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<sup>4</sup>The zone-refined 4,4'-bis(heptyloxy)azoxybenzene was generously provided by Dr. J. T. S. Andrews, Liquid Crystal Institute, Kent State University.

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### BIAXIAL LIQUID CRYSTALS\*

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Convergent light observations have been made on three liquid crystalline substances with nematic and smectic phases. The nematic phase was observed to be uniaxial as expected, but smectic *C* was found to be biaxial. The optic axial angle  $2V$  for smectic *C* was found to be on the order of  $20^\circ$  for all three compounds. Also, the tilt angle for smectic *C* is nearly  $45^\circ$  for all three liquid-crystal systems. Anisotropy of the degrees of order is suggested as a partial explanation for the biaxial character of smectic *C*.

We have studied three compounds which exhibit a nematic phase and a single smectic phase of the type classified by Sackmann and Demus<sup>1</sup> as type *C*. The smectic phases classified by Sackmann and Demus correspond to different molecular arrangements and are separated from each other by first-order phase transitions.

The nematic phase has a long-range order such that the long axes of the molecules are nearly parallel and the parallelism of the long axis generally varies continuously throughout the bulk of the liquid. Assuming a random arrangement of the center of mass and free rotation about the long molecular axis, nematic liquids would be uniaxial. All observations do show the nematic phase to be uniaxial. Smectic *A* is a system in which the molecules are arranged in layers with the long molecular axis perpendicular to the layer.<sup>2</sup> If the centers of mass within the layers are random and free rotation is assumed, then smectic *A* would be uniaxial. Experimentally, all smectic-*A* phases have been shown, in general, to be uniaxial. Smectic *C* is generally considered to have a structure such that the molecules are arranged in layers but with the long molecular axis tilted with respect to the layer normal.<sup>2</sup> In general, it has been assumed that all smectic phases including smectic *C* would show uniaxial character and, in fact, Friedel<sup>3</sup> argued that all smectic phases would be uniaxial. Recently, Saupe<sup>2</sup> pointed out that due to symme-

try considerations, smectic *C* should be biaxial. The observations we report in this Letter show that smectic *C* phases are biaxial.

An alternate molecular arrangement, from x-ray data, for smectic-*C* phases has been proposed by Chistyakov *et al.*<sup>4</sup> Chistyakov proposes that instead of the direction of alignment being parallel throughout the sample, the direction of tilt is constant but alternates from layer to layer. This results in a herringbone type of molecular structure.

It has been reported by Arora, Fergason, and Saupe<sup>5</sup> that smectic-*C* phases can be oriented by surface action so that the layers are parallel to the surface. It was also reported that apparently the smectic-*C* phase of the compound bis-(4'-*n*-decyloxybenzal)-2-chloro-1,4-phenylenediamine (DOBCP) has a tilt angle of nearly  $45^\circ$ . Since the smectic *C* can be oriented, it was assumed that an area large enough for convergent-light observation could be uniformly oriented.

Optical observations upon compounds which have a smectic-*C* phase were made using a Leitz Panphot-pol polarizing microscope, equipped with a heated stage. Since the orientation of the layers of smectic *C* are parallel to the containing surfaces and the molecules are tilted with respect to the layers, the interference figure is off center if flat glass slides perpendicular to the microscope axis are used. In fact, the interference figure is so badly off center that little in-

formation can be obtained from convergent light observations. In order to center the interference figure, two  $45^\circ$  prisms of dimensions  $7 \times 7 \times 8$  mm were used as slides on the Leitz stage. The prisms were rubbed with paper parallel to the 8-mm dimension to orient the smectic *C* by surface action, as shown in Fig. 1. Since, with the prisms, a large objective working distance is needed, work with a small numerical objective was necessary. An objective of  $32\times$  magnification with a 0.4 numerical aperture was used for convergent light. The numerical aperture corresponds to an angular field of view of  $45^\circ$  in air.

The first compound examined was DOBCP since it was known that the smectic *C* had a tilt angle of approximately  $45^\circ$ ; with the  $45^\circ$  prisms, observations would then be made looking down the optic axis if smectic *C* was uniaxial. The first rather surprising observation was that in plane-parallel light between crossed polars, the

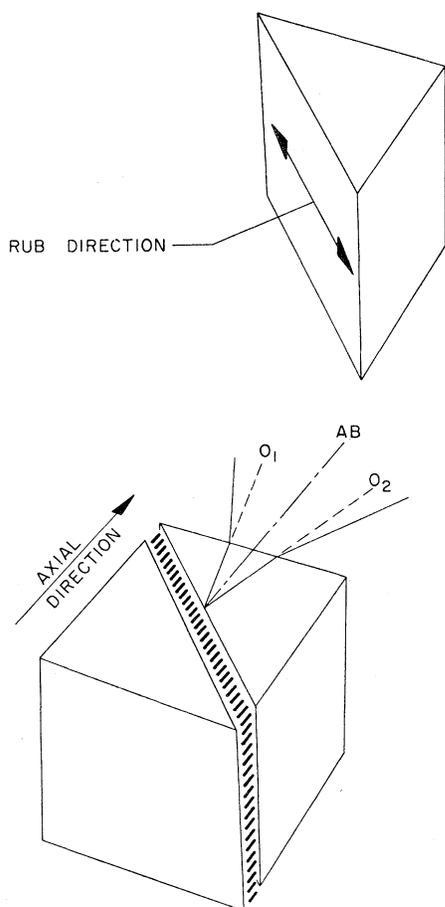


FIG. 1. Schematic representation of the method used to center the interference figure of oriented liquid crystals. *AB* is the acute bisectrix and the angle between rays  $O_1$  and  $O_2$  is the optic axial angle  $2V$ .

nematic phase of DOBCP showed no birefringence at all stage positions. This indicates that the observation is made looking down the optical axis and, hence, the molecular axis (optic axis) is at a  $45^\circ$  angle to the normal to the prism face. This result is rather surprising since it is generally assumed that the optic axis would be parallel to the rub direction of the prism faces. In convergent light, a perfect uniaxial cross was observed which did not change on rotation of the stage. The cross is slightly off center, but this is probably due to a mismatch between the indices of refraction of the liquid crystal and the glass prisms rather than the optic axis being at an angle other than  $45^\circ$  to the prism normal. The interference figure for the nematic phase of DOBCP is shown in Fig. 2. In order to obtain good orientation, thin layers of the liquid crystal were used and, hence, the birefringence is small and interference rings are not observed. The interference figure could be observed from the clearing point to the nematic-smectic-*C* transition and showed no changes over this temperature range. At the nematic-smectic-*C* transition temperature the uniaxial cross disappears and a split cross typical of a biaxial material appears. The interference figure shows a cross every  $90^\circ$  on stage rotation, and at  $45^\circ$  between extinction positions, the maximum splitting of the isogyres of the interference figure is observed. In the extinction position the biaxial cross appeared the same as the uniaxial cross and was off center the same amount. A typical interference figure for smectic *C* is shown in Fig. 3.

4-*n*-nonyloxybenzoic acid (NOBA) and 4, 4'-di-*n*-heptyloxyazoxybenzene (HOAOB) also show nematic and smectic-*C* phases. Convergent-

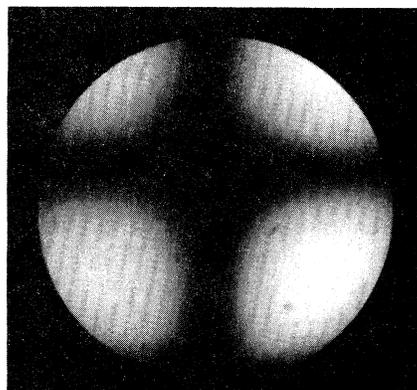


FIG. 2. Interference figure observed in the nematic phase of bis-(4'-*n*-decyloxybenzal)-2-chloro-1, 4-phenylenediamine.

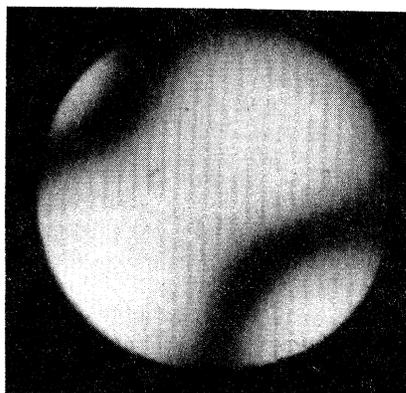


FIG. 3. Interference figure observed in the smectic-C phase of bis-(4'-*n*-decyloxybenzal)-2-chloro-1,4-phenylenediamine at 45° from extinction position. The optic axial angle is 18°

light observations on NOBA and HOAOB show the same behavior as for DOBCP; that is, a uniaxial cross is observed in the nematic phase and on transition to smectic C, the uniaxial cross becomes a biaxial cross. As for DOBCP, both crosses were off center the same amount. Observation with a quarter-wave plate showed that all nematic phases were optically positive. Use of a full-wave plate showed that all of the smectic-C phases were also optically positive. In all cases, the extinction position for smectic-C phases occurred when the rubbed direction of the prisms was parallel or perpendicular to the direction of polarization of the polarizer. This indicates that the optic axes are in a plane parallel to the rubbed direction and the surface normal. The angle between the optic axes ( $2V$ ) was calculated to be  $18 \pm 3^\circ$  for DOBCP and HOAOB and  $22 \pm 3^\circ$  for NOBA. The  $2V$  angle was calculated using Mallard's law<sup>6</sup> with aragonite ( $2V = 30^\circ$ ) as a calibration substance.

Since all the smectic-C phases observed were optically positive, Chistyakov's suggestion of a herringbone structure can be ruled out; Chistyakov made the suggestion after studying NOBA.<sup>4</sup> This conclusion can be made because in the herringbone model the molecular axes would be at  $90^\circ$  angles to one another and, therefore, the maximum index of refraction  $\gamma$  and the intermediate index  $\beta$  would have to be of the same order of magnitude. Thus, the herringbone structure would require that smectic C be optically negative. Also, the tilt angle for all three substances must be nearly  $45^\circ$ , whereas Chistyakov and Chaikowsky<sup>7</sup> find  $30^\circ$ , using x rays, for HOAOB. The value of  $45^\circ$  for HOAOB is in agreement with the value deVries<sup>8</sup> obtained with

x rays. The fact that all three substances showed tilt angles of  $45^\circ$  is itself surprising. Our studies on compounds having different terminal substituents and showing smectic-C phases suggest that smectic-C phases with relatively long-chain alkoxy terminal substituents have a tilt angle of about  $45^\circ$ .

The fact that smectic C is a biaxial liquid crystal could be explained in two ways: the anisotropic inner field, and an anisotropic degree of order  $S$ . Since the effect of the anisotropic inner field is very difficult to estimate, we will consider the possible effects of having an anisotropic degree of order. Since fluctuations in the direction of the tilt change the layer thickness, it seems reasonable to assume that fluctuations in the direction of the tilt are less than fluctuations in a direction perpendicular to the tilt axis. If we assume free rotation about the long axis, then by a method analogous to that used by Saupe and Maier<sup>9</sup> for uniaxial materials, we find that for no inner field correction,

$$\eta_\beta^2 - \eta_\alpha^2 = \frac{1}{3}(\eta_\gamma^2 - 1)(S_2 - S_1) = \frac{1}{3}(\eta_\gamma^2 - 1)\Delta S,$$

where we define the degree of order as  $S_i = \frac{1}{2}(3 \cos^2 \theta_i - 1)$  with  $\theta_i$  the angle between the  $\gamma$  axis and the direction of the long axis of the molecule; thus,  $\theta_1$  and  $\theta_2$  denote fluctuations parallel and perpendicular to the tilt direction. The intermediate index of refraction is  $\eta_\beta$ ;  $\eta_\alpha$  is the minimum index; and  $\eta_\gamma$  is the maximum index. If we assume  $\eta_\gamma \sim 1.7$ ,  $\eta_\beta \sim \eta_\alpha \sim 1.5$  from values for similar uniaxial liquid crystals, and  $\eta_\beta - \eta_\alpha \sim 0.01$ , then an anisotropy  $\Delta S \sim 0.05$  is obtained. Since  $\eta_\beta - \eta_\alpha \sim 0.01$  is the maximum difference conceivable for a  $2V$  of  $20^\circ$ , the  $\Delta S$  value of 0.05 is not unreasonable. For more realistic values of  $\eta_\beta - \eta_\alpha$ , smaller values of  $\Delta S$  would be obtained and, hence, it seems easily possible for there to be an anisotropy of  $S$  large enough to cause smectic-C phases to be biaxial. Therefore, at least a part of the biaxial character of smectic C may be due to an anisotropy ordering parameter.

For matters of completeness, the transition temperatures for DOBCP,<sup>5</sup> HOAOB,<sup>10</sup> and NOBA<sup>11</sup> are, respectively: nematic-isotropic transition, 166.5, 122.5, and  $143.0^\circ\text{C}$ ; nematic-smectic C transition, 112.5, 92.0, and  $117.0^\circ\text{C}$ ; and the smectic-C-solid transitions, 66.0, 74.0, and  $94^\circ\text{C}$ .

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versations about smectic phases.

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### FEEDBACK STABILIZATION OF HIGHLY CONDUCTING PLASMAS\*

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A general result for the feedback stabilization of surface instabilities of a highly conducting plasma is presented. Explicit form of feedback for stabilizing a linear pinch with a uniform axial static magnetic field is given.

Recently, various attempts have been made in using feedback to quench certain forms of plasma oscillations<sup>1-5</sup> and to stabilize drift waves in plasmas.<sup>6-11</sup> Marked improvement in plasma confinement has been reported in a number of experiments.<sup>4-8</sup> Here, we report the results of theoretical studies on the feedback stabilization of highly conducting plasmas with applications to linear pinch stabilization.

Consider a perfectly conducting plasma immersed in a static magnetic field whose motion is describable by the hydromagnetic equations with an isotropic stress tensor.<sup>12</sup> We shall assume that a static equilibrium exists but that it

is unstable. The feedback signal is in the form of a time- and space-dependent perturbation of the external magnetic field in the vacuum region surrounding the plasma about a given static field configuration. Since the time-varying component of the external magnetic field cannot penetrate into the plasma interior, it can only influence the plasma motion by varying the magnetic pressure on the plasma surface. The feedback, in effect, introduces a correction of the local magnetic field depending on the instantaneous deviation of the plasma surface from its equilibrium. Using the linearized hydromagnetic equations with linearized boundary condition,<sup>13</sup>

$$-\gamma p_0 (\nabla_0 \cdot \vec{\xi}) + \mu_0^{-1} \vec{B}_0 \cdot [\nabla_0 \times (\vec{\xi} \times \vec{B}_0)] + \frac{1}{2} \mu_0^{-1} \vec{\xi} \cdot (\nabla_0 |\vec{B}_0|^2 - \nabla_0 |\hat{B}_0|^2) = \mu_0^{-1} \vec{B}_0 \cdot (\delta \hat{B}_f + \delta \hat{B}_c), \quad (1)$$

it can be shown that a feedback correction field  $\delta \hat{B}_c$  at the vacuum-plasma boundary for local stabilization of surface instabilities is given by

$$\delta \hat{B}_c(t, \vec{r}_0) = K [\vec{n}_0(\vec{r}_0) \cdot \vec{\xi}(t, \vec{r}_0)] \hat{B}_0(\vec{r}_0), \quad (2)$$

where  $K$  satisfies

$$K \mu_0^{-1} \min_{\vec{r}_0 \in \partial D_0} |\hat{B}_0(\vec{r}_0)|^2 + \min_{\vec{r}_0 \in \partial D_0} \left\{ \nabla_0 \left( \frac{|\vec{B}_0(\vec{r}_0)|^2}{2\mu_0} \right) - \nabla_0 \left( \frac{|\vec{B}_0(\vec{r}_0)|^2}{2\mu_0} + p_0(\vec{r}_0) \right) \right\} > 0, \quad (3)$$

where  $\vec{r}_0$  denotes a point on the equilibrium plasma-boundary surface  $\partial D_0$ ;  $\vec{n}_0(\vec{r}_0)$  is the outward unit normal to  $\partial D_0$  at  $\vec{r}_0$ ;  $\vec{B}_0$  and  $\hat{B}_0$  are the equilibrium magnetic fields in the plasma and vacuum, respec-

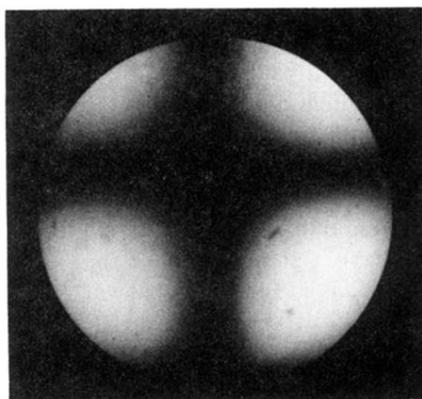


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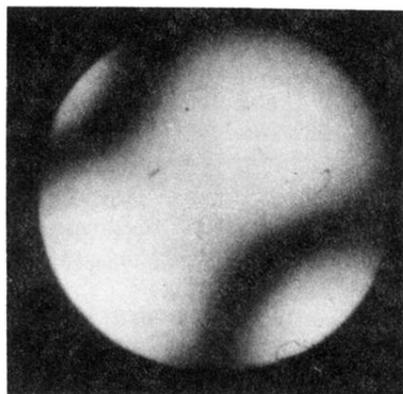


FIG. 3. Interference figure observed in the smectic-*C* phase of bis-(4'-*n*-decyloxybenzal)-2-chloro-1,4-phenylenediamine at 45° from extinction position. The optic axial angle is 18°