., 2% cholesterol propionate in diheptylazoxybenzene, showing these properties.

The director was oriented in an applied electric field giving rise to good contrast between the two states. In large pitch materials reorientation proceeds via rolling of the director around its cone (cf. Fig. 1). This corresponds essentially to the approximation $\sin\phi \approx \phi$ in Eq. (1), and
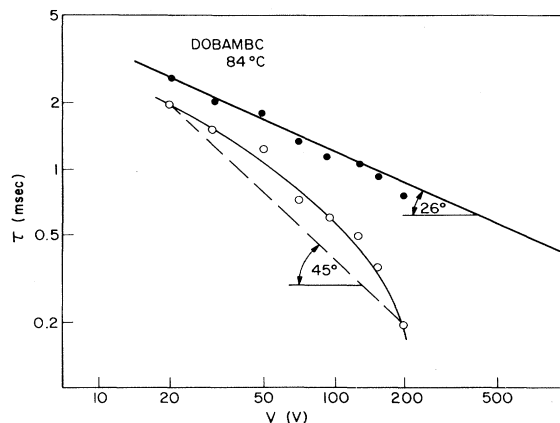

FIG. 2. τ vs V (O) and $\tau - \tau_0$ vs V (●) for DOBAMBC at 84 °C. τ vs $\sqrt{V}$ plot $\tau_0 = -5.74$ msec. The slope of the ($\tau - \tau_0$) plot is 26° verifying $\tau - \tau_0 \propto 1/\sqrt{V}$ as given by Eq. (3).

there are no domain walls. However, bistability was *not* observed, even in fields as high as 6×10^5 V/cm. Therefore we conclude that reduction of the sample thickness below the value of the pitch does not, in general, lead to bistability in ferroelectric smectic liquid crystals as claimed by Clark and Lagerwall.

To summarize, we have provided evidence for the fact that switching in ferroelectric chiral smectics proceeds via the motion of domain walls. In contrast to cholesterics, the termination of walls in chiral smectics requires the nucleation of dislocations. Furthermore, we have demonstrated that, in general, production of samples with a thickness smaller than the pitch does not lead to bistability in chiral smectics.

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