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Electric Field Induced Transitions in Polar Liquid Crystals with Frustrating Interlayer Interaction

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*Using Landau theory of phase transitions, behaviour of antiferroelectric liquid crystal and first order transition to the nonpolar Sm*A *phase were studied in electric field. Influence of frustrating interaction on structure above the temperature of the transition into a nonpolar phase was investigated. We have calculated an electric field - temperature phase diagram, including structures with spatial variation of both the phase and the modulus of the order parameter.*

Keywords smectic liquid crystals, antiferroelectrics, frustations, phase transitions

In recent years an essential breakthrough was achieved in experimental and theoretical studies of polar smectic phases [1]. An important step forward were the pioneering works of Mach *et al*. [2,3] on resonant x-ray scattering. They demonstrated that besides well-known ferroelectric smectic-C^* (SmC^*) and antiferroelectric smectic-C^*_A (SmC^*_A) phases with one- and two-layer periodicity there exist structures with three-layer (SmC^*_{d3}), four-layer (SmC^*_{d4}) period and a helicoidal structure (SmC^*_α) with a short-pitch period from about three to several tens of molecular layers. The period of the helicoidal structure can be incommensurate with smectic layer thickness. In subsequent x-ray and optical studies [4–9] commensurate structures were observed with larger periodicities up to 12 molecular layers [9].

Polar liquid crystals have layered smectic structure. The long axes of molecules are tilted in the layer plane (Fig. 1). Orientation of molecules in i-th smectic layer can be described by a two-component two-dimensional vector ξ_i parallel to the plane of smectic layers. Modulus of ξ_i characterizes the tilt of molecules (angle θ_i), direction of ξ_i – the azimuthal orientation of molecules (angle φ_i). Different structures can be formed by a layer-by-layer change of the azimuthal orientation of molecules and order parameter modulus (Fig. 1b). Interlayer interactions induce two-level structure of phases: (a) local periodic ordering of ξ_i, which leads to formation of different phases with period from one (SmC^*) to about ten molecular layers, (b) local periodic ordering is modulated by a long-pitch helix, related to chirality, with a typical period from about a hundred to several thousand layers. The deviation of mutual azimuthal orientation of molecules in adjacent layers $\Delta\varphi_i = \varphi_{i+1}\text{-}\varphi_i$ in commensurate phases from 0 (synclinic ordering) or π (anticlinic ordering) is due to chirality. In our calculations we shall not account for the chiral interaction, i. e. we shall consider commensurate structures to be planar (that is, $\Delta\varphi_i = 0$ or π).

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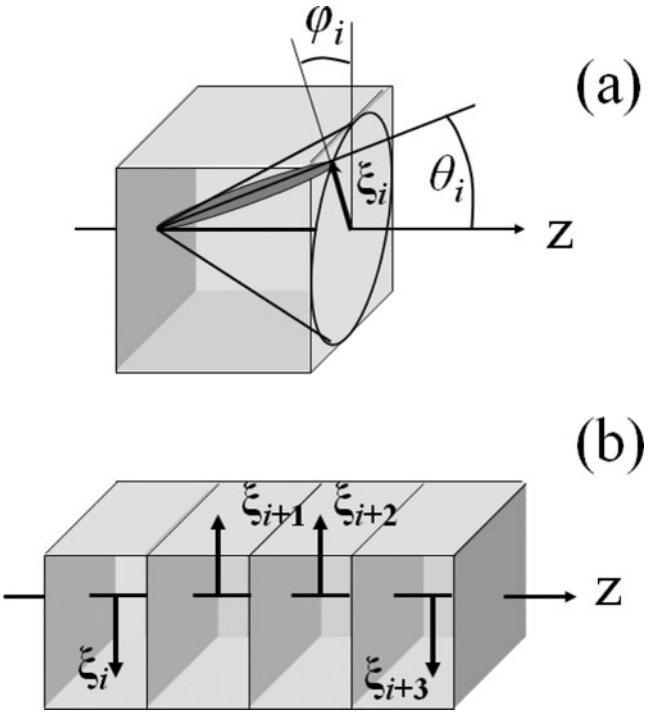

Figure 1. (a) Polar (angle θ_i) and azimuthal (angle φ_i) orientation of molecules in i-th smectic layer can be described by a two-component vector ξ_i. Modulus of ξ_i characterizes the tilt of molecules in the layers (angle θ_i). (b) Different polar phases are formed by variation of ξ_i from layer to layer.

To describe different polar phases and transitions between them such an approximation is justified to be used in calculations [10–12] since chiral interlayer interaction, which induces a long-pitch helix, is essentially weaker than interactions responsible for formation of different smectic phases.

Different polar smectic structures and phase transitions between them upon change of temperature have been extensively studied experimentally and theoretically [1,13–22]. An effective way of acting on polar structures is applying electric field to the sample. In field, transformation of the structure with a change of phase and modulus of the order parameter can take place. We shall concentrate on two effects:

(1) In electric field polar structures can be formed which are not observed in absence of the field. One of the first models of multistage transition from antiferroelectric SmC^*_A to ferroelectric SmC^* was proposed by Fukuda *et al*. [23]. In a general case the transition can occur quasicontinuously via a manifold of ferroelectric structures (Farey sequence). Upon increasing field, structures with larger polarization form. However in most cases the transition occurs either directly into SmC^* or via a limited number of structures.

(2) Another little-studied subject is the field-induced transformation of anticlinic structures above the temperature of the transition into the paraelectric Smectic-*A* (Sm*A*) phase. The electroclinic effect has been studied thoroughly in ferroelectric liquid crystals [24–27]. In ferroelectrics both the interlayer interactions and the electric field promote parallel orientations of ξ_i in different layers. In contrast, interlayer interactions in antiferroelectric structures, frustrating next-nearest layer interaction promote anticlinic orientation of

molecules which leads to a competition with electric field stabilizing the synclinic structure. In the case of a first order transition formation of unusual structures was predicted [28,29] near the temperature of the transition into the paraelectric phase.

In Landau theory of phase transitions between polar phases vector ξ_j (Fig. 1) can be used as the order parameter. The two-component order parameter can be written in the form $\xi_j = |\xi_j| e^{i\varphi_j}$ and can be characterized by the modulus and phase of the order parameter. The expansion of the free energy F over the order parameter includes conventional Landau terms F_L for isolated layers, which depend on the order parameter modulus [30]

$$F_L = \sum_i \left[\frac{1}{2}\alpha(\mathrm{T} - \mathrm{T}^*)\xi_i^2 + \frac{1}{4}b_0\xi_i^4 + \frac{1}{6}c_0\xi_i^6 \right]. \quad (1)$$

T^* is the temperature of the second order phase transition ($b_0 > 0$) into the paraelectric phase in absence of interaction between layers. First order phase transition into the paraelectric phase ($b_0 < 0$, $E = 0$) occurs at a higher temperature $T_0 > T^*$. Interlayer interactions F_{int} in polar liquid crystals depend on the azimuthal orientation of molecules in layers, i. e. on the phase of the order parameter [1,31]

$$F_{\mathrm{int}} = \frac{1}{2}a_1 \sum_i \xi_i\xi_{i+1} + \frac{1}{8}a_2 \sum_i \xi_i\xi_{i+2} + \sum_i b\,[\xi_i \times \xi_{i+1}]^2. \quad (2)$$

Interaction of layer polarization P_i with external electric field $F_E = \sum P_i E$. Layer polarization is proportional to the modulus of the order parameter $P_i = P_0|\,\xi_i|$. The interaction between nearest layers is described by the first term in (2). For $a_1 < 0$ the interaction favours the synclinic orientation of molecules ($\Delta\varphi_i = 0$), for $a_1 > 0$ the anticlinic orientation ($\Delta\varphi_i = \pi$). The interaction of next nearest smectic layers (the second term in (2)) is mainly due to existence of layer polarization [31]. As was demonstrated by Čepič and co-workers [31,32] $a_2 > 0$. For positive a_2 the interaction favours anticlinic orientation of molecules in i-th and $i + 2$-th layers. Such an orientation is incompatible both with ferroelectric (SmC^*) and with antiferroelectric (SmC^*_{A}) structures, which leads to frustration. The release of frustration can occur via formation of multilayer structures [1]. The biquadratic interlayer interaction (the third term in (2)) describes the energetic barrier between synclinic and anticlinic orientation of molecules. Structures of the polar phases were obtained by minimization of the free energy $F = F_L + F_{int} + F_E$ over the phase and modulus of the order parameter for different structures and comparing their energies.

In this work we investigate antiferroelectric with a first order transition to the SmA phase at $E = 0$. The transformation of antiferroelectric in electric field for different values of frustrating interaction and upon changing temperature was studied. Figure 2 shows the phase diagram in coordinates frustrating interaction – P_0E. At $E = 0$ and fixed $|\,\xi_i|$ the antiferroelectric phase exists up to $a_1/a_2 < 0.5$. If in different layers the order parameter modulus is fixed, $a_1/a_2 = 0.5$ is the point of global degeneracy, in which an infinite number of structures have the same energy [33,34]. If the values of ξ_i are not fixed and can change from layer to layer, in the vicinity of the point $a_1/a_2 = 0.5$ the three-layer SmC^*_{d3} structure (Fig. 3c) becomes energetically preferable [34]. For large values of frustrating interaction the SmC^*_{d4} phase is formed (right part of Fig. 2, Fig. 3d). For small a_2 in electric field the transition to ferroelecrtic (SmC^*) occurs directly from the antiferroelectric phase (left part of Fig. 2). Increase of the frustrating interaction induces formation of an intermediate ferrielectric SmC^*_{d3} structure. With increasing a_2 the range of the SmC^*_{d3} increases, and

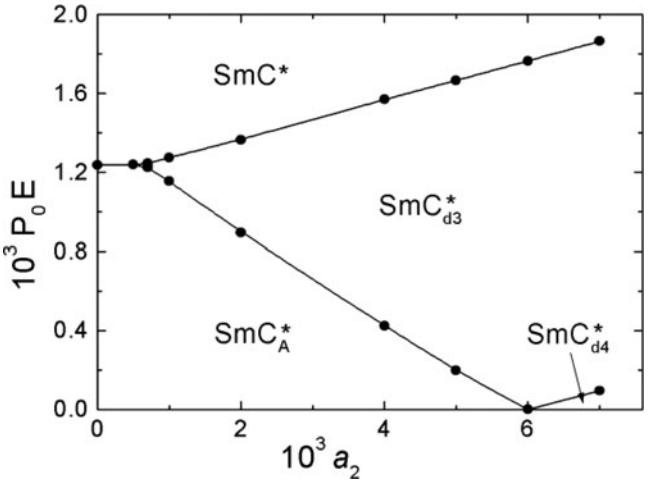

Figure 2. Phase diagram: frustrating interaction (a_2) – (P_0E). For a small value of frustrating interaction (a_2) the transition in electric field occurs between antiferroelectric and ferroelectric structures. Upon increase of frustration the ferrielectric SmC^*_{d3} structure is formed. At high value of a_2 the SmC^*_{d4} phase appears. Model parameters are: $\alpha = 0.01\text{K}^{-1}$, $b_0 = -0.5$, $c_0 = 6$, $a_1 = 0.003$, $b = 0.02$, T_0–$T = 7.25$K. T_0 is the temperature of the transition into the paraelectric phase in absence of the field.

the magnitude of the electric field decreases for the SmC^*_A-SmC^*_{d3} transition. Figure 4a shows the dependence of polarization on P_0E for $T < T_0$ and for a value of frustrating interaction which is sufficient to induce the SmC^*_{d3} structure in field. Upon the transition from SmC^*_{d3} to SmC^* the polarization increases in approximately three times. Some increase of polarization with field in the SmC^*_{d3} phase (Fig.4a) is due to changes of the

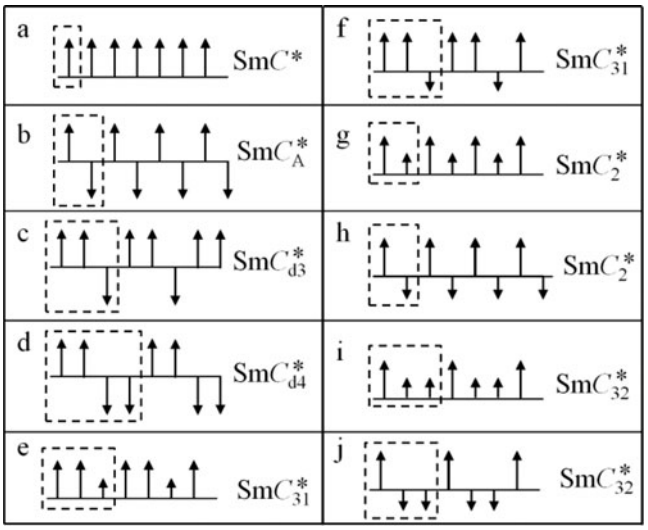

Figure 3. Schematic representation of polar phases. The period of ordering is perpendicular to the layer planes and is denoted by a rectangle.

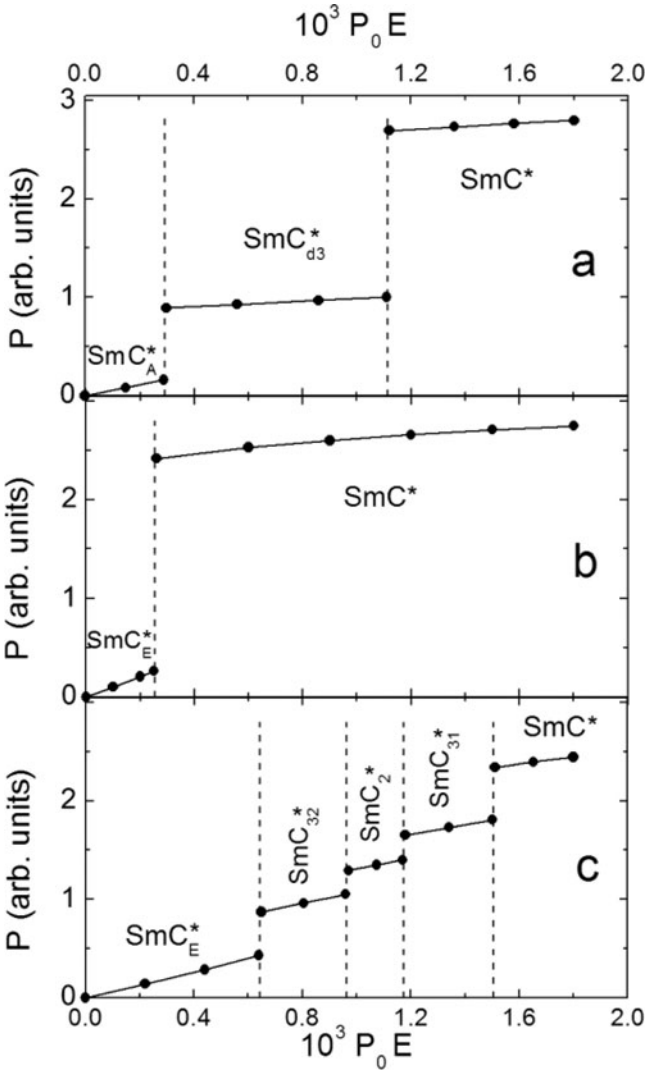

Figure 4. Electric field induced transition between polar structures at low $(T_0–T) > 0$ (a) and high $(T_0–T) < 0$ (b,c) temperature. (a) The transition from Sm$C_A{}^*$ to SmC^* in electric field at low temperature ($T < T_0$) in presence of frustrating interaction can occur via an intermediate ferrielectric Sm$C^*{}_{d3}$ structure. Model parameters are: $\alpha = 0.01\text{K}^{-1}$, $b_0 = -0.5$, $c_0 = 6$, $a_1 = 0.003$, $a_2 = 0.004$, $b = 0.02$, $T_0 - T = 0.5\text{K}$. (b) The first order Sm$C_E{}^*$ to SmC^* transition in electric field for $T > T_0$ in a ferroelectric liquid crystal. Model parameters are: $\alpha = 0.01\text{K}^{-1}$, $b_0 = -0.5$, $c_0 = 6$, $T_0 - T = -0.2\text{K}$. (c) In antiferroelectric with frustrating interlayer interaction the transition from Sm$C_E{}^*$ to SmC^* can occur at $T > T_0$ via a series of intermediate structures. Model parameters are: $\alpha = 0.01\text{K}^{-1}$, $b_0 = -0.5$, $c_0 = 6$, $a_1 = 0.003$, $a_2 = 0.004$, $b = 0.02$, $T_0 – T = -0.2\text{K}$ (c).

order parameter ξ_i (increase of ξ_i in layers with polarization parallel to the field, decrease in layers with polarization antiparallel to the field). A ferrielectric structure in electric field was observed by Shtykov *et al*. [35] in a compound in which the Sm$C^*{}_{d3}$ phase exists at a higher temperature in absence of electric field. In field the stability range of the Sm$C^*{}_{d3}$

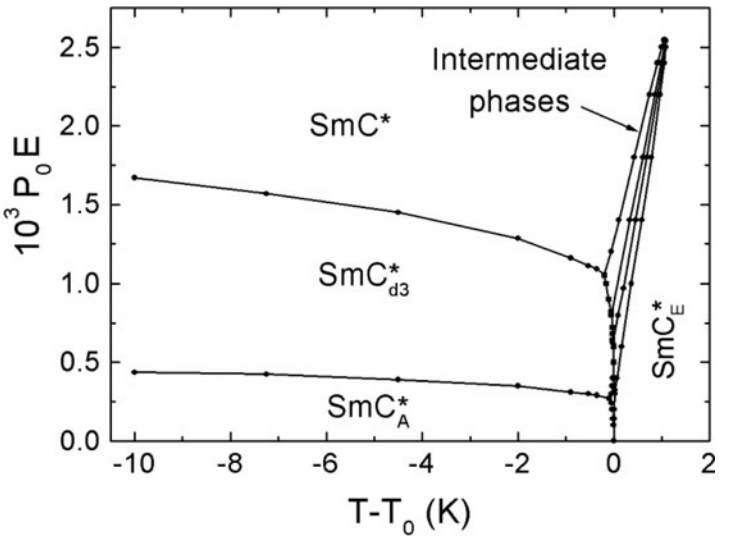

Figure 5. *(T–T₀) – (P₀E)* phase diagram. The region near T_0 is shown in Fig. 6 on an enlarged scale. Model parameters are: $\alpha = 0.01\text{K}^{-1}$, $b_0 = -0.5$, $c = 6$, $a_1 = 0.003$, $a_2 = 0.004$, $b = 0.02$.

phase was shifted to lower temperatures. Figures 2 and 4a illustrate the situation when in absence of the field heating of the SmC^*_A does not lead to a transition to the ferrielectric phase. Analogous formation of the SmC^*_{d3} was observed by Song *et al*. [36] in mixtures of ferroelectric and antiferroelectric liquid crystals. Formation of SmC^*_{d3} leads to two-step switching in optical devices based on antiferroelectric liquid crystals.

Transformation of polar structures in electric field, analogous to the one shown in Fig. 2, occurs in a wide temperature range T_0–$T > 0.3$K (Fig. 5). Polar structures in the low-temperature range mainly differ by azimuthal orientation of molecules, that is, the phase of the order parameter (Fig. 3b–d), which can be considered a "soft" component of the order parameter. The value of the order parameter modulus is determined mainly by Landau free energy F_L. Spatial change of the order parameter modulus can be small ("rigid" component of the order parameter).

Structures and phase transitions of essentially different type can be observed near T_0. In this temperature range for the first order phase transition Landau energies F_L with zero and finite values of the order parameter are close. Interlayer interactions and interaction of polarization with electric field can induce structures consisting of a combination of layers with different values of the order parameter. New types of structures are formed which are not observed at low temperatures. The high-temperature part of the phase diagram (Fig. 6) demonstrates appearance of new types of structures shown in Fig. 3e–j with a spatial modulation of the order parameter.

With increasing temperature the transition from the ferroelectric SmC^* structure (Fig. 6) with a large value of the order parameter $|\xi_i|$, determined mainly by Landau terms, to the SmC^*_E structure with a small value of the order parameter related to electrostriction, occurs via intermediate structures. These structures are composed of layers with large and small value of the order parameter (Fig. 3e,g,i). Upon increasing temperature and upon the transition from one structure to another the fraction of layers with a small value of the order parameter increases (1/3 in Sm$C_{31}{}^*$, 1/2 in Sm$C_2{}^*$, 2/3 in Sm$C_{32}{}^*$, Fig. 3). In large field all layers in intermediate structures have synclinic orientation of molecules

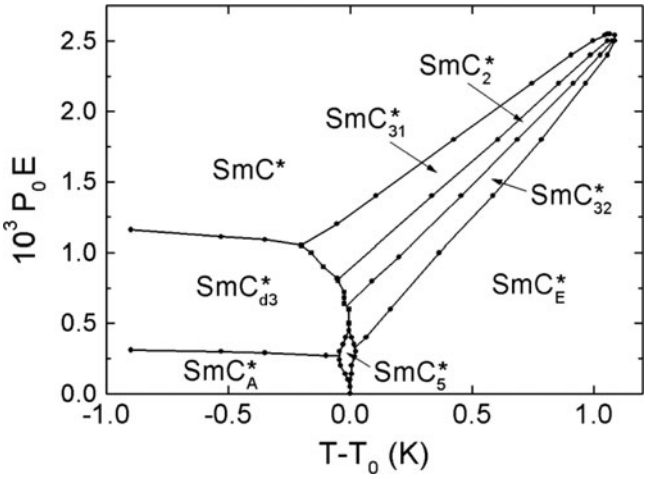

Figure 6. *(T–T$_0$) – (P$_0$E)* phase diagram near T_0. Model parameters are: $\alpha = 0.01\mathrm{K}^{-1}$, $b_0 = -0.5$, $c = 6$, $a_1 = 0.003$, $a_2 = 0.004$, $b = 0.02$.

(Fig. 3e,g,i). Near the border with the SmC^*_{d3} phase the orientation of molecules with a small and large values of the order parameter is anticlinic (Fig. 3f,h,j). In low field near $T–T_0 = 0$ a small region of five-layer ferroelectric SmC_5^* is formed (Fig. 6). This structure has anticlinic ordering in three layers and small value of the order parameter in two other layers. Polarization of this structure $P_5 \approx 1/5$ of the polarization of the structure with synclinic orientation of molecules (i.e. of the SmC^* structure).

Figure 4b shows the dependence of polarization on electric field in ferroelectric liquid crystal without frustration interaction in the temperature range above T_0. Electric field leads to two well-known effects: (a) electroclinic effect for small values of the field, when order parameter and polarization continuously increase with increasing field; (b) a stepwise transition into a ferroelectric structure with polarization which is close to the value of polarization near the transition to the paraelectric phase in absence of the field. The latter behaviour was observed by Bahr and Heppke in ferroelectric liquid crystal C7 with a first order phase transition from the SmC^* to SmA structure [26]. In this temperature range in a ferroelectric liquid crystal tristable switching can be observed, which is analogous to the one observed in antiferroelectrics at $T < T_0$ [23].

Interlayer interactions for $a_1, a_2 > 0$ (antiferroelectric liquid crystal) induce a splitting of the first order transition from SmC^*_E to SmC^* into several transitions forming intermediate structures (Fig. 4c, Fig. 6). Upon increasing field, multistep switching can be observed. The polarization increases in a stepwise manner upon increasing the field. It is worth noting once more that the described manifold of phases is formed in antiferroelectric liquid crystals. In ferroelectric a usual picture of the transition between SmC^* and field-induced SmC^*_E structure is observed (Fig. 4b).

According to calculations, near T_0 structures can be formed in which the order parameter and polarization differ significantly in adjacent layers. So one may ask whether such variations of polarization and the order parameter can be realized in polar compounds or other interactions exist which are not accounted in Landau theory that prohibit a strong change of order parameter and polarization in nearest layers. Experiments on free standing

films of liquid crystals [37] show that the order parameter profile can be strongly nonuniform near the surface [38−41]. Near T_0 the difference of molecular tilt angles in the surface layer and in the adjacent one can be of the order of ten or more degrees [38,41]. These experiments suggest the possibility of formation of modulated structures in antiferroelectrics.

In this paper in the framework of Landau theory of phase transitions we calculated temperature - electric field phase diagrams in antiferroelectric liquid crystals below and above T_0. Interlayer interactions, responsible for appearance of the antiferroelectric phase, and the frustrating long-range interaction can sufficiently transform the structure above T_0. In electric field formation of a sequence of structures with different value of polarization is possible. The appearance of these structures is due to the competition of the interaction of layer polarization and electric field with interlayer interactions. Regions of stability of intermediate ferroelectric phases were determined. These structures are formed by spatial change of phase and modulus of the order parameter. Such structures can be formed not only in polar liquid crystals but in other systems which are described by a two-component order parameter. Interlayer interactions in solid antiferroelectrics and magnetics can lead to effects analogous to the ones described in this work. A variety of experimental methods can be used to study intermediate structures: electrooptical and polarization studies, resonant x-ray diffraction, and diffraction of polarized neutrons.

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