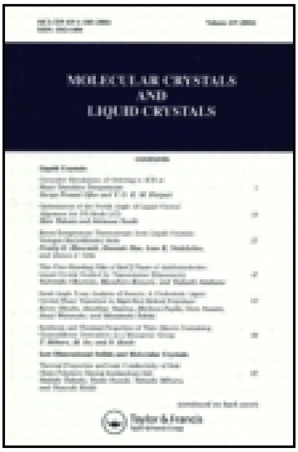

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Short-Pitch Modes Approach to the Problem of Antiferroelectricity in Liquid Crystals

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SHORT-PITCH MODES APPROACH TO THE PROBLEM OF ANTIFERROELECTRICITY IN LIQUID CRYSTALS

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Abstract The short-pitch modes (SPM) model is proposed for the description of various antiferroelectric and ferrielectric phases in the framework of phenomenological approach taking into account short-range correlations with different characteristic lengths for non-chiral and chiral terms in the free energy expansion, this small difference being dependent on the temperature. The ratio of correlation length to molecular length determines the number of SPM with wave numbers close to rational numbers of the inverse molecular length. The weak chiral interaction determines the wave number of long-pitch mode and the temperature intervals of the existence of each phase. The free energies of all the SPM are equal at the critical point that causes a mixing, i.e., a coexistence of many modes in the vicinity of this point. The alternation of ferri- and antiferroelecric phases near the critical point depends on material parameters of compounds. The existence of ferrielectric phases is determined by a neighbourhood of long-pitch mode. Thermodynamic properties and dielectric relaxation processes in such phases are discussed.

INTRODUCTION

During last several years, a variety of antiferroelectric and ferrielectric structures in several compounds and in their mixtures was discovered [1–6]. It was noted [7,8] that the observed phenomena are typical for the compounds with a characteristic structure of molecules. Hori *et al.*[8] marked that largely bent molecules can be responsible for the antiferroelectricity in liquid crystals. Isozaki *et al.*[7] underlined that the repulsive interactions and packing entropy of molecules play an important role in these phenomena. The sequence of phases is dependent on compounds. For binary mixtures of MHPOOCBC and EHPOCBC, the possible phases observed stably are [7] SmC^*_A—FI_L—SmC^*_γ—FI_H—AF—SmC*—SmC^*_α—SmA. The phase SmC^*_α is considered [9–11] as a coexistence of a variety of ferrielectric and antiferroelectric structures but not as a single structure. The direct phase transition SmC^*_A—SmC* is more typical for a mono compound [12]. The phase sequences were investigated in the papers [13–15] on the basis of phenomenological models of Landau-type theory. The appearance of the sequence FI_L—Sm^*_γ—FI_H—AF was described [9] by the one-dimensional Ising model with long-range repulsive interactions. One should note that one-dimensional Ising models describe phase transitions in a non-realistic case of a spin with all the rest spins in the system. The expansions in terms of small wave numbers q are correct only for $ql \ll 1$, where l is the length of a molecule. The present paper makes an attempt to look on the antiferroelectricity phenomenon from the opposite side in sense of q-expansions. This phenomenological model serves to understanding of some general features of the phenomenon under consideration: the closeness of pitches of these phases to rational numbers, mixing of several SPM, the alternation of ferri- and antiferroelectric states, the limitation on the number of such states and the relation of these properties to a structure of molecules.

THE SHORT-PITCH MODES MODEL

The set of physical features mentioned above cannot be occasional. A number of ferro-, ferri- and antiferroelectric states shows a relatively large radius of alternating interactions between molecules. The physical reason for this can be long-range smectic correlations with an effective correlation length ξ larger than l. Such correlations can be related to steric interactions between molecules with strongly curved shape. The permanent molecular electric dipole moments do not interact in such a way directly, but they can correlate indirectly due to the interaction between chiral and polar molecules. In general, the dipole ordering can be caused by a polar asymmetry of the molecular shape, which modulates the chiral intermolecular interaction through the excluded volume effects[16]. One can assume that the excluded volume effects increase the correlation radius ξ for chiral centers with permanent dipoles. Inside such an area, without taking into account the effects of chirality, dipoles can be oriented along the polar axis with equal probability for opposite orientations. It means that the energies of all these states are equal and they contribute to the main term of the free energy. Such states and corresponding modes $\exp(i\pi kz)$ can be characterized by wave numbers $k = m/nl$, where $m = 1, 2..., n = m, m+1, ...$ which represent the packing possibilities for molecules. The periods (pitches) of these structures $2nl/m$ are equal or larger than double molecular length. The wave numbers k have a sense if they are larger than ξ^{-1}, i.e., if $n/m \prec \xi/l$, the short pitches being inside the correlated area. The main term of the free energy, in such a case, can be presented in the form $\sin^2(m\pi/ql)$, it is equal to zero at $q = k$, but it sharply increases at $q \neq k$. If the ratio n/m is larger than ξ/l, then, in fact, the modulations with small q should be taken into account, the main term in the free energy being proportional to q^2. In both cases, this main term is independent of the sign of q.

Taking into account the weak effects of chirality, one should include the term depending on the sign of q to the free energy, for example in the form $\beta \sin(m\pi/ql)$at large q or the term βql at small q, where the small constant β characterizes the relative weakness of the chirality effects. These effects can change slightly the correlations between dipoles, for example the correlation length $\lambda \simeq \xi$, the difference $\lambda - \xi$ being essentially dependent on the temperature, especially near SmA—SmC* phase transition point.

Thus, instead of the magnitude q characterizing the heterogeneous fluctuation terms in the free energy, one can choose the more suitable magnitudes $ql^{-1}/(q^2 + \xi^{-2})$ and $ql^{-1}/(q^2 + \lambda^{-2})$which help to describe both long-pitch and short-pitch modes. These magnitudes have maximum values of the order of ξ/l which are larger than 1. Therefore it is not correct to develop the free energy as a series in powers of these magnitudes and to set a limit on the powers. It is good practice to think of the free energy as a series in harmonic functions of the magnitudes. The simple assumption is done in the present phenomenological SPM model, just the first harmonics are taken into account to describe the effects under consideration. One can write the non-chiral and chiral terms mentioned above as

$$\sin^2\left(\frac{m\pi ql^{-1}}{q^2 + \xi^{-2}}\right) + \beta\left[\alpha - \sin\left(\frac{m\pi ql^{-1}}{q^2 + \lambda^{-2}}\right)\right]^2 \tag{1}$$

The first term in (1) is the main term, the second one describes the weak chirality effects, the parameters α and β are much smaller than 1. It is clear from (1) that one has the ordinary expression for the q-dependent free energy term at small values of q. If $q\xi$ and $q\lambda$ are much larger than 1, the main term in (1) shows that the SPM with $q = k$ correspond to minimum energies. Thus, for the fixed m, the modes with n smaller than $(m\xi/2l)$can really exist, the most essential m-terms being determined by the molecular structure. The magnitude $(m\xi/2l)$ determines, in fact, the possible number of phases and transitions between them for a given m-set.

THE PHASE TRANSITIONS IN THE SPM MODEL

The set of chiral smectic phases can be described by the minimization of the expression (1) with respect to the wave number q. To do this, let us consider, first of all, the dependence of the free energy on q in vicinity of the points $q = k$. Substituting the magnitudes $q = k + \delta q$ into (1) and developing this expression as a series in powers of δq, one can find that, for a given m-set, each short-pitch n-mode corresponds to the existence of the minimum energy

$$f_n^m\left(\delta q_n^m\right) \simeq \frac{n^6\pi^2}{m^4}\beta\left(1-\beta\right)\left[\frac{l^2}{\lambda^2}-\frac{l^2}{\xi^2}+\left(-1\right)^n\frac{m^2\alpha}{n^3\pi}\right]^2 \qquad (2)$$

$$\delta q_n^m \simeq -\frac{1}{l\left(1+\beta\right)}\left[\frac{nl^2}{m\xi^2}+\beta\left(\frac{nl^2}{m\lambda^2}+\left(-1\right)^n\frac{m\alpha}{n^2\pi}\right)\right].$$

It is seen from (2) that the small parameter

$$\tau = \frac{l^2}{\lambda^2}-\frac{l^2}{\xi^2}$$

determines the temperature dependence of the free energy for each SPM. Each SPM has zero minimum energy at

$$\tau = \tau_n^m = \left(-1\right)^{n+1}\frac{m^2\alpha}{n^3\pi}. \qquad (3)$$

All SPM, in accordance with (2), have the same energy

$$f_c = f_n^m(\tau = 0) \simeq \alpha^2\beta(1-\beta) \qquad (4)$$

at the point $\tau = 0$.

It is clear from (2) that the total number of SPM is limited by the condition $l\delta q_n^m \leq 1$. If $l\delta q_n^m > 1$,one should find another minimum of the free energy which is related to a long-pitch mode. Assuming that such a minimum corresponds to $q\xi$ and $q\lambda$ much smaller than 1 and neglecting the corresponding magnitudes in (1), one can easily calculate by the minimization of (1) that the value

$$q = q_0 \simeq \frac{\alpha\beta\lambda^2 l}{m\pi\xi^4} \qquad (5)$$

determines another minimum of the free energy

$$f_0 \simeq \alpha^2\beta\left(1-\frac{\beta\lambda^4}{\xi^4}\right) \simeq f_c + 2\left(\frac{\alpha\beta\lambda}{l}\right)^2\tau \qquad (6)$$

It is important to note that the magnitudes $f_c$and f_0 are equal at $\tau = 0$ accordingly to the expressions (4) and (6), the value $q_0 l \sim \alpha\beta\left(l/\xi\right)^2$ being much smaller than 1. In vicinity of the point $\tau = 0$, the magnitudes $f_n^m\left(\tau\right)$ can be written in the form

$$f_n^m\left(\tau\right) \simeq f_c + \left(-1\right)^n\frac{2\pi\alpha\beta n^3}{m^2}\tau. \qquad (7)$$

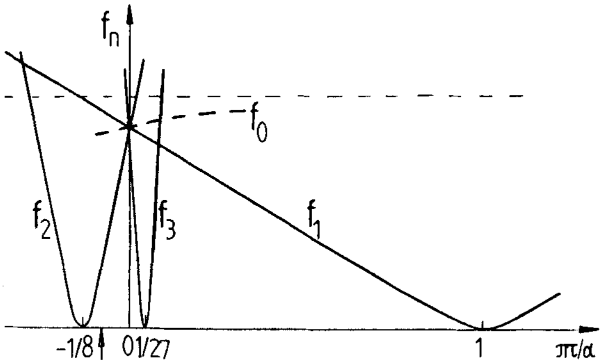

Figure 1: SPM energy branches as functions of a referred temperature.The fat dotted curve corresponds to the long-pitch mode, the light dotted straight line shows the energy of homogeneous structure

Thus, comparing the expressions (6) and (7), one can conclude that the slopes of the lines $f_0(\tau)$ and $f_n^m(\tau)$ at the point $\tau = 0$ can drastically differ or can be almost equal in dependence on the material parameters α, β, λ/l and on SPM numbers. Figure 1 shows these features of SPM behaviour.

The obtained results can throw some light on the observed phase diagrams. It is clear from (2)-(7) that there is the temperature ($\tau = 0$) where all the modes (short-pitch and long-pitch) have the same energy f_c. In vicinity of this point , a mixing of different modes can actually occur, the long-pitch mode being obviously attended. The point $\tau = 0$ more than likely is in the neighborhood the point $\tau(T_{AC})$ corresponding to the SmA—SmC* phase transition temperature T_{AC} because chirality effects vanish at this temperature. It is assumed that increasing positive values of τ correspond to a decrease of T below T_{AC}. The point $\tau(T_0) = 0$ may be slightly smaller than T_{AC}, i.e., $\tau(T_{AC}) < 0$ because of the interaction between SPM shifting the transition temperature. The point $\tau(T_{AC})$ is shown in Fig. 1 by the arrow. In the temperature interval corresponding to the interval $\tau(T_{AC}) < \tau < 0$, one can consider various scenarios of the phase transitions between SPM states (see Figure 2). For a given m-set of SPM with the total number $m\xi/2l$ of n-modes, where $n = m$, $m + 1$, ..., the odd n-modes are at positive values of τ, but the even n-modes are at negative values of τ (in the framework of the present model). In the considered narrow interval of negative values of τ, for a compound with material parameters satisfying the inequality

$$\frac{\alpha\beta\lambda^2}{l^2} > \frac{\pi n^3}{m^2} \tag{8}$$

for all even n-modes, the ordinary long-pitch SmC* has the lowest free energy according to (6) and (7) (see Figure 2a). For a compound in which, instead of the

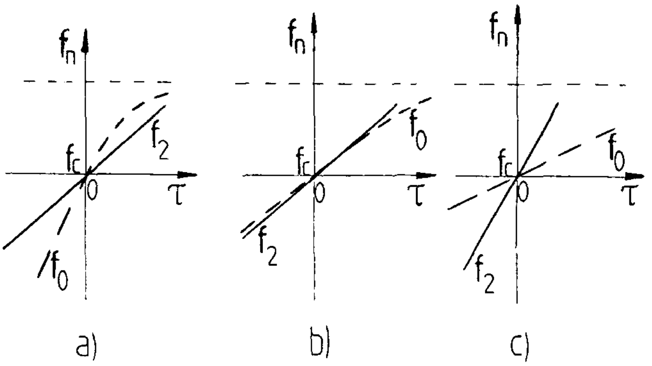

Figure 2: The scenarios of phase transitions at various values of material parameters. SmA phase can be followed by a) SmC* phase, b)FI phase as a mixing of long-pitch and short-pitch modes, c)AF phase

inequality (8), one has the approximate equality at a maximum even n, the mixing of the long-pitch mode with even $n_{\max}$-mode is more than likely (see Figure 2b). Such a state can be considered as SmC^*_α which is rather a ferrielectric phase. For compounds with material parameters satisfying the inequality which is inverse to (8), one should expect the existence of the antiferroelectric state, i.e., the SPM with a maximum even $n = n_{\max}$ (see Figure 2c). Thus, the phase transitions from SmA to SmC*, SmC^*_α (i.e., FI) and SmC^*_A (i.e., AF) depend on material parameters of compounds and their mixtures.

At the point $\tau = 0$, the situation is rather complicated because of mixing of many modes which can interact between themselves. In fact, it was assumed above that all the modes have the same amplitude A_0 (i.e., the tilt angle θ). In reality, these amplitudes A_n can differ. To describe their mutual influence, one should take into account additional terms in the free energy expansion, for example one can consider the typical term

$$-vA_0^2A_n^2 \tag{9}$$

corresponding to the interaction between the long-pitch and n-short-pitch modes, which induces the appearance of both modes, the parameter v being positive. One can imagine to himself a more complicated situation when, due to similar reasons, the long-pitch mode coexists with several SPM. One can affirm that the larger the magnitude $m\xi/2l$ related to molecular properties of compounds the larger is the quantity of SPM and the larger is the quantity of various ferrielectric states in vicinity of the temperature T_{AC}.

The steepness of the τ-behavior of the SPM free energy in vicinity of the point $\tau = 0$ allows to make another principal conclusion. With an increase of positive τ, possible ferrielectric states must be followed by a purely SPM, i.e., by an antiferroelectric state. Figure 1 shows, for example, that for the SPM with the maximum odd number n, there is deep and narrow pit in the function $f_n(\tau)$ in the immediate vicinity of the point $\tau = 0$. Therefore the phase transition to this antiferroelectric state from a ferrielectric one is highly probable. At the point τ^*, one should expect the first order phase transition from AF state with $n = 3$ to AF state with $n = 1$ (probably the observed phase SmC$^*_A$). But it should be noted that the difference between the free energies $\beta\alpha^2$ (for a non-modulated structure), f_c and f^* is small. Thus, again the interaction term (9) between existing amplitudes $A_1$and A_3 And the hidden one A_0 can induce the appearance of the long-pitch mode in a certain temperature range in vicinity of the point τ^*. Correspondingly, the mixing of these two SPM with the long-pitch mode creates the new ferrielectric state (probably the phase SmC$^*_\gamma$) in-between the phases AF$_3$ and AF$_1$ on the scale of temperatures. Depending on the structure of molecules, one can expect the existence of several such ferrielectric states, for example one can obtain the mixing of the long-pitch mode with the first, third or first plus third SPM, i.e., three FI phases in the situation described above (probably the observed phases FI$_H$, FI$_L$ and SmC$^*_\gamma$).

Let us consider more detailed the phase transitions between AF and FI states under an influence of the long-pitch mode (the ferroelectric state) when the corresponding phase transition temperatures are close with each other. One can write the corresponding free energy expansion in the form

$$F = (f_c + a)\,A_0^2 + (f_c + b)\,A_1^2 + \frac{1}{2}A_0^4 + \frac{1}{2}A_1^4 - vA_0^2A_1^2. \tag{10}$$

The temperature dependence of a and b, in such a model, is shown in Figure 3. It is seen that the lines $a(T)$ and $b(T)$ intersect at some point T^*, $a(T^*) = b(T^*) = a^*$, $|\, a^* \,|$ being larger than f_c. The minimization of the functional F with respect to the amplitudes A_0 and A_1 results in the equations:

- AF state

$$A_0 = 0, A_1 = \sqrt{|\, f_c + b \,|} \quad \text{at} \quad T < T_{AF} \simeq T^* - \,|\, (f_c + a^*)\,(da/dT)^{-1} \,|; \tag{11}$$

-the ferroelectric state

$$A_1 = 0, A_0 = \sqrt{|\, f_c + a \,|} \quad \text{at} \quad T > T_{FI} \simeq T^* + \,|\, (f_c + a^*)\,(db/dT)^{-1} \,|; \tag{12}$$

-FI state (at temperatures $T_{AF} < T < T_{FI}$)

$$A_0 \simeq \sqrt{|\, da/dT \,|\,(T - T_{AF})}, A_1 \simeq \sqrt{(db/dT)\,(T_{FI} - T)}; \tag{13}$$

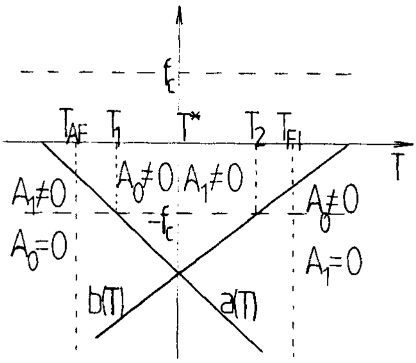

Figure 3: AF-FI phase diagram. The temperature intervals corresponding to AF, FI and ferroelectric phases are separated by vertical light dotted lines

where a smallness $v \ll 1$ is taken into account. The temperature T_{FI} is the point of the transition to FI phase from the ferroelectric one, the point T_{AF} corresponds to the transition from FI state to AF state, these phase transitions being of the second order. It is well known that a sufficiently strong interaction v results in the first order transitions.

DIELECTRIC RESPONSE OF FI AND AF STATES

The response of various phases to actions of the field E can be described by taking into account the additional terms $-A_n E$ in the functional F, the n-modes with the amplitudes A_n being induced by the electric field. For example, one can obtain the induced amplitude $A_0 \sim E/(T_{AF} - T)$ in AF state near the phase transition point T_{AF}, which is similar to a FI soft mode. In fact, the response A_0 cannot be infinite at T_{AF} because of long-pitch modulations with the wave number q_0, i.e., a relatively small correction of the order of Kq_0^2 should be included to the corresponding relaxation frequency $\omega_{AF,0}$:

$$\omega_{AF,0} \sim \gamma^{-1} \left[\mid da/dT \mid (T_{AF} - T) + Kq_0^2 \right], \tag{14}$$

where γ is the viscosity coefficient. The relaxation strength $\varepsilon''_{AF,0}$ related to the frequency (14) is inversely proportional to the magnitude $\gamma\omega_{AF,0}$, i.e., $\varepsilon''_{AF,0} \sim (T_{AF} - T)^{-1}$ must increase when the temperature comes closer to the phase transition temperature T_{AF}. The temperature dependence of the relaxation frequency $\omega_{AF,0}$ is more complicated due to the temperature dependence of viscosity. The decrease of viscosity, when the temperature increases, can result in an increase of $\omega_{AF,0}$ in relatively large temperature interval. To suppress such a response by the BIAS field, one needs the larger field when the temperature approaches T_{AF} point since the response increases near this point.

Above, in fact, we assumed that the magnitude q_0 is independent of temperature. But it is possible to consider the temperature dependence $q_0(T)$ in the framework of the SPM model. For this aim, one should add the term

$$gA_0^2A_1^2\sin^2\frac{m\pi ql}{q^2+\xi^{-2}}$$

to the free energy (1), where g is the positive constant. In such a case, one can calculate the corrections to the magnitude q_0 at temperatures close to the phase transition point T_{FI} at which the amplitude A_1 arises. In the assumption of small amplitudes A_1 and finite amplitudes A_0, one obtains the expression

$$q_{FI,0}\sim q_{C^*,0}\left(1-gA_1^2\right)$$

which shows that the long-pitch mode has a smaller wave number (larger pitch) in FI phase. If the phase transition FI—SmC* is of the first order, then one can expect a coexistence of two long-pitch modes with the wave numbers $q_{C^*,0}$ and $q_{FI,0}$, the difference $(q_{C^*,0}-q_{FI,0})$ is proportional to the amplitude A_1 squared and it becomes larger at temperatures below T_{FI}. In such a case, two Goldstone modes can be observed. The dielectric response in FI phase, which is related to the presence of the long-pitch mode, has the term $\mu_p^2/Kq_{FI,0}^2$, where μ_p is the piezoelectric coefficient. A general decrease of such a response, at temperature decreasing, must slow down at T_{FI} point due to the decrease of $q_{FI,0}$.

The dielectric response of the short-pitch mode A_1 (in FI and AF states) must be much smaller in comparison with the response of the long-pitch mode since the large wave number q_1 gives rise to large values of elastic energy. One can use ordinary expressions [17] to estimate qualitatively the effects of SPM on this response ε_{SPM}:

$$\varepsilon_{SPM}\simeq\frac{\mu_p^2}{Kq_1^2+i\omega\gamma_\varphi}+\frac{\left(\mu_p-\mu_fq_1A_1^2\right)^2}{const\,(T_{FI}-T)+Kq_1^2+i\omega\gamma_\theta}. \tag{15}$$

It is seen from (15) that $\mid\varepsilon_{SPM}\mid$ is much smaller than the response μ_p^2/Kq_0^2 since $q_1\gg q_0$. Eq. (15) takes into account a flexoelectric effect [17] which can be essential when the flexoelectric coefficient μ_f and wave number q_1 are large, this effect being related both to the modulations of azimuth (φ) and tilt (θ) angles of the director. If the value q_1^{-1} is equated to a molecular length l, the magnitude Kq_1^2/γ describes, in fact, some molecular motions. Of course, in such a case, the values of K and γ (γ_φ and γ_θ) are not correctly determined, but one can expect that this magnitude represents a certain relaxation frequency on the molecular level. Therefore the magnitude $A_1^2\sim(T_{FI}-T)$ in Eq. (15) describes some deviations of the relaxation frequency and of the relaxation strength from the molecular values. When the magnitude $const\,(T_{FI}-T)$ is much smaller than the magnitude Kq_1^2, but the magnitude

$\mu_f q_1 A_1^2$ is larger than μ_p, in a certain temperature range in FI phase (and even in AF phase), the molecular relaxation frequency is approximately constant, but the relaxation strength of the molecular mode can increase, at temperature decreasing, proportionally to the magnitude $\mu_f^2 q_1^2 A_1^4$.

CONCLUSIONS

The obtained results are based on the assumption that the free energy as a series in terms of harmonic functions of the variables $q/(q^2+\xi^{-2})$can be cut off by two first harmonics. The advantage of this phenomenological approach is that one can follow changes in micro (short-pitch) and macro (long-pitch) -modulated structures of chiral smectics with molecular inclinations. The natural limitation on the number of SPM is imposed by the smectic correlation length ξ (by the ratio ξ/l). Due to the presence of small parameters $(l/\xi)$and β (the weak chiral interaction potential), one can consider ordinary expansions of the free energy in terms of small deviations of the products ql from the rational numbers m/n describing SPM. Besides, they allow to consider the temperature dependence of the free energy near SmA—SmC*phase transition point, the corresponding narrow temperature range contains all the possible ferri- and antiferroelectric phases.

There is the critical intersection point $\tau = 0$ on the temperature scale, in which the energies of all SPM are equal that causes a mixing (i.e., a coexistence) of many modes in the vicinity of this point. Alternation of ferri- and antiferroelectric phases near this critical point totally depends on material parameters of compounds. The existence of ferrielectric phases is determined by a neighborhood of the long-pitch mode in the vicinity of the critical intersection point. Relatively far from this point, the intersections of the energy branches of two following SPM (pure antiferroelectric modes) are also in a neighborhood of the long-pitch mode branch. Therefore the interactions between the long-pitch mode and two SPM can induce up to three ferrielectric states due to a mixing with each SPM separately or with both SPM. The most simple antiferroelectric state with the shortest pitch exists at the lowest temperatures.

The presented theory can qualitatively explain many of observed physical features of the liquid-crystalline antiferroelectricity. Other applications of this approach are in progress. The results of this study are compared with the dielectric behavior of different substances showing antiferroelectric and ferrielectric properties in the paper presented at the 15th ILCC by Hiller *et al.*[18].

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