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THE ULTRASONIC INVESTIGATION OF MAGNETIC FIELD INDUCED STRUCTURAL CHANGES OF A SMECTIC- C PHASE.

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Abstract The experimental research results of orientational structure changes in smectic-C phase with the magnetic field applied are presented. Variations of the acoustical parameters (attenuation coefficient α and ultrasound velocity c) were registered as functions of the magnetic field in various conditions. The different models of smectic-C were applied to explain the experimental results.

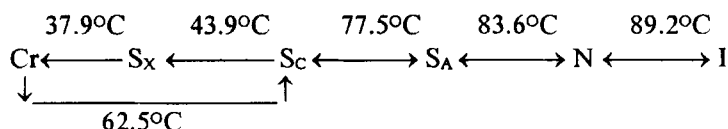
INTRODUCTION

The smectic-C phase (S_c) is of special interest because its orientational structure can be altered (rotation of the local C-director or the projection of the director \mathbf{n} on the layer plan) easily enough under the action of a moderate magnetic field. Compared with nematics the study of molecular alignment in the smectic-C phase and action of the magnetic field seems to be more complicated and more interesting. The existence of the layer structure and thus the limitation of the director displacement leads to various possible reactions of the sample under the action of the magnetic field for different experimental conditions. There are a few publications devoted to models and experimental study of smectic-C phase by acoustic method.¹⁻⁶ The experimental data have clearly indicated efficiency of the ultrasound method for studying complicated changes in orientational structure of various liquid crystal modifications. The method being employed gives a high degree of accuracy and sensibility.

In this paper we report the behaviour of the S_c phase of thermotropic liquid crystal in the magnetic field by means of the ultrasound method.

MATERIALS AND METHODS

We have investigated 4-n-hexyloxyphenyl-4-n-decyloxybenzoate having the following phase sequence:



The smectic-C sample was prepared by slow-speed cooling it from the nematic phase to the temperature of 65°C with the magnetic field applied. The field of induction \mathbf{B}_0 was used for an orientation of the nematic phase of liquid crystal sample and was fixed during the cooling of the sample starting from the nematic phase through the smectic-A to the smectic-C phase. In our experiments \mathbf{B}_0 was always directed along ultrasound wave vector \mathbf{q} . We used the field of induction \mathbf{B} to change the structure of smectic-C. Values both \mathbf{B} and \mathbf{B}_0 could be changed to induce the orientational variations of molecules in the smectic-C. After preparing the S_c sample the magnetic field \mathbf{B}_0 was turned off and relative variations of longitudinal ultrasound attenuation coefficient $\Delta\alpha/f^2 = \{\alpha(\mathbf{B}) - \alpha(0)\}/f^2$ and velocity $\Delta c = c(\mathbf{B}) - c(0)$ were registered as functions of the magnetic field \mathbf{B} which was directed along or normal to wave vector \mathbf{q} . The measurements were carried out by modified pulse-phase method at the frequency $f=3\text{ MHz}$.⁷ The measured cell consists of two quartz transducers 15 mm in diameter of X-cut plates. The transducer spacing was 30 mm. Hence the sample volume was $\approx 5.3\text{ cm}^3$. The method accuracy was $\approx 10^{-3}\%$ and 2% for measuring the ultrasound velocity and the attenuation coefficient variations respectively.

RESULTS AND DISCUSSION

The simplest smectic-C model describes the monodomain layer structure $\mathbf{m}=\mathbf{B}_0/B_0$ (\mathbf{m} - layer normal) with the director reorienting along the slant side of the cone. Note that for smectic-C prepared from a nematic in the very strong magnetic field the polydomain model seems to be more suitable.^{1,2} In this model the director is restricted to a cone with the apex angle of 2Ψ , and the axes of the various domain cones are randomly distributed on an alignment cone, which has its axes along the magnetic field direction \mathbf{B}_0 and also has the apex angle of 2Ψ . Therefore,

smectic layers are inclined relative to \mathbf{B}_0 direction, and one obtains a polycrystalline material. Domains are considered to be sufficiently large so that thermal motion cannot destroy their orientation by the magnetic field. In general, in a polydomain material the distribution of layer normals of the domains can be more complicated and varies with temperature.

The experimental dependencies of $\Delta\alpha(B)/f^2$ are plotted in Fig.1. The data indicate the increasing of the magnetic field to cause the significant changes of the attenuation coefficient for the two orientation \mathbf{B} and \mathbf{q} when $B > 0.15$ T. For a small magnetic field (less than 0.15 T) the parameter $\Delta\alpha(B)/f^2$ practically does not depend on B . We should note that $B = 0.15$ T corresponds to the magnetic field of the molecular alignment saturation in the nematic phase. When field is being changed in nematics the similar variations of attenuation coefficient begin at a much less magnetic field ($B \approx 0.01$ T). This situation may be explained by large activation energy of molecular alignment in a smectic-C as compared to a nematics.

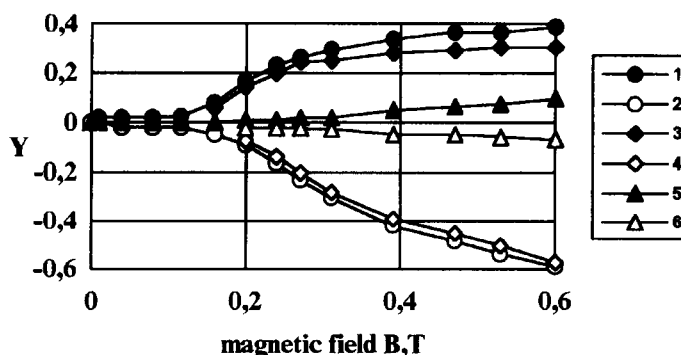


FIGURE 1 Dependencies of $\Delta\alpha/f^2$ on magnetic field B :
 $Y = \Delta\alpha/f^2$, $10^{-12} \text{ m}^{-1} \text{ s}^2$.
 B_0 : 1 - 0.6 T, 3 - 0.16 T, 5 - 0 T, (\mathbf{B} along \mathbf{q})
 B_0 : 2 - 0.6 T, 4 - 0.16 T, 6 - 0 T (\mathbf{B} normal to \mathbf{q}).

If the sample is cooled from the nematic phase into the smectic-C phase in the absence of the magnetic field one should expect a spherical symmetry of domain orientation, i.e. all directions of the layer normals should be equally probable. In this case the weak dependence of attenuation coefficient versus the magnetic field is observed (Fig.1). The data in Fig.1 probably also testify that the sufficiently large smectic-C sample prepared in the magnetic field as described above

cannot be considered as an ideal monodomain. Indeed, there will be no variations of parameter $\Delta\alpha(\mathbf{B})$ if we suggest that the sample layer structure is not affected by variations of the magnetic field and the attenuation depends only upon the angle between the director and wave vector in the same way as in nematics. It should be noted that the value of B used in the experiment is probably not enough for orientation of the director in the single direction on the side of the cone. The existence of the dependencies $\Delta\alpha(B, B_0)$ points to a distortion of the sample layer structure. Considering the orientation variations of the domain normals to be small (much less than Ψ) the model similar to the monodomain one with small deflections of layer normals from the magnetic field direction \mathbf{B}_0 in which the sample was cooled seems to be more efficient for our case.

Indeed, in the examined compound the smectic-C phase is formed from the smectic-A phase (S_A), so that the tilt angle Ψ at smectic-A - smectic-C phase transition is small compared to Ψ by about 45° in the S_C phase formed from the nematic, in which case the polydomain model evidently is appropriate. In our case the prepared layer structure of the S_C phase cannot differ in principle from the layer structure S_A especially near S_A - S_C phase transition. To destroy the layer structure of the smectic-C phase a stronger field ($B_0 \approx 1.4 \text{ T}$) or a strong boundary influence are needed.^{1,2,8} The model assumptions agree with our other experiments the results of which have been published earlier.³⁻⁶ In the proposed model the smectic-C has two stable angular states of director.⁶ This structural bistability probably explains the existence of the two relatively flat regions on $\alpha(\varphi)/f^2$ dependence Fig.2 (φ - angle between \mathbf{B} and \mathbf{q}). These regions disappear when the magnetic field \mathbf{B}_0 is decreased and the sample layer structure is like of polydomain. The magnetic field was rotated in the plane normal to smectic layers. As can be seen from Fig.2 the obtained curves have a period of 180° , but differently from the corresponding dependencies in the nematic phase. Also, we do not observe the variations of ultrasound velocity when the field is being changed. This fact is a good evidence that the smectic layers, once formed, conserve their orientation when field is rotated away from its original direction or when it changes its value in a constant direction.

At a fixed temperature the variations of α are due to the corresponding

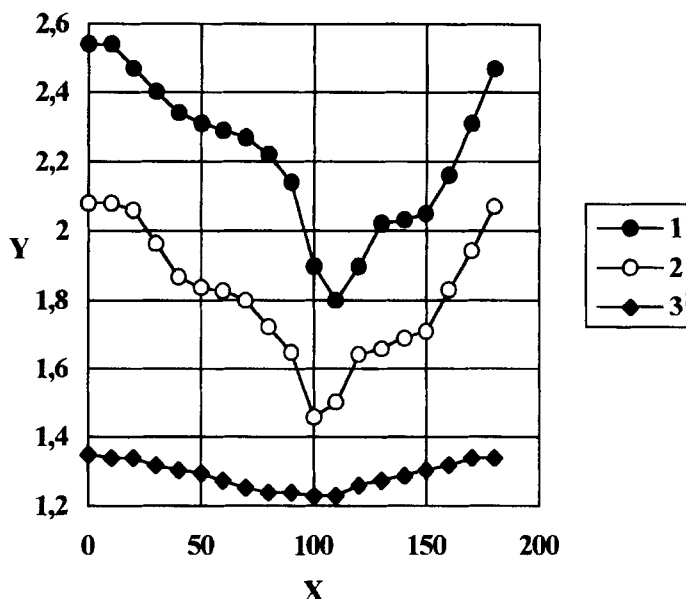


FIGURE 2 Angular dependencies of the attenuation coefficient:
 X - angle φ (degrees), Y - α/f^2 , $10^{-12} \text{ m}^{-1} \text{ s}^2$,
 1 - $B_0=0.6 \text{ T}$, 2 - $B_0=0.16 \text{ T}$, 3 - $B_0=0 \text{ T}$.

variations of B . Assuming the behaviour of the director to be quasinematic we have attempted to explain the dependence $\Delta\alpha(B)/f^2$ obtained using the expression found for nematics:⁹

$$\alpha/f^2 = a \cdot \cos^2(\theta) + b \cdot \cos^4(\theta) + \alpha_0/f^2 \quad (1)$$

where a and b are combinations of the viscosity coefficients; α_0 is constant; θ is angle between director \mathbf{n} and wave vector \mathbf{q} . In order to compare the observed behaviour with that produced from the smectic-C models we have calculated the $\langle \cos^2(\theta) \rangle$ and $\langle \cos^4(\theta) \rangle$ as functions of the magnetic energy E_H

$$E_H = 1/2 \cdot \Delta\chi \cdot N \cdot (\mathbf{n} \cdot \mathbf{H})^2 \quad (2)$$

and the geometrical model parameters for the two orientations \mathbf{B} and \mathbf{q} . The brackets mean statistical average with the distribution function:

$$W = A \cdot \exp(-E_H/kT) \quad (3)$$

$\Delta\chi$ is the anisotropy of the magnetic susceptibility; \mathbf{H} is the magnetic field strength; N is the number of the molecules with correlated orientation. The geometrical model parameters are the molecular tilt angle (Ψ), angle between smectic

layer normal and the \mathbf{B}_0 direction, azimuth angle of the layer normal projection on the layer plan and azimuth angle of the director projection on the perpendicular plan to the \mathbf{B}_0 direction. The molecular tilt angle in S_c phase of the compound chosen by us was determined experimentally equal to 15° at the temperature of 65°C .¹⁰ Assuming $\Psi=15^\circ$ we have found that our experimental data can be satisfactorily explained in the framework of the model taking into account the relatively small (about 10°) inclination angle of the smectic layer normals from the \mathbf{B}_0 direction. The influence of the possible layer structure defects on experimental data is not clear.

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