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The semi-phenomenological model of antiferroelectricity in chiral smectic liquid crystals† III. Dielectric spectroscopy

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In the framework of the model of short pitch modes (SPM) developed for antiferroelectric liquid crystals, the sets of various relaxation modes related to ferro-, antiferro- and ferri-electric phases are explained. The 'soft-like' modes describe the amplitude perturbations of short pitch modes, while the Goldstone modes describe the azimuthal perturbations of the molecular packings related to short pitch modes. The frequencies of the Goldstone modes are mainly determined by the chirality parameter, depending on the number of short pitch modes. The relaxation modes described correspond to the observed phase diagrams in liquid crystal materials for which the measurements of dielectric spectra are presented.

1. Introduction

Along with the models [1–8] applied for the consideration of very complicated problems related to the antiferroelectric structures and properties of new liquid crystalline substances, in refs [9–11] the short pitch modes (SPM) approach was proposed for the characterization of observed families of ferro-, ferri- and antiferro-electric phases in different chiral smectic materials with inclined molecules. This approach was related to the assumed structures for such smectic phases possessing complicated spatial director distributions $\Xi(z)$ along the crystalline z-axis. Function $\Xi(z)$ is, in fact, the sum of terms from SPM (with amplitudes Θ_n and large wave numbers q_n) and long pitch mode (LPM with amplitude Θ_L and small wave number q_L). The n-SPM, in the absence of any incommensurability, describe pure antiferroelectric (AF) structures with unit cells along the z-axis. In such a case, $q_n = k_n = m/nl$, where $m = 1, 2, \ldots, n = \pm m, \pm(m+1), \ldots, l$ is the thickness of a monomolecular layer, and the polarization vectors form closed manifold geometries in the smectic planes, which correspond to various stars in n layers-unit cells. The total number of SPM in a given family n_{max} is of the order of $m\xi/l$, where ξ is the correlation length (along the crystalline z-axis) measured in molecular lengths l [9, 10]. In the presence of a small incommensurability $\delta q_n = q_n - k_n$, the n-SPM describe weak ferrielectric (FI) states. In the case when SPM and LPM coexist, the ferrielectric properties are more pronounced. The resulting incommensurate structures, the phase transitions between them and the corresponding phase diagrams were described in the framework of such an approach in ref. [11].

The experimental study of the resulting structures and of possible SPM is the hard problem. One of the promising experimental methods is dielectric spectroscopy. In the present paper, in the framework of our approach, we give an interpretation of the relaxation modes in ferri-, and antiferro-electric phases and discuss recent experimental data in this area.

2. Soft modes and Goldstone modes in the SPM model

It was shown [10] that, in a simple approximation, when only the terms quadratic in Θ_j are taken into account, the free energy density takes the form

$$F = F^* + F_q = \sum_j F_j = \sum_j A_j \Theta_j^2$$

$$= \sum_j \left[\frac{R}{T_A}(T - T_j) + r f_j \right] \Theta_j^2 \qquad (1)$$

where the positive constants R and r have the dimensions of coefficients A_j (i.e. of energy density), the functions $f_j(T)$ describe the spatially heterogeneous part of the energy, and the first phase transition from the smectic A to a chiral smectic with inclined molecules occurs at the

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temperature T_A. The appearance of the j-waves is determined by the coefficients A_j vanishing in the F_j terms at some points T_{cj}. The magnitudes $|A_j|$ play the role of 'soft mode' frequencies

$$\omega_{sj} \propto |A_j|/\gamma_\Theta \tag{2}$$

(multiplied by viscosities γ_Θ for the polar angle motions) corresponding to the j-modes, which can be observed in the dielectric spectrum. In general, the response of various modes to the actions of an external field E is described by taking into account the terms $[-\Theta_j(\tau)E]$ in the functional F, the j-modes being induced by the electric field. In fact, in an antiferroelectric phase, these 'soft modes' are related to complicated (both the azimuthal and polar angles) perturbations of the director orientation. The difference between such frequencies, for example between ω_{s1} and ω_{s2}, is related to the difference in perturbations of the molecular short range packing [the packings with wave numbers $k_1 = l^{-1}$ and $k_2 = (2l)^{-1}$] inside the correlated area with radius ξ. It should be mentioned that the activation of such relaxation modes may be difficult in the SmC* and FI phases with a strong director twisting.

We remind the reader that the short range packings with different wave numbers k_n are assumed to be almost equivalent in their energies which have almost the same minimum value. The incommensurability effects may change this minimum value for different SPM [10]. It was assumed in [10] that the Landau coefficients $a_j = a'(T - T_j)$ for the j-modes vanish at different temperatures T_j if q_j are not exactly equal to k_j ($j = n$ and $T_j = T_n$ for the n-SPM), $T_j = T_L$ being also different for the LPM with $q_j = q_L$. Since $T_n = T^*$ for $q_n = k_n$, magnitudes T_n are equal to

$$T_n(q_n) \equiv T_n(q_n - k_n) = T^* + C(q_n - k_n)^2 \tag{3}$$

because the transition temperature cannot depend on the sign of the incommensurability $\delta q_n = q_n - k_n$. Constant C is positive if the incommensurability is favourable, i.e. if it results in an energy gain. The incommensurability effects, accordingly to equation (3), can effectively shift the transition temperatures similarly to the increase in the T_{CA} temperature due to the existence of a conventional helix [12]. It was shown [11] that $T_1(\delta q_1) < T_2(\delta q_2) < T_3(\delta q_3)$ and $A_3(0) < A_2(0) < A_1(0)$ if $\pi \geqslant \alpha\nu^{-1}\xi^2$, where α is the chirality parameter, magnitude $\nu^{-1/2}\xi$ characterizes the correlation length in the smectic plane which may be larger than the correlation length ξ along the crystalline axis. These assumptions (as a model) correspond to the observed increase in the phase transition temperatures when the period of the independent SPM structures increases. In the case of mixing of j-waves, the mentioned inequalities can even change to the inverse inequalities but in such a case, at higher temperatures the n-SPM with a smaller n does not appear independently of the SPM with a larger n. The inverse inequalities $T_3(\delta q_3) < T_2(\delta q_2) < T_1(\delta q_1)$ also occur if $27\pi \leqslant \alpha\nu^{-1}\xi^2$.

Under the condition $27\pi \leqslant \alpha\nu^{-1}\xi^2$ it is more than likely that the azimuthal perturbations of these packings are related to the chirality properties and described by Goldstone-like modes with the relaxation frequencies

$$\omega_{Gn} \propto (r/\gamma_\varphi)(\delta q_n)^2 \propto \frac{r}{l^2\gamma_\varphi}\left(\frac{\alpha}{n^2\pi}\right)^2 \tag{4}$$

$$\omega_{GL} \propto a'\delta T_L \gamma_\varphi^{-1} \propto r\alpha^2\gamma_\varphi^{-1} \propto (K/\gamma_\varphi)(q_L)^2 \propto \frac{K}{l^2\gamma_\varphi}\left(\frac{\alpha\nu}{\pi\lambda^2}\right)^2 \tag{5}$$

where magnitudes δq_n and q_L are given by equations (10) and (15) from [10]:

$$\delta q_n^m \cong -\frac{1}{l}\left[\frac{n\nu}{m\xi^2} + (-1)^n\frac{m\alpha}{n^2\pi}\right],\ q_L \cong \frac{\alpha\nu}{m\pi\lambda^2},\ \frac{\nu}{\lambda^2} = \frac{\nu}{\xi^2} + \tau. \tag{6}$$

Here, K is the elastic modulus, γ_φ is the viscosity coefficient for azimuthal orientational motions, and it is assumed that τ is equal to zero at the temperature T_A.

The ω_{Gn} values describe, in fact, the induced deviations from the energy minimum. The ω_{GL} value is conventional for the SmC* phase and, by this relation, we can estimate parameter r,

$$r \propto \frac{K\nu^2}{\pi^2 l^2 \lambda^4}. \tag{7}$$

Substituting this r value into the expression for ω_{Gn} we obtain:

$$\omega_{Gn} \propto \frac{\omega_{GL}}{\pi^2 n^4}. \tag{8}$$

Thus, the ω_{Gn} values become much less than ω_{GL} for $n > 1$, ω_{G1} being approximately one order less and ω_{G2} being approximately two orders less than ω_{GL}, i.e. the LPM-Goldstone relaxation mode has a larger frequency than the 1-SPM-Goldstone relaxation mode.

In accordance with equations (4) and (6), for $\alpha\xi^2\nu^{-1} \ll n^3\pi$, i.e. for large numbers n, we obtain the value

$$\omega_{Gn} \propto \omega_{GL}\nu^2(n^2/\alpha^2\xi^4) \tag{9}$$

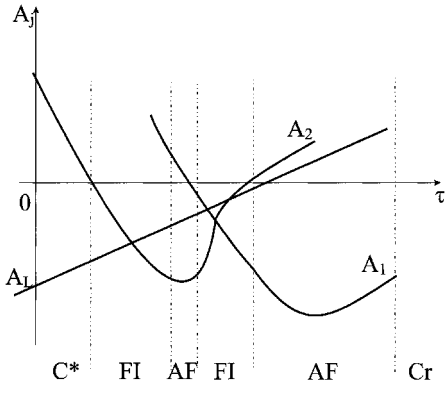
which is valid at $n \geqslant (\alpha\xi^2/\pi\nu)^{1/3}$. In such a case, the ω_{Gn} values increase with increase in the n-number, but they may be less than ω_{GL} when parameter $\alpha\xi^2\nu^{-1}$ is larger than n.

The activation of these relaxation modes may be difficult in the antiferroelectric phases because of the lack of an average polarization value inside a short

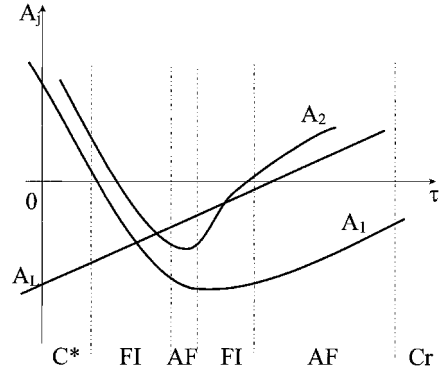
range area. If an AF phase possesses a weak proper incommensurability, i.e. it is a weak FI phase, then a weak Goldstone mode may be activated in such an AF phase. The application of the external electric field must induce the long range mode, i.e. the average polarization, and thus the interaction of this polarization with the field, must produce the azimuthal director perturbations and activate the Goldstone relaxation modes discussed.

3. The model phase diagrams

Let us consider, as an example, the phase diagrams presented in figures 1 and 2 corresponding to the inequality $A_{1\min} < A_{2\min}$, their absolute values being large with respect to the modulus of $A_L(\tau)$ at the points of intersection of these three lines. The inequality $A_{1\min} < A_{2\min}$ means that we consider the situation $\alpha\xi^2 \nu^{-1} \gg n^3 \pi$. Here, the 1-SPM does not appear independently of the 2-SPM

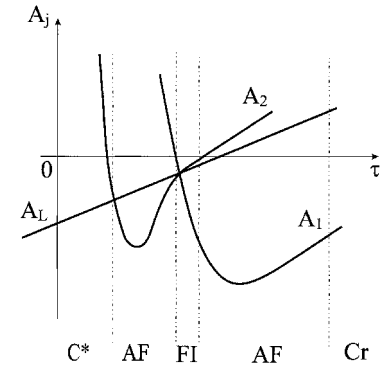


(*a*)



(*b*)

Figure 1. The two SPM and LPM branches of energy as functions of the reduced temperature. Two cases for the SPM lines are shown: (*a*) with intersection, (*b*) without intersection. The induction of LPM by weak SPM in the high temperature region is assumed. The SmC* phase, and two antiferro- and two ferri-electric phases are present.



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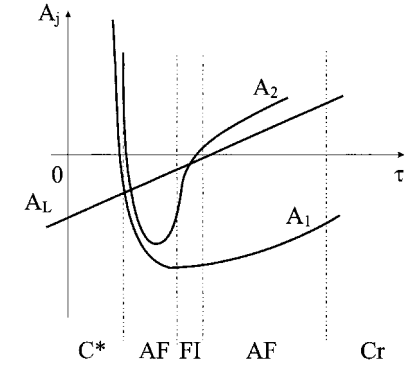

(*b*)

Figure 2. The two SPM and LPM branches of energy as functions of the reduced temperature. Two cases for the SPM lines are shown: (*a*) with intersection, (*b*) without intersection. The suppression of LPM by strong SPM in the high temperature region is assumed. The slopes of the SPM curves essentially differ in low and high temperature regions. The SmC* phase, two antiferroelectric phases and one ferrielectric phase are present.

at a high temperature, i.e. near the point of the transition to the SmC* phase, both the SPM are mixed with LPM at high temperatures. Both figures 1 and 2 describe two hypothetical situations when lines $A_2(\tau)$ and $A_1(\tau)$ are intersecting (*a*) and are not intersecting (*b*). The difference between figures 1 and 2 is the following: at the high temperature points of intersection with line $A_L(\tau)$, the slopes of lines $A_2(\tau)$ and $A_1(\tau)$ in figure 1 are less than in figure 2, and, correspondingly, the temperature range of comparable values of these functions is broader in figure 1 than in figure 2. At a lower temperature, i.e. at large values of τ, the values of functions $A_2(\tau)$ and $A_L(\tau)$ are rather more comparable than the values of functions $A_1(\tau)$ and $A_L(\tau)$ (in both figures). It should be noted, that at low temperatures, i.e. at large values of τ, the slopes of these lines can change and be less due to the presence of higher order terms, for example τ^3, in functions $f_n(\tau)$.

In accordance with the discussed properties of the effective intermode interaction (see equations (21)–(23) in [11]), we can expect that a rapid increase in the amplitudes Θ_1 and Θ_2 in the vicinity of the high temperature points of the intersections of line $A_L(\tau)$ with lines $A_1(\tau)$ and $A_2(\tau)$ (see figure 2) practically suppresses LPM, but there are some temperature intervals for the coexistence of these modes in the phase diagrams shown in figure 1. Thus, in the latter case, the high temperature FI(2L) and FI(12L) phases may arise [see figures 1(*a*) and 1(*b*) respectively], since here relatively weak 1- and 2-SPM induce the LPM, but such phases are absent in the phase diagrams shown in figure 2, since here strong 1- and 2-SPM suppress the LPM. At low temperatures, for all the phase diagrams, a certain competition between the 2-SPM and 1-SPM occurs for survival of the LPM: a weaker 2-SPM induces the LPM, but a stronger 1-SPM suppresses it [11]. In both figures 1 and 2, therefore, a possibility for occurrence of the FI(12L) state exists at low temperatures. At lower temperatures, only the AF(1) phase is present. In the vicinity of deep minima of the $A_2(\tau)$ and $A_1(\tau)$ functions, the AF(12) phase occurs in figures 1(*b*) and 2(*b*), and the AF(2) phase occurs in figures 1(*a*) and 2(*a*). The AF(12) phase may also be present at low temperatures [see figures 1(*b*) and 2(*b*)] if a mixing of the 1-SPM and 2-SPM takes place.

These phase diagrams must result in a different dielectric spectroscopy. First of all, the 'quasi-soft-mode' frequencies ω_{s1} and ω_{s2} [see equation (2)] must occur in the antiferroelectric phases AF(1) and AF(2), respectively. In the low temperature FI phase, the strongest 1-SPM-Goldstone mode must be observed, but the observation of the low-frequency 2-SPM-Goldstone mode [see equation (8)], as well of the very weak LPM-Goldstone mode (see figures 1 and 2), may be impossible. In the high temperature FI phase, the 1-SPM- and LPM-Goldstone modes must be observable, the frequency ω_{GL} being much more than the frequency ω_{G1}. Because of a strong twisting of the high temperature FI state and SmC* phase, the 'soft modes' may be non-observable in both phases.

The hypothetical antiferroelectric phase AF(12) has a rather complicated structure due to the coexistence of the 1-SPM and 2-SPM, the unit cell being a combination of the orientation stars with zero-value total polarization and a certain value of the effective tilt angle [10]. Unfortunately, because of the unknown interaction between these modes, we cannot exactly describe the activation of various relaxation modes in such a unit cell. If the 2-SPM is weak, the AF(12) phase possesses the ω_{s1} relaxation frequency. The latter mode must also occur and be observable in the low temperature ferrielectric phase FI(12L) if the helicoidal mode is sufficiently weak in this phase. But the observation of both the soft-like relaxation modes with frequencies ω_{s1} and ω_{s2} in the AF(12) and FI(12L) phases is under question. Probably, certain complex soft-like perturbations of the unit cell result in such a collective mode.

Some molecular motions in the individual unit cells may be responsible for a relaxation process [9] related to perturbations of the stars. To check the effects of molecular motions, the effect of decrease in chirality should be applied to the substances studied. For instance, in a racemic mixture with $\alpha = 0$, the phase diagrams discussed must radically change, only the SmC and SmC_A phases being observable. In such a case, the molecular mode may be measured in the latter phases which have no macroscopic twisting. In the SmC_A phase, in which the 'antiferro-packing' of dipoles occurs only locally, the induced molecular motions may be related to the tumbling of dipoles and molecules under the action of the electric field. During the process of such a tumbling of molecules around their short axes, the tilt angle and monolayer thickness do not change.

4. Discussion of experimental data

We assume that such an approach can explain the experimental results obtained in ref. [13] and in the present work (see figure 3). In the substance AS-573 [13], the SmC* phase, two FI phases and two AF phases were observed. In substance MHPBC (this work), the SmC* phase, two AF phases and one FI phase in between the AF phases were observed. For both the AFLC, the conventional LPM-Goldstone mode with frequency ω_{GL} is only observed well in the SmC* phase (see figure 3).

For AS-573 [13], in the high temperature FI phase, two relatively strong Goldstone modes are observed with frequencies ω_{GL} (close to ω_{GL} in the SmC* phase) and ω_{G1} which is one order less than ω_{GL}. We ascribe this second relaxation mode to the 1-SPM-Goldstone mode in the SPM model and recall that the corresponding relaxation frequency ω_{G1} must be really much less than ω_{GL}, see equation (8). Since the 2-SPM-Goldstone mode is weaker than the 1-SPM-Goldstone mode [compare $A_2(\tau)$ and $A_1(\tau)$ in figures 1 and 2] and ω_{G2} is much less than ω_{GL} and ω_{G1} [see equation (7)], the 2-SPM-Goldstone mode is hardly observable. The 'quasi-soft-modes' are hardly observable because of twisting of this FI phase.

In the high temperature AF phase, two relaxation modes with relatively large frequencies were observed for AS-573 [13], but only the highest one is observed by us (see figure 3). For AS-573, we ascribe the highest mode to the 'quasi-soft mode' frequency ω_{s1}, discussed above. The second frequency is much less than the value of ω_{s1} [13]. If the second mode is the collective one [13], for instance a 'quasi-soft mode', then its frequency

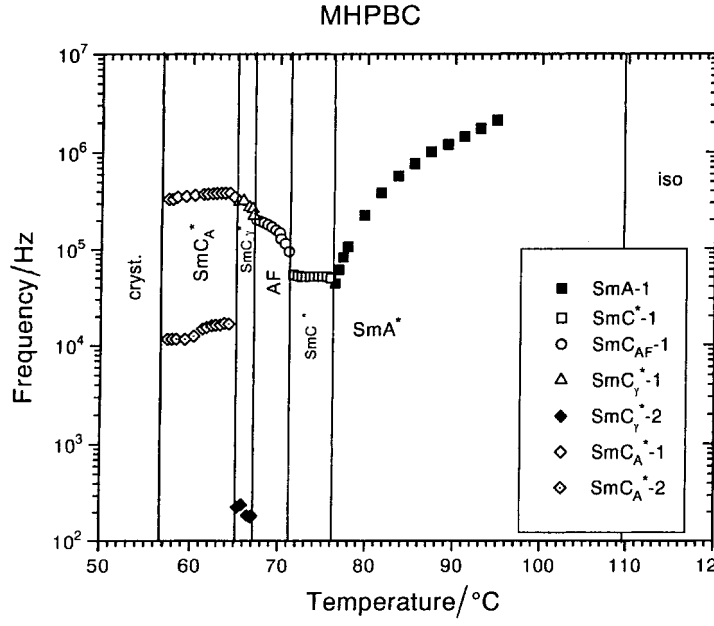

Figure 3. Fitting results for applying up to three terms of a Cole–Cole function to the measured dielectric spectra of MHPBC [4-(1-methylheptyloxycarbonyl)-phenyl 4′-octylbiphenyl-4-carboxylate] versus temperature. For obtaining these dielectric data, 25 µm thick EHC-cells (E.H.C. Ltd. Tokyo, Japan) with ITO-electrodes covered with rubbed polyimide to obtain a unique planar alignment were used.

ω_{s2} could be much less than ω_{s1} in accordance with the different $|A_j|$ values (see figures 1 and 2). Besides, the weakness of 2-SPM could be a reason for the observation only of 1-SPM in the present work. But our study of the racemic variant of substance AS-573 shows that the second relaxation mode is present in the SmC_A and SmC phases, while the other modes disappear. Therefore, we think that this relaxation mode is rather a molecular mode.

In the low-temperature FI phase, two relaxation modes with drastically different frequencies are observed (see figure 3 and ref. [13]). The highest frequency is close to the ω_{s1} value in the high temperature AF phase and to the similar value in the low temperature AF phase; therefore we ascribe this strong mode to the 1-SPM-soft mode with frequency ω_{s1}. A molecular mode is not observable here, probably because of its weakness and the twisting of this FI phase. The weak LPM-Goldstone mode (see figures 1 and 2) is also not observable here. But a stronger SPM-Goldstone mode may be observable though its frequency is much less than ω_{GL}. Therefore, here we ascribe the lowest measured frequency to the SPM-Goldstone mode frequency ω_{G1} since it is close to the Goldstone mode relaxation frequency, observed in the high temperature FI phase for AS-573 [13]. Besides, we conclude that the phase diagram for AS-573 [13] probably corresponds to figure 1(*b*), showing the existence of two FI phases with comparable frequencies for the 1-SPM-Goldstone mode. The phase diagram shown in figure 3 is likely to correspond to figure 2(*a*) showing only the low temperature FI phase with the 1-SPM-Goldstone mode. Furthermore, figure 2(*a*) shows that the high temperature AF phase possesses the soft-like mode with frequency ω_{s2}, which is noticeably less than the ω_{s1} value ascribed to the substance AS-573.

In the low-temperature AF phase, two relaxation modes are observed (see figure 3 and ref. [13]). The highest mode has a frequency which is close to the discussed magnitude ω_{s1} characterizing the high temperature AF phase in AS-573 and the low temperature FI phases in both substances. Therefore, we ascribe this mode to the 1-SPM-soft-like mode we have mentioned. The second relaxation mode is present in both the SmC_A^* and racemic SmC_A phases, and it should be ascribed, as discussed above, to a certain molecular mode. Recent investigations [14] show that the third very weak relaxation mode ('zero' mode) is also observable in the low temperature AF phase, and this Goldstone-like mode may be ascribed to a weak proper incommensurability of such a phase as discussed above.

It is useful to discuss the temperature dependences of the relaxation frequencies. Equations (5), (6) and (8) show that the Goldstone modes have relatively smooth temperature dependences related to the effective τ-dependence of the correlation length λ, see equation (6); all these frequencies have a tendency to decrease with increasing temperature. The experimental data (see figure 3 and ref. [13]) demonstrate such a temperature behaviour. But the temperature behaviour of 'quasi-soft modes' is rather complicated and determined by the temperature dependences of the $|A_j|$ amplitudes and viscosities. According to figures 1 and 2, there is a general tendency for these amplitudes to decrease when the temperature approaches the limits of existence of tilted smectic phases,

and to increase when the temperature corresponds approximately to a central part of this temperature interval. The experimental data (see figure 3 and ref. [13]) show such a tendency.

5. Conclusions

Due to the chiral and polar interactions inside the correlation area, the real wave numbers q_n and energies of the short pitch packings slightly differ from the basic ones: the wave numbers become incommensurate and change for small magnitudes δq_n which are the measures of the incommensurate structures, and the energies of such structures are shifted at magnitudes $r\delta q_n^2$ where parameter r characterizes an energy scale of the incommensurate structures with respect to the energy scale R of the homogeneous part of the free energy. Magnitudes $r\delta q_n^2$ are analogous to magnitudes Kq_L^2 characterizing the shift of the SmA–SmC* phase transition temperature; the first magnitudes are proportional to the relaxation frequencies of the Goldstone-like modes, while the latter one is proportional to the relaxation frequency of the Goldstone mode in the conventional SmC* phase with a long pitch, i.e. with small wave number q_L. The soft mode-like behaviour of the relaxation modes in various SPM structures is also possible, the corresponding frequencies being dependent on the amplitudes of the SPM modulations.

The dielectric spectroscopy of such materials is described by quasi-soft modes and quasi-Goldstone modes with their relaxation frequencies determined by the main physical parameters discussed. These relaxation modes correspond to the observed phase diagrams. For instance, different quasi-soft modes are observed in the low temperature antiferroelectric phases, and several Goldstone modes are observed in the high temperature ferrielectric phase. The tendencies of a relatively smooth monotonous temperature change of the relaxation frequencies for Goldstone-like modes and of a more rapid temperature change (with a maximum at an intermediate temperature) of the relaxation frequencies for soft-like modes are demonstrated.

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