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MOLECULAR RELAXATION IN HIGHLY ORDERED ORTHOGONAL SMECTIC S_A , S_B AND S_F PHASES

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ABSTRACT: Dielectric relaxation and calorimetric investigations were performed on different smectic phases. Discontinuities of the critical frequency were found at both S_A – S_B and S_B – S_E phase transitions. S_A and S_E structures can be regarded as two-dimensional liquid and two-dimensional crystal, respectively. The low frequency dielectric dispersion presented here demonstrates the difference between the S_E and a three-dimensional crystal where this phenomena is absent.

INTRODUCTION. - The characteristic low frequency dielectric relaxation phenomenon is well known in nematic liquid crystals appearing as a consequence of the long range orientational ordering of the molecular long axes and disappearing in the isotropic liquid and solid state. This relaxation process is connected with the rotational motion of the molecules around their short axes 1 . It was found $^{2-4}$ that this low frequency dielectric dispersion is present in some smectic liquid crystals, such as S_A where the elongated molecules oriented parallel to each other form layers but the structure is liquid-like inside the layer; S_B which is characterized by a hexagonal pattern of the

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centres of gravity of the molecules with a free rotation around the molecular axis.

In the present communication we concentrate on the S_E phase, which represents a two-dimensional crystal with a herring-bone structure formed by the planes of the molecules (see Fig. 2). The question arises about the difference between the molecular dynamics in the S_B and S_E phases.

$$C_{8}^{H}_{17}^{O-}$$
 $OOC_{2}^{H}_{5}$

which has

$$Cr \xrightarrow{348 \text{ K}} S_E \xrightarrow{356 \text{ K}} S_B \xrightarrow{367 \text{ K}} S_A \xrightarrow{383 \text{ K}} I$$

phases and was synthesized in our laboratory. The thermal properties were studied by polarizing microscope and a Perkin-Elmer DSC-2 calorimeter. The dielectric relaxation was measured in a wide frequency range up to 250 MHz. The calorimetric run is shown in Fig. 1 from which the high peak at the S_A - isotropic transition is easily apparent. The measured transition heats are $\Delta H_{AI} = 9.65^{+}_{-}0.18 \text{ kJ/mole}, \ \Delta H_{BA} = 1.56^{+}_{-}0.28 \text{ kJ/mole}$ and $\Delta H_{EB} = 2.45^{+}_{-}0.18 \text{ kJ/mole}$ for the S_A - Iso, $S_B - S_A$ and $S_E - S_B$ phase transitions respectively.

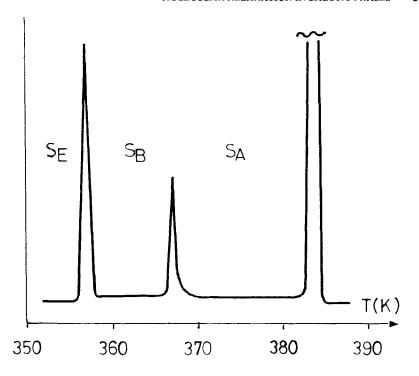


FIGURE 1 DSC trace of the compound

The dielectric critical frequencies connected with the rotational motion of the molecule around its short axis are shown in Fig. 2.

Apart from the jump about a factor of 3 in f_C at the S_A - S_B transition (which is less pronounced than the effect registered in 2) there appears a very intensive change (x20) at the S_B - S_E transition which was not observed in 5 . Contrary to the usual decrease towards the more ordered phases we found a strong increase in the activation energy from S_A to S_B .

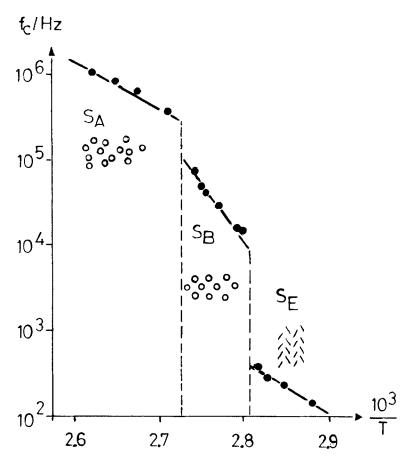


FIGURE 2 Arrhenius plot of the critical frequency

experimental points;

mean square fit

<u>DISCUSSION.</u> - The measured high values of the transition enthalpies show that both S_A - S_B and S_B - S_E transitions are of first order. Discontinuities in the critical frequency indicate a hindrance increasing by steps of the rotational motion. Decrease of f_C by a step at the S_A - S_B phase transition was interpreted by the appearance

of a collective libration in the two-dimensional hexagonal layer 3 . The further decrease of f_c found at the S_B - S_E transition is connected with the creation of the herring-bone structure. Whereas in the S_B phase librational waves can be created in any direction in the layers, in S_E this collective motion becomes anisotropic due to the structure formed by the planes of the molecules.

X-ray investigations show a close similarity between the S_E and crystalline phases. The measured here relaxation times in S_E (τ = 1/2 πf_C) show a prominent difference between them. This relaxation phenomena is frozen in solid phase.

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