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## Electrohydrodynamic instability and anisotropy of the electrical conductivity in the smectic *A* phase of a liquid crystal

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The phenomenon of electrohydrodynamic instability has been detected in the smectic *A* phase of a liquid crystal with positive dielectric anisotropy. The threshold, frequency, and contrast characteristics have been investigated both for this instability and for the electrically induced confocal-homotropic transition. The variation of the parameters of EHD instability and of the confocal and homotropic textures has been compared with the rules of variation of the electrical conductivity of the corresponding structures. A possible mechanism for the onset of instability is in many respects analogous to the Carr-Helfrich mechanism for nematic liquid crystals.

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The possibility of an onset of field-induced instability in the smectic *A* phase of liquid crystals (LC) is predicted by theory both in weak<sup>[1]</sup> and in sufficiently strong<sup>[2]</sup> electromagnetic fields. In the first case, because of the small value of the deformation, the effect has not yet been confirmed experimentally, whereas in fields of large intensity an electrically induced transition has been observed in a number of smectic liquid crystals (SLC).<sup>[3]</sup> The occurrence of electrohydrodynamic (EHD) instability in SLC is also predicted theoretically,<sup>[4]</sup> but experiments confirming this phenomenon are not, to our knowledge, reflected in the literature.

In the present paper, we report results of an investigation showing the presence of EHD instability in the smectic *A* phase, and we compare the behavior and peculiarities of the observed instability with the rules of variation of the electrical conductivity in SLC.

Chosen as object of investigation was 4-nitrophenyl-4-octyl oxybenzoate, which possesses both smectic and nematic mesophases and passes from the solid crystal following sequence of temperatures: SC 49 °C SLC-A (SC) state to the isotropic liquid (IL) state in the 61 °C NLC 68 °C IL. The smectic phase formed on cooling extended to ~33 °C. On slow transition from the isotropic phase to the nematic liquid crystal (NLC) phase, the LC molecules aligned themselves predominantly perpendicular to the surfaces of the electrodes, producing a homotropic orientation, which persisted also in the smectic state.

Application to the sample of a low-frequency (~20 Hz) voltage above a certain threshold value ( $U_{th}$ ) causes the appearance in individual parts of the cell of nuclei of turbulent motion, which, after spreading, fill the whole field of view. The rate of spreading of the turbulence depends on the temperature of the sample and on the value of the applied voltage. Within the interval 35 °C <  $t$  < 55 °C, 0 <  $U - U_{th}$  < 15 V the numerical values of these quantities satisfy the empirical relation

$$V[\mu\text{m/sec}] = (1.8 - t_{tr}/t)(U - U_{th}),$$

where  $t_{tr}$  is the temperature of the transition SLC-A → NLC.

The value of  $U_{th}$  increases with lowering of temperature and with increase of the frequency of the applied field (Fig. 1). Each temperature has its own "critical" frequency ( $f_{cr}$ ), above which EHD instability does not occur. With increase of temperature,  $f_{cr}$  shifts toward high frequencies. It should be noted that the smectic phase formed on heating a solid crystal has a predominantly planar orientation of the molecules and that the  $U_{th}$  of the corresponding texture is somewhat below the threshold for the homotropic texture.

The optically transparent homotropic structure is destroyed in an electric field, and in the process, turbulence severely scatters the transmitted light. After the applied voltage is turned off, a stable confocal texture is formed, and a scattering condition persists. The relaxation time of such a state is very

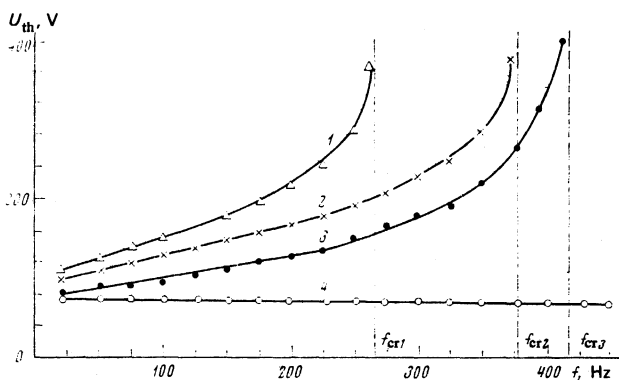


FIG. 1. Frequency dependence of the threshold voltage for EHD instability (1-3) and of the confocal-homotropic transition (4);  $d = 15 \mu\text{m}$ . 1 and 4,  $36^\circ\text{C}$ ; 2,  $45^\circ\text{C}$ ; 3,  $53^\circ\text{C}$ .

long, and consequently we may speak of an electrically induced memory effort in SLC.<sup>[5]</sup> The substance under investigation has a positive value of the dielectric anisotropy, and therefore the reverse confocal-homotropic transition can also be produced by an electric field above a certain threshold value (Fig. 1, Curve 4). The threshold of this transition is only slightly frequency-dependent for  $f > f_{cr}$  and increases somewhat for  $f < f_{cr}$ .

As is evident from the figure, it is possible to choose a fixed frequency such that with increase of voltage one can pass successively from the confocal texture through the homotropic again to EHD instability. The accompanying change of intensity of the transmitted light is shown in Fig. 2. The indicated transitions *A* and *B* correspond to a change of the current through the sample; for  $f < f_{cr}$ , the current decreases at the first transition point and increases by a small amount at the second (Fig. 2, Curve 2).

The low-frequency volt-ampere characteristic (VAC) consists of two regions with different slopes (Fig. 3a). The first region corresponds to the homotropic texture, the second to the development of EHD instability. The onset of turbulent motion is accompanied by a discontinuous increase of the current; with increase of the frequency of the applied field, this jump shifts toward higher voltages. This relation is in agreement

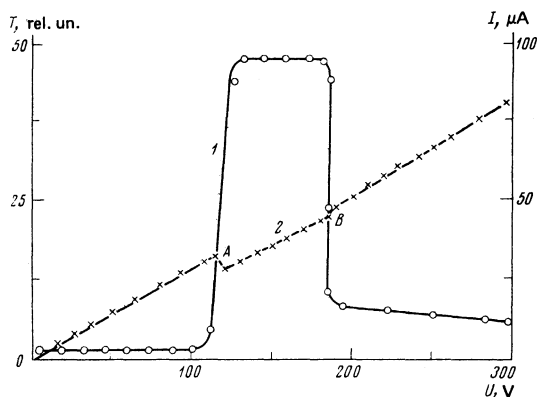


FIG. 2. Variation of intensity  $T$  of transmitted light (1) and of current (2) with applied voltage. Initial state, confocal texture;  $t = 53^\circ\text{C}$ ,  $d = 15 \mu\text{m}$ ,  $f = 200 \text{ Hz}$ .

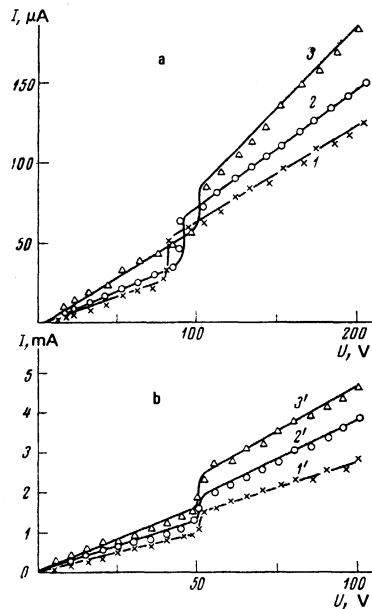


FIG. 3. Volt-ampere characteristics. Initial texture: a, homotropic; b, conformal. 1, 20 kHz; 2, 100 kHz; 3, 200 kHz; 1', 6 kHz; 2', 8 kHz; 3', 10 kHz.

with the frequency dependence of the threshold for EHD instability.

The form of the section of the VAC that corresponds to the confocal-homotropic transition depends on the frequency of the exciting field. While for  $f < f_{cr}$  the current decreases in the transition range, for  $f > f_{cr}$  there is observed at this point an abrupt increase of the current, in response to destruction of the confocal texture and homotropic alignment of the molecules. In this case the location of the "step" on the VAC is independent of frequency.

The observed features of the VAC can be explained on the basis of a difference in the mechanisms of the electrical conductivity  $\sigma$  corresponding to one or another range of frequency. At low frequencies, the value of  $\sigma$  is primarily determined by the value of the mobility of the charge carriers. It is known<sup>[6-9]</sup> that in SLC the mobility component along the smectic layers is larger than the component perpendicular to the plane of a layer. A SLC in the process of turbulent motion, and also a smectic confocal structure, can be regarded as a collection of individual, irregularly arranged microregions, within which short-range smectic order persists. Therefore the local mobility changes from section over the whole sample. Nevertheless the mobility averaged over the whole volume of the sample, and correspondingly the electrical conductivity, of the scattering state ( $\sigma_{sc}$ ), corresponding to turbulent motion or to a confocal texture, will be larger than the electrical conductivity of the transparent state ( $\sigma_{tr}$ ), corresponding to a homotropic structure. The measurements of the VAC at the low-frequency confocal-homotropic transition (Fig. 2, point *A*) and at the onset of EHD instability (Fig. 2, point *B*; Fig. 3a) support the arguments set forth above.

At relatively high frequencies, the value of  $\sigma$  is primarily determined by the value of the permittivity measured in the direction of the applied field. For the SLC under investigation,  $\epsilon_{\parallel} > \epsilon_{\perp}$ ; consequently,  $\sigma$  for the homotropic structure should be larger than  $\sigma$  for the confocal texture or for turbulent motion. In other words, at high frequencies the electrical conductivity  $\sigma_{tr} > \sigma_{sc}$ , as is confirmed by the form of the corresponding VAC (Fig. 3b).

The parameter  $\Delta\sigma = \sigma_{tr} - \sigma_{sc}$  can serve as a criterion of the electrical-conductivity mechanism. The frequency dependence of the reduced value of this parameter,  $\Delta\sigma/\sigma$ , reflects well the sequence of observed changes (Fig. 4). It should be noted that the frequency  $f_0$  at which the electrical conductivity of the transparent and scattering states is the same coincides approximately with the critical frequency  $f_{cr}$  of the threshold for EHD instability. The numerical values of  $f_{cr}$ ,  $f_0$ , and  $\sigma$  vary with temperature, but over the whole range of existence of the smectic A phase, they satisfy the relation

$$f_0 \approx f_{cr} = 24.8\sigma/\epsilon_0,$$

where  $\epsilon_0$  is the electrical constant.

The electrical conductivity varies with temperature according to an exponential law (Fig. 5) and can be characterized by the values of the activation energies  $E$  for the various phase states of the LC. In the isotropic-liquid and nematic phases, the activation energies are the same for the structures obtained in the heating and cooling processes and are, respectively,  $E_{IL} = 0.35$  eV/deg and  $E_{NLC} = 0.59$  eV/deg. For the smectic phase, however, the values of  $E$  corresponding to the structures formed on heating and on cooling of the sample are significantly different. On heating of this substance, as has been indicated, the smectic A phase has a predominantly planar texture. The activation energy then determined coincides with  $E_{IL}$  and is 0.35 eV/deg. In a cooling cycle, a structure with homotropic orientation of the molecules is formed; the corresponding activation energy is close to  $E_{NLC}$  and is 0.61 eV/deg.

It should be mentioned that it was noticed earlier,<sup>[7]</sup> for two smectic phases, that values of  $E$  obtained from measurements of  $\sigma$  in the direction parallel to the

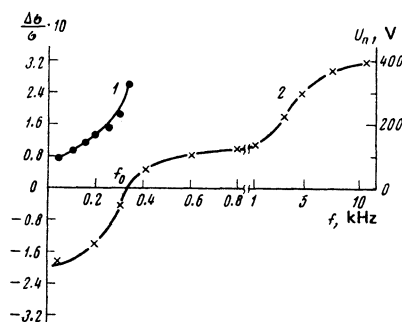


FIG. 4. Frequency dependence of: 1, the threshold voltage for EHD instability; 2, the parameter  $\Delta\sigma/\sigma$ ;  $t = 40^\circ\text{C}$ ,  $d = 15 \mu\text{m}$ .

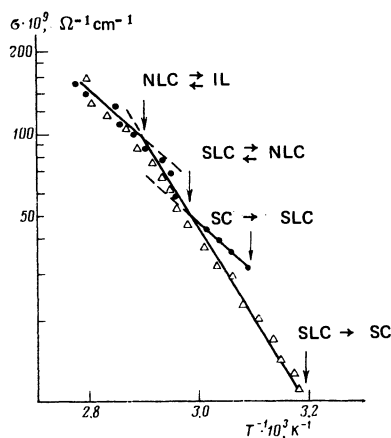


FIG. 5. Temperature dependence of the electrical conductivity of 4-nitrophenyl-4-octyl oxybenzoate.

orientation of the molecules were higher as compared with  $E$  determined in the perpendicular direction.

The mechanism of onset of EHD instability in a smectic A phase is apparently in many respects analogous to the Carr-Helfrich mechanism for NLC!<sup>[10-12]</sup> In the present case, the initial prerequisite for the onset of instability may be considered to be the presence of anisotropy of  $\sigma$  and the destruction of the parallel orientation of smectic layers with homotropically aligned molecules. Such a distortion may originate, in particular, from the presence of some micro-defect on the surface of the glass plates. The strong interaction between the layers promotes the successive transmission of the deformation to the interior of the LC volume. The large value of the components of  $\sigma$  along the smectic layers causes a distribution of space charge in accordance with the Helfrich<sup>[11]</sup> model. The field of the space charge, added to the external electric field, leads to the appearance of a torque and to a local onset of turbulence. Under the microscope it is seen that when the switching on is repeated, the instability always originates at the same places and, as it develops, covers the whole area of the cell. If the LC is kept in an

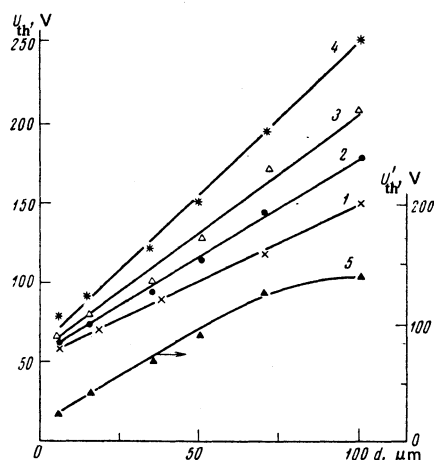


FIG. 6. Variation with sample thickness of the threshold voltage for: 1-4, EHD instability; 5, conformational-homotropic transition;  $t = 53^\circ\text{C}$ ; 1, 20 kHz; 2, 50 kHz; 3, 100 kHz; 4, 150 kHz; 5, 5 kHz.

alternating field for a long time,  $U_{th}$  increases somewhat. Apparently this is due to some smoothing out of the distortion and to a corresponding diminution of the amount of the space charge. At high frequencies, the space charge lags in phase behind the applied voltage in the direction perpendicular to the field. Therefore with increase of the frequency of the external field,  $U_{th}$  increases.

While having a number of common features, EHD instability in the smectic A phase differs from its analog in the nematic state. First, motion of a smectic liquid at once acquires turbulent character, and destruction of the layered structure occurs without the rotary flow that is usually observed in the form of nematic Williams domains. Second, in contrast to instability in NLC,  $U_{th}$  in SLC depends on the thickness  $d$  of the sample (Fig. 6). There is a relation of direct proportionality between  $U_{th}$  and  $d$ , in confirmation of theoretical calculations (see formula (9) of Ref. 4). At the same time, the threshold of the conformational-homotropic transition is proportional to  $\sqrt{d}$  (Fig. 6, Curve 5).

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## Variation of the connectivity of the electron constant-energy surface in Bi under pressure

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The Shubnikov-de Haas (SdH) effect in single-crystal samples of Bi and Bi-Te alloys is investigated at hydrostatic pressures,  $p$ , of up to 20 kbar in magnetic fields of up to 60 kOe at helium temperatures. It is found that hydrostatic pressure induces a transition of the electron constant-energy surface (ECES) from a quasi-ellipsoidal to a dumbbell-like shape and then to a doubly connected surface. A magnetic-field-induced change in the connectivity of the ECES is observed in the region of pressures where the cross section of the neck of the dumbbell becomes sufficiently small. The shape of the ECES at different pressures  $p$  is established from the angular dependences of the SdH-oscillation frequencies. The obtained pressure dependences of the extremal cross sections,  $S$ , of the ECES are discussed on the basis of the McClure band spectrum model for materials of the Bi type. The computed  $S(p)$  functions agree with the experimental functions if it is assumed that the spectrum at the  $L$  point of the Brillouin zone is inverted and  $\epsilon_{gL} \sim -7$  meV. It is found that the parameter ratio  $Q_{22}^2/\alpha_c < 0.0005$  a.u. in the McClure model.

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### INTRODUCTION

The band structure and the Fermi surface (FS) of the current carriers in Bi have been investigated in a large number of experiments by different methods (comprehensive lists of references on this question are given in Fal'kovskii's<sup>[1]</sup> and Édel'man's<sup>[2]</sup> review articles). The data obtained in the investigations on Bi were up until very recently interpreted on the basis of two different band-spectrum theories: the Lax theory<sup>[3]</sup> and the Abrikosov-Fal'kovskii (AF)<sup>[4,5]</sup> theory. It follows from Lax's two-band model that the electron constant-energy surfaces (ECES) in Bi are ellipsoidal, while the electron

and hole spectrum at the  $L$  point of the Brillouin zone is a mirror spectrum and is the same whether the spectrum at the  $L$  point is inverted ( $\epsilon_{gL} \equiv \epsilon(L_a) - \epsilon(L_s) < 0$ ; the bottom of the conduction band is formed by the  $L_s$  term, while the bottom of the valence band is formed by the  $L_a$  term, it being then possible for saddle points to exist in the spectrum) or direct ( $\epsilon_{gL} > 0$ ; the spectrum cannot contain saddle points).

In principle, the Lax model cannot explain the experimentally established deviation<sup>[6,7]</sup> of the ECES in Bi from the ellipsoidal shape. The AF model satisfactorily describes the angular dependences of the cyclotron