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ANOMALOUS ALIGNMENT IN THE SMECTIC PHASE OF A LIQUID CRYSTAL OWING TO AN ELECTRIC FIELD*

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Measurements of the dielectric loss in a liquid crystal, exhibiting both nematic and smectic phases, show an ordering with the long axes of the molecules preferring a direction papallel to both dc and 5000-Hz electric fields in the nematic phase. In the smectic phase the preferred direction is parallel to a 5000-Hz electric field and perpendicular to a dc field. The observed alignment of the smectic phase in a dc field is believed to be anomalous and it is suggested that this effect may be associated with the conductivity anisotropy.

Anomalous alignment in nematic liquid crystals owing to electric fields has been observed by many workers. Recent work[1-3] employing microwave dielectric techniques has shown that the process primarily responsible for this anomalous behavior is a well-behaved process. Interfacial polarization resulting from the conductivity anisotropy of clusters of molecules was suggested[1] as a mechanism that could produce an interaction with the electric field and account for the anomalous effect. This work has recently been treated theoretically by Helfrich.[4]

In the previously mentioned nematic materials the anomalous effect involves an ordering with the long axes of the molecules preferring a direction parallel to a dc electric field and perpendicular to an ac electric field for frequencies above the audio region. Results reported here show that in a smetic phase of ethyl-p-[(p-methoxybenzylidene)amino] cinnamate (hereafter referred to as EMC) the long axes of the molecules prefer a direction perpendicular to a dc electric field and parallel to a 5000-Hz electric field.

Vorlander[5] reported finding three liquid-crystalline phases in EMC, and these were later investigated by Demus and Sackmann,[6] and recently by Chistyakov, Schabischev, Jarenov, and Gusakova.[7] The work reported here involves only the nematic phase (118-140°C) and smectic phase A. The sample was purified by recrystallization from ethanol. An attempt to further purify by zone refining was not successful.

The experimental techniques were discussed previously.[1] Measurements of the dielectric loss in EMC at a microwave frequency of 24.5 GHz have been reported earlier.[8] The measurements of the dielectric loss in a magnetic field reported here are not identical to those reported earlier because of higher purity of the sample. Some of the effects mentioned previously[8] are now believed to be due to impurities.

Figure 1 shows measurements of the dielectric loss at a microwave frequency of 24.5 GHz as a function of temperature which were obtained by measuring the power transmitted through the sample while cooling from 120 to 110°C. When the sample was cooled in the presence of an external magnetic field of 10 000 G applied perpen-

dicular to the microwave electric field, an increase in absorption was observed as the sample passed into the smectic phase at about 118°C. It is believed that this increase is due to an increase in the degree of alignment with the long molecular axes parallel to the magnetic field. However, when the sample was cooled in the presence of a 10 000-G magnetic field applied parallel to the microwave electric field, the expected drop in the dielectric loss at $T = 118$°C was not observed. The well effects appear to be very significant in producing an ordering with the long axes of the molecules parallel to the walls of the container. This would tend to decrease the degree of alignment for a magnetic field applied parallel to the microwave electric field and thus prevent a significant drop in the dielectric loss at 118°C. Preliminary measurements employing NMR techniques support the suggestion concerning wall effects.

When an electric field is applied to the nematic phase of EMC an ordering with the long axes of the molecules parallel to the field is preferred for both dc and ac fields. A comparison of the relative effectiveness of electric and magnetic fields for producing molecular alignment showed that a magnetic field of 1000 G was comparable with an electric field of approximately 270 V/cm for both dc and ac fields. The measurements were made at $T = 125$°C using the methods reported earlier[1] and appeared to be independent of temperature in the nematic range.

Figure 1 shows measurements of the dielectric loss as the sample was cooled from 120 to 110°C with a dc electric field applied parallel to a 10 000-G magnetic field and the microwave electric field. As the value of the electric field was increased from 2000 to 5000 V/cm the dielectric loss showed an increase in the smetic phase. This implies an ordering with the long axes of the molecules preferring a direction perpendicular to the dc electric field. When either the electric field was increased to 7500 V/cm or the magnetic field was reduced to zero, the results were not appreciably different from those with a 5000-V/cm field applied parallel to the magnetic field. The relative effectiveness of the two fields is difficult to establish because of wall effects, but one would probably need an electric field of at least 3000 V/cm to predominate over a 10 000-G magnetic field. In addition, measurements of the dielectric loss show that a high magnetic field instigates a greater degree of ordering with the long molecular axes perpendicular to the microwave electric field than a high dc electric field.

Figure 2 shows measurements of the dielectric loss in EMC which were obtained while cooling from 120 to 110°C in the presence of a 5000-Hz electric field applied parallel to the microwave electric field and perpendicular to a 5000-G mag-

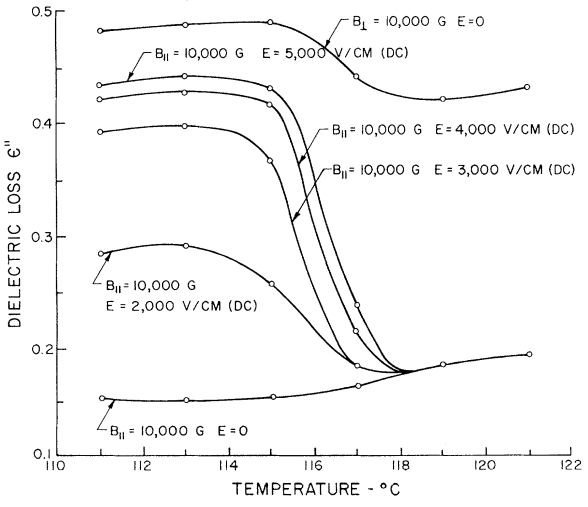

FIG. 1. Dielectric loss in EMC at a microwave frequency of 24.5 GHz as a function of temperature. A magnetic field of 10 000 G was applied parallel and perpendicular to the microwave electric field. dc electric fields of 2000, 3000, 4000, and 5000 V/cm were applied parallel to a 10 000-G magnetic field and the microwave field.

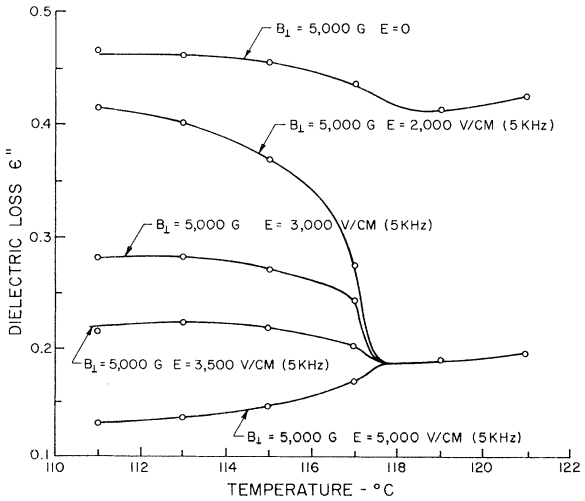

FIG. 2. Dielectric loss in EMC at a microwave frequency of 24.5 GHz as a function of temperature. A magnetic field of 5000 G was applied perpendicular to the microwave electric field. 5-KHz electric fields of 2000, 3000, 3500, and 5000 V/cm were applied perpendicular to the 5000-G magnetic field.

netic field. As the value of the electric field is increased the dielectric loss becomes less, which implies that an ordering in the 5000-Hz field is preferred with the long molecular axes parallel to the electric field. Because of wall effects, the relative effectiveness of electric and magnetic fields is difficult to estimate, but 5000 G is probably equivalent to an electric field between 2000 and 3000 V/cm. This implies that the relative effectiveness of a 5000-Hz field compared with a magnetic field is less in the smectic than in the nematic phase. Measurements at other frequencies indicated that the results were independent of frequency in the neighborhood of 5000 Hz. This implies that the ordering due to the 5000-Hz field was due only to the dielectric anisotropy. The absence of low-frequency dielectric measurements in this material makes it difficult to further discuss the dielectric anisotropy.

Preliminary measurements of the conductivity showed that the conductivity was greatest parallel to the long axes of the molecules in the nematic phase, but in the smectic phase the conductivity of the sample was at least 2 times greater in a direction perpendicular to the long axes when a dc voltage of 20 V was applied. It was difficult to establish the direction for maximum conductivity in the smectic phase employing ac fields with the available equipment, but this will be checked later. It is possible that charge carriers can move along the planes between layers of molecules more easily than perpendicular to the planes. The anomalous effect in the smectic phase seems to be consistent with the anomalous effect reported earlier[3] for the nematic phase in that the preferred ordering is in a direction such that the conductivity is a maximum, and it is possible to obtain a higher degree of alignment in a high magnetic field than in a high dc electric field. Measurements employing an ac electric field of approximately 100 Hz showed a tendency for the molecules to favor an orientation with their long axes perpendicular to the field, which implies that the anomalous effect is frequency dependent at very low audio frequencies. This is consistent with the results on nematic materials, in that a frequency dependence was also found in the audio region.

A model employed by the author which suggested that the experiment reported here might show an anomalous effect is illustrated in Fig. 3 of an earlier article.[3] This is a very crude model and lacks detail, but it may provide an idea for a mechanism that allows for an interaction involving external electric fields. The clusters of molecules, which are illustrated in this diagram, and aligned with their long axes parallel to each other within a cluster but are aligned at an angle with the long molecular axes of an adjacent cluster, should resemble a smectic rather than a nematic phase. Since the conductivity in the smectic phase is greatest perpendicular to the long molecular axes rather than parallel as in the nematic phase, the interfacial polarization at the boundaries between clusters owing to conductivity anisotropy will be reversed from that shown in Fig. 3(b) of the earlier article.[3] Since the dielectric constant in the material reported here is the greatest parallel to the long molecular axes, the polarization in Fig. 3(c) of the earlier article[3] owing to the dielectric anisotropy will also be reversed. By reversing the polarization for both the conductivity and dielectric anisotropy, the model can be applied to the smectic phase, and two competing aligning processes are illustrated. The preferred direction will depend on which process predominates and can be frequency dependent as suggested for nematic materials.[3]

The response time for a dc electric field in the smectic phase is much longer than in the nematic phase. The data for Fig. 1 were obtained by cooling at the rate of approximately 0.2°C/min between 114 and 118°C. It is believed that the results reported here are sensitive to the amount and type of impurity in the sample, and that results on a sample prepared by other techniques might show some variation.

The results reported here for a dc electric field appear to be consistent with those reported by Chistyakov, Schabeschev, Jarenov, and Gusakova[7] in that it is possible for the molecules to align with their long axes parallel to a dc field in the nematic phase and perpendicular to the field in the smectic phase.

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