# Light scattering and the dispersion of susceptibilities in an incommensurate phase

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A consistent investigation of light scattering and the dispersion of the dielectric and elastic susceptibilities in the incommensurate phase is carried out. It is shown that the Goldstone excitation (phason) is always active in light scattering whether or not there is a change in the polarization. The intensity of the phason line vanishes at the symmetric–incommensurate phase transition point, and depends essentially on the direction of the scattering vector. In the scattering spectrum the phason line is a narrow central peak whose width is proportional to the square of the scattering vector. The phason also contributes always to the elastic susceptibility, but its contribution to the dielectric susceptibility is nonzero only if the crystal is piezoelectric in the symmetric phase. These contributions depend essentially on the direction of propagation of the elastic or electromagnetic waves, and manifest themselves only at very low frequencies (of the order of a hertz or a fraction of a hertz). The question of the appearance in the incommensurate phase of new spectral lines corresponding to non-Goldstone excitations is also considered. The selection rules are determined, and the intensities of the new lines and the scattering geometry in which they can be observed are discussed. The temperature dependence of the frequencies corresponding to these lines is also discussed. The analysis is carried out primarily for the case of a two-component order parameter, but the distinctive features that arise when the order parameter has a large number of components are analyzed also in the particular case of a four-component order parameter.

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The experimental investigations of light scattering and dielectric dispersion in incommensurate phases have greatly gained in scope (see, for example, Refs. 1–11). At the same time, the corresponding theory seems to us to be insufficiently developed. For example, the literature contains the most diverse opinions about the manifestation in such experiments of the characteristic excitation of the incommensurate phase, viz., the Goldstone phason mode. Thus, Cowley, actually asserts in his review article[12] that the phason mode is equivalent to an acoustic mode, and should similarly manifest itself in Mandel'shtam–Brillouin scattering. On the other hand, it is claimed in Refs. 13–15 that the Goldstone mode is inactive in light scattering and light absorption. The opinion also exists[3] that the phason is active in light scattering only if the crystal does not possess a center of inversion. The absence of a definite theory also leads to lack of coordination in the interpretation of experiment. Some experimenters have sought the manifestation of phasons in light scattering accompanied by a change in the polarization[1,4,6,7]; others, in light scattering not involving a change in the polarization.[2,3] Attempts have been made to detect the contribution of the phason to both the central peak[2,3] and the Mandel'shtam–Brillouin sideband components.[11]

The purpose of the present paper is to give a consistent and, as far as possible, exhaustive description of light scattering and the dispersion of the dielectric and elastic susceptibilities in the incommensurate phase on the basis of a phenomenological theory that goes back to the Landau theory of phase transitions. Let us emphasize that such an approach is standard, having been repeatedly used to investigate light scattering in the vicinity of phase-transition points.[14-16] As we shall see, the specific nature of the incommensurate phase manifests itself only in the necessity of the consideration of certain terms containing the spatial derivatives of the order parameter, which are usually neglected since they turn out to be insignificant in the analysis of commensurate phase transitions.

We note that in such an approach, as always in the Landau theory, we need consider only the symmetry of the high-temperature phase (which is the normal symmetry), the properties of the incommensurate phase being then determined automatically. In particular, it is not at all necessary to describe the symmetry of the incommensurate phase with the aid of the superspace groups that have been introduced in a number of papers (see, for example, Ref. 17).

## 1. MANIFESTATION OF THE PHASON IN THE CASE OF A TWO-COMPONENT ORDER PARAMETER

There are in the literature two equivalent methods of describing the transition into the incommensurate phase. In the unified description of the most frequently observed case of two transitions—from the high-temperature phase into the incommensurate phase, and from this phase into the low-temperature commensurate phase—the incommensurate phase is treated as a spatial modulation of the commensurate phase, and only the order parameter corresponding to the high-temperature—commensurate phase transition is introduced. The structure of the incommensurate phase is then described as an inhomogeneous structure with a "frozen-in" order-parameter wave, and a special role is played in the study of the transition into the incommensurate phase by the Lifshitz invariant, which is that term in the thermodynamic-potential density which contains the first derivatives of the order parameter.[18,19] The presence of the Lifshitz invariant makes a

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second-order transition into a homogeneous (commensurate) phase impossible, and, as a result, the incommensurate phase arises. On the other hand, we can introduce an order parameter corresponding directly to the high-temperature—incommensurate phase transition.[20,21] Such an order parameter is the normal coordinate $Q(\mathbf{K}_0)$ corresponding to the wave vector $\mathbf{k}=\mathbf{K}_0$ of the incommensurate superstructure formed just below the high-temperature—incommensurate phase transition temperature $T_i$. This parameter is spatially homogeneous in the incommensurate phase, even though the Lifshitz invariant is allowed by the symmetry, since the value of $\mathbf{K}_0$ is characterized by the fact that, for such $\mathbf{k}$, the coefficient of the Lifshitz invariant in the thermodynamic potential vanishes at the transition point. It is precisely the second approach that will be convenient for us to use in the present paper.

The order parameter used has at least two components, since the vector $\mathbf{K}_0$ lies inside the Brillouin zone of the high-temperature phase and the star of the physically irreducible representation contains a vector $-\mathbf{K}_0$ that is not equivalent to it [the basis functions of such a representation are proportional to $\exp(\pm i\mathbf{K}_0\cdot\mathbf{r})$]. We note that, for the majority of dielectrics with incommensurate superstructures,[22,23] the order parameter is precisely a two-component one. The case in which the order parameter has more than two components will be discussed in the third section. As the components of the order parameter we can take $Q(\mathbf{K}_0)$ and $Q(-\mathbf{K}_0)=Q^*(\mathbf{K}_0)$.

Since $Q$ and $Q^*$ correspond to one and the same frequency $[\omega(\mathbf{k})=\omega(-\mathbf{k})]$, the corresponding vibrations can formally be considered to be degenerate. This degeneracy is lifted below the phase-transition point because of the formation of the superstructure, which leads to the splitting of the branches and the replacement of the single soft-mode frequency by two frequencies. One of them remains equal to zero in the entire region of existence of the incommensurate phase (the Goldstone mode), and the corresponding excitation is called a phason, while the other increases with decreasing temperature, i.e., behaves just like the frequency of the soft mode associated with a commensurate phase transition, and the corresponding excitation is called an amplitudon.[24-26] Figure 1 shows the Brillouin zone and the phason 1 and amplitudon 2 branches below $T_i$, the normal coordinate $Q$ corresponding to the point $A$. If we set $Q=\rho e^{i\varphi}$, then the phason represents the oscillations of the phase $\varphi$, while the amplitudon represents the $\rho$ oscillations.

Let us see how these excitations manifest themselves in light scattering and susceptibility dispersion. As is well known, light scattering is connected with fluctuations of the permittivity tensor $\varepsilon_{ik}$ (Refs. 27 and 16). To elucidate the manifestations of these excitations in the scattering spectra, we must consider first those changes in $\varepsilon_{ik}$ which are due to changes in the order parameter. To do this, we must determine the dependence of $\varepsilon_{ik}$ on the order-parameter components, i.e., find out which combinations of these components transform like the quantities $\varepsilon_{ik}$, which, like the components

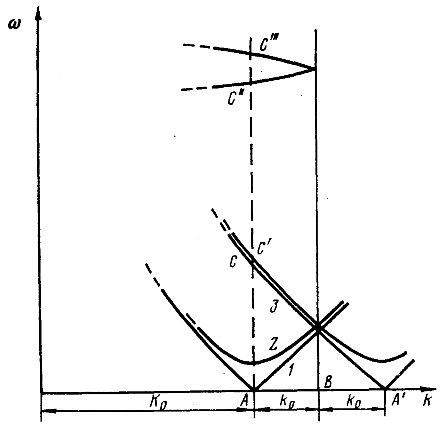


FIG. 1. Brillouin zone of the symmetric high-temperature phase. The point B represents the zone boundary; a portion of the branches has been continued into the second zone. The bottom branches are for $T<T_i$.

of any tensor or vector, are invariant under translation (let us recall that we are now talking about the dependence of $\varepsilon_{ik}$ on $Q$ in the symmetric phase). Similarly, in order to find out whether the excitations under discussion will make a contribution to the dielectric or elastic dispersion, we must find out whether there can be any coupling between the order parameter and the components of the polarization vector $\mathbf{P}$ or the strain tensor $u_{ik}$. Let us note that, if we assume that the vector $\mathbf{P}$ depends on the normal coordinates, then we actually have dielectrics in mind, since we do not then take into account the contribution to the polarization of the conduction electrons.

If the vector $\mathbf{K}_0$ is not commensurate with any reciprocal-lattice vector of the symmetric phase, then the only quantities that are invariant under translation in the symmetric phase, and depend on, but do not contain derivatives of, the order parameter are the quantity $QQ^*=\rho^2$ and its powers. The invariants containing spatial derivatives of the order parameter play an insignificant role and are, as a rule, neglected in the investigation of light scattering and susceptibility dispersion in the vicinity of the critical points of normal (commensurate) phase transitions. Because $QQ^*$ does not contain the phase $\varphi$, it is concluded in Refs. 14 and 15 that the phason is inactive.[1)] But this conclusion is incorrect, since, as we shall see below, the translation invariants containing linear combinations of the first space derivatives of the order parameter (we shall call them gradient invariants) also play an important role in light scattering and susceptibility dispersion in the incommensurate phase.

Let us discuss the question of the activity of the phason first for light scattering. The simplest gradient invariant is

$$\partial(QQ^*)/\partial x_i=\partial\rho^2/\partial x_i.$$

It does not depend on $\varphi$, and is therefore of no interest in connection with the question of the activity of the phason. There, however, exist gradient invariants that contain the phase, e.g., the Lifshitz invariant:

$$\frac{i}{2}\left(Q\frac{\partial Q^*}{\partial x}-Q^*\frac{\partial Q}{\partial x}\right)=\rho^2\frac{\partial\varphi}{\partial x},$$

where by $x$ we mean the axis directed along the wave vector $\mathbf{K}_0$ of the incommensurate superstructure. From the fact that the Lifshitz invariant is invariant not just under translation, but under the full symmetry-transformation group of the high-temperature phase (we shall call such invariants full invariants) it follows that the phase $\varphi$ transforms like $x$. Therefore, the quantities $\rho^2\partial\varphi/\partial x_i$ are also gradient invariants that transform like the elements of a second-rank tensor. There are no other gradient invariants of second order in $Q$ in the case of the two-component order parameter.

Let us, for concreteness, limit ourselves to crystals whose symmetric phases belong to the orthorhombic system, assuming that the vector $\mathbf{K}_0$ is parallel to one of the crystallographic axes, as in the majority of dielectrics possessing the incommensurate phase.[22,23] The case of other systems can similarly be discussed without any difficulty. We consider first the $\varepsilon_{ik}$ diagonal-element fluctuations that cause light to be scattered without a change in the polarization if the incident light is polarized along one of the crystallographic axes.[16] Since in the indicated system such elements are full invariants, their dependence on $\varphi$ can be due only to the presence of the Lifshitz invariant, e.g.,

$$\varepsilon_{xx}=\varepsilon_{xx}^{(0)}+a\rho^2\frac{\partial\varphi}{\partial x}. \quad (1)$$

The corresponding integrated intensity of the scattered light is proportional to the mean square fluctuation of the Fourier component $\varepsilon_{xx}(\mathbf{q})$, where $\mathbf{q}$ is the scattering vector, i.e., the difference between the wave vectors of the incident and scattered light.[27,16] Using (1), we find the mean square $\varepsilon_{xx}(\mathbf{q})$ fluctuation due to the $\varphi$ fluctuations:

$$\langle|\Delta\varepsilon_{xx}(\mathbf{q})|^2\rangle=a^2\rho_e^4q_x^2\langle|\varphi(\mathbf{q})|^2\rangle, \quad (2)$$

where $\rho_e$ is the equilibrium value of $\rho$ and $\varphi(\mathbf{q})$ is the Fourier transform of the fluctuation $\Delta\varphi$ of the phase.

To compute the $\varphi(\mathbf{q})$ fluctuations, let us write the thermodynamic potential density in the variables $\rho$ and $\varphi$:

$$\Phi=\frac{\alpha}{2}\rho^2+\frac{\beta_1}{4}\rho^4+\frac{1}{2}\sum_{i=1}^{3}\delta_i\left[\left(\frac{\partial\rho}{\partial x_i}\right)^2+\rho^2\left(\frac{\partial\varphi}{\partial x_i}\right)^2\right]. \quad (3)$$

Notice that, for the chosen order parameter, the Lifshitz invariant does not enter here (see above). Proceeding in standard fashion,[16] we find on the basis of (3) the mean square fluctuation

$$\langle|\varphi(\mathbf{q})|^2\rangle=\frac{k_BT}{V\rho_e^2(\delta_1q_x^2+\delta_2q_y^2+\delta_3q_z^2)}, \quad (4)$$

where $V$ is the volume of the crystal (see also Ref. 15). Substituting this expression into (2), we obtain

$$\langle|\Delta\varepsilon_{xx}(\mathbf{q})|^2\rangle=\frac{a^2\rho_e^2q_x^2k_BT}{V(\delta_1q_x^2+\delta_2q_y^2+\delta_3q_z^2)}. \quad (5)$$

Let us note that the expressions for the fluctuations of the other diagonal elements of $\varepsilon_{ik}$ will differ only in the values of the coefficient $a$, i.e., the numerator will contain $q_x^2$ as before.

Let us now consider the off-diagonal elements of $\varepsilon_{ik}$, the fluctuations of which cause polarization-nonpreserving light scattering.[16] In accordance with the assertion made above, their dependence on $\varphi$ is given by the relations

$$\varepsilon_{xy}\sim\rho^2\frac{\partial\varphi}{\partial y},\quad \varepsilon_{xz}\sim\rho^2\frac{\partial\varphi}{\partial z}.$$

The element $\varepsilon_{yz}$, on the other hand, does not at all depend on $\varphi$. Proceeding as above, we obtain formulas of the type (5) for the mean square fluctuations, but the quantity $\langle|\Delta\varepsilon_{xy}|^2\rangle$ will have in its numerator the factor $q_y^2$ in place of $q_x^2$, while the quantity $\langle|\Delta\varepsilon_{xz}|^2\rangle$ will contain $q_z^2$.

Proceeding to the discussion of the results, we point out first that, in contrast to light scattering in the vicinity of normal phase transition points, the terms in $\varepsilon_{ik}$ that depend on the derivatives of the order parameter make a contribution that does not vanish in the limit $\mathbf{q}\to 0$. This is due to the fact that the phase fluctuations (4) diverge as $\mathbf{q}\to 0$. It is also clear that allowance for powers of the first or higher derivatives in the dependence of $\varepsilon_{ik}$ on the order parameter is of no interest, since the contribution of such terms will vanish at $\mathbf{q}\to 0$. An interesting characteristic of light scattering by phasons is the critical dependence of its intensity on the direction of $\mathbf{q}$, a dependence which does not disappear in the limit $\mathbf{q}\to 0$. In other words, the $\mathbf{q}$ dependence of the scattering intensity is not analytic at $\mathbf{q}\to 0$, and it is in this circumstance that the similarity between the phason branch and the acoustic branches in a solid manifests itself, although the characters of the $\mathbf{q}$-direction dependences in these two cases are, of course, different. Since $\rho_e^2$ stands in the numerator of (5), the intensity is low near the transition point, and increases in proportion to $T_i-T$ as the temperature is lowered.[2)] Let us also note that the phason contributes to both polarization-preserving and polarization-nonpreserving scattering of light. Contrary to the assertion made in Ref. 3, the activity of the phason has nothing to do with the absence or presence of a symmetry center in the crystal.

Let us emphasize that the formula (5) only reflects in a simplified manner the character of the dependence of the quantity $\langle|\Delta\varepsilon_{xx}(\mathbf{q})|^2\rangle$ and the scattered-light intensity proportional to it on the direction of the vector $\mathbf{q}$. The point is that, as noted in Ref. 16, we should, in considering light scattering below the phase transition point, take into account the dependence of the tensor $\varepsilon_{ik}$ not only on the order parameter, but also on the other quantities whose fluctuations are linearly connected with the order-parameter fluctuations (such a quantity is, for example, the strain tensor). This allowance leads to the renormalization of the coefficients in expression (5), and it is significant that this renormalization has a nontrivial character: the noted nonanalyticity at $\mathbf{q}\to 0$ may occur in the renormalized coefficients as well. As a result, the angular dependence of the scattered-light intensity will be quite complicated, but the main features of this dependence are accurately reflected by the formula (5): the intensity vanishes at $q=0$. Let us note here that a similar observation can be made about the role played by other quantities together with the order parameter in the problem, considered below, of the dispersion of the dielectric and elastic susceptibilities.

Let us now discuss the spectral intensity of light scattered by phasons. As usual, we introduce the kinetic energy $K$ and the dissipation function $R$ that correspond to the normal coordinate $Q$:

$$K=\frac{\mu}{2}\frac{\partial Q}{\partial t}\frac{\partial Q^*}{\partial t}=\frac{\mu}{2}\left[\left(\frac{\partial\rho}{\partial t}\right)^2+\rho^2\left(\frac{\partial\varphi}{\partial t}\right)^2\right],\quad R=\frac{\nu}{2}\frac{\partial Q}{\partial t}\frac{\partial Q^*}{\partial t}. \qquad (6)$$

Since the phason and the amplitudon are normal excitations of the incommensurate phase,[24-26] as can also be seen directly from (3), the equations of motion for $\Delta\varphi$ and $\Delta\rho$ separate, and for $\Delta\varphi$ we have

$$\mu\frac{\partial^2\Delta\varphi}{\partial t^2}+\nu\frac{\partial\Delta\varphi}{\partial t}-\sum_{i=1}^{3}\delta_i\frac{\partial^2\Delta\varphi}{\partial x_i^2}=h_\varphi(\mathbf{r},t),$$

where $h_\varphi$ is the corresponding generalized force. Using the standard procedure,[16] we find the spectral density of the fluctuations

$$\langle|\varphi(\mathbf{q},\Omega)|^2\rangle=\frac{\nu k_BT}{\pi V\rho_e^2[(\mu\Omega^2-\delta_1q_x^2-\delta_2q_y^2-\delta_3q_z^2)^2+\nu^2\Omega^2]}. \qquad (7)$$

Since, as can be seen from (6), the quantity $\nu$ has the meaning of a damping constant for the optical phonon in the symmetric phase and therefore there is no reason why it should not be large (see also below), the quantities $\mu\delta_iq_i^2$ are negligible in comparison with $\nu^2$. Now we can see from (7) that the spectral density $\langle|\varphi(\mathbf{q},\Omega)|^2\rangle$ decreases monotonically as the frequency $\Omega$ increases, i.e., the $\varphi$ fluctuations have a relaxation character. In the most interesting region (low $\Omega$) we have from (7) the expression

$$\langle|\varphi(\mathbf{q},\Omega)|^2\rangle=\frac{\nu k_BT}{\pi V\rho_e^2[(\delta_1q_x^2+\delta_2q_y^2+\delta_3q_z^2)^2+\nu^2\Omega^2]}. \qquad (8)$$

It can be seen from this that the width of the central peak of the spectrum of the $\varphi$ fluctuations is proportional to $(\delta_1q_x^2+\delta_2q_y^2+\delta_3q_z^2)/\nu$, as in the case of fluctuations of a diffusional nature. The spectral intensity of the scattered light can be obtained from (2) by replacing $\langle|\varphi(\mathbf{q})|^2\rangle$ by $\langle|\varphi(\mathbf{q},\Omega)|^2\rangle$, $\Omega$ being the scattering-induced change in the light frequency.

We emphasize that, although the phason branch is sometimes called a new acoustic branch in the incommensurate phase, the scattering of light by phasons differs significantly from Mandel'shtam–Brillouin scattering, since the phasons contribute a central peak in the spectrum rather than sideband components. This is due to the difference in the phason and acoustic-phonon damping. If the wave vector $\mathbf{q}$ tends to zero, the acoustic vibration does not lead to energy dissipation, since such a vibration corresponds to the displacement of the whole crystal without a change in the relative positions of the atoms. On the other hand, at $\mathbf{q}\to 0$ the phason corresponds to a shift, as a unit, of the static wave of atomic displacements occurring below the phase transition point. The various positions of this wave in the crystal are energetically equivalent, i.e., the phason frequency tends to zero as $\mathbf{q}\to 0$, but the transition from one position of the wave to another is connected with changes in the relative positions of the atoms, i.e., with dissipative processes. A reflection of this is that the phason-damping constant $\nu$ is virtually independent of the wave vector $\mathbf{q}$.

Let us discuss the width of the phason central peak, whose maximum value $\Delta\Omega_{max}$ can be estimated by taking as $\mathbf{q}$ the wave vector of light waves. We have

$$\Delta\Omega_{max}\approx\frac{\delta_iq^2}{\nu}=\frac{\delta_ik_a^2}{\nu}\frac{q^2}{k_a^2}\sim(1-10)\frac{q^2}{k_a^2}\omega_a\sim(10^{-6}-10^{-5})\,\omega_a. \qquad (9)$$

Here $k_a$ is the dimension of the Brillouin zone of the symmetric phase, and we have taken into account the fact that $\delta_ik_a^2/\mu\sim\omega_a^2$, where $\omega_a$ is the characteristic phonon frequency ($\sim 10^{13}$ Hz), and the fact that the $Q$ factor for normal optical phonons $\omega_a\mu/\nu\sim 1-10$. It can be seen from this that even the maximum width of the phason peak is quite small.

This difference between scattering by phasons and Mandel'shtam–Brillouin scattering has not been noted in the literature, not even where it is asserted that the phason is active in light scattering (see, for example, Ref. 12). As a result, in some experiments[11] the manifestation of the phason was sought in the lateral Mandel'shtam–Brillouin components, and not in the central peak. In a number of experiments,[1,4,6] attempts were made to detect the scattering by the phasons in the incommensurate phase in the same scattering geometry and in the same frequency range as for the phasons in the low-temperature commensurate phase. The negative result of these experiments is entirely natural, since in the commensurate phase the phason-induced scattering is practically determined by those terms in the expansion of $\varepsilon_{ik}$ in powers of the order parameter which do not contain derivatives, and do not occur in the incommensurate phase, a fact which can result in the manifestation of the phason in another scattering geometry. Furthermore, the phason frequency in the commensurate phase does not vanish at $\mathbf{q}\to 0$, i.e., such a phason contributes in the scattering either sideband components or a central peak whose width does not differ much from $\omega_a$ and does not depend on $\mathbf{q}$.

We note that allowance for the coupling of the order-parameter fluctuations to the fluctuations of other quantities does not lead in the investigation of the spectral density of the phason-scattered light to changes of a qualitative nature. This is due to the fact that the phason relaxation rate $\delta_iq^2/\nu$ is much lower than the other characteristic frequencies of the crystal that correspond to the given $\mathbf{q}$, i.e., the variations of all the other quantities have time to adjust themselves to the variation of $\varphi(\mathbf{q})$.

Let us proceed to discuss the contribution of the phason to the dispersion of the dielectric susceptibility. To do this, we must first find out which combinations of the order-parameter components transform like the components of the polarization vector $\mathbf{P}$. Above we said that the $\varphi$-dependent translation invariants have the form $\rho^2\partial\varphi/\partial x_i$, transforming like the elements of a second-rank tensor. Therefore, they can be linearly coupled to $\mathbf{P}$ only if the symmetric phase belongs to the piezoelectric class, since some of the strains leading to the piezoelectric effect are described by at least one of the components $u_{xx}$, $u_{xy}$, or $u_{xz}$.

Assume that this condition is fulfilled; then some component $P_i$ transforms like $\rho^2\partial\varphi/\partial x_j$. Retaining in the thermodynamic potential only the terms that are important for what follows, we have

$$\Phi=\frac{\rho_e^2}{2}\sum_{k=1}^{3}\delta_k\left(\frac{\partial\varphi}{\partial x_k}\right)^2+a_1P_i\rho_e^2\frac{\partial\varphi}{\partial x_j}+\frac{\varkappa}{2}P_i^2-P_iE_i. \quad (10)$$

Since, as we shall see below, the region of extremely low frequencies is the one of importance, we can neglect the intrinsic inertia and the viscosity of the polarization. Now, varying (10) with respect to $P_i$, we find

$$P_i=-\frac{a_1\rho_e^2}{\varkappa}\frac{\partial\varphi}{\partial x_j}+\frac{E_i}{\varkappa}. \quad (11)$$

Substituting this in (10), varying the resulting expression with respect to $\varphi$, and taking (6) into account, we obtain an equation relating $\varphi$ with the field intensity $\mathbf{E}$:

$$\mu\frac{\partial^2\varphi}{\partial t^2}+\nu\frac{\partial\varphi}{\partial t}-\sum_{k=1}^{3}\delta_k\frac{\partial^2\varphi}{\partial x_k^2}=\frac{a_1}{\varkappa}\frac{\partial E_i}{\partial x_j}. \quad (12)$$

From (11) and (12) we find the dielectric susceptibility

$$\chi(\mathbf{q},\omega)=\frac{1}{\varkappa}+\frac{a_1^2\rho_e^2q_j^2}{\varkappa^2(\delta_1q_x^2+\delta_2q_y^2+\delta_3q_z^2-\mu\omega^2-i\omega\nu)}, \quad (13)$$

where $\mathbf{q}$ and $\omega$ are respectively the wave vector and the frequency of the field $\mathbf{E}$.

We examine (13) first in the static (i.e., $\omega=0$) case. The temperature dependence of $\chi$ has a kink at the critical point of the transition into the incommensurate phase, since $\rho_e^2\sim(T_i-T)$. Note that a similar—in its dependence on $T$—contribution to $\chi$ is made not only by the above coupling of $\mathbf{P}$ with the order parameter, but also by the universal term of the form $\rho^2P_i^2$ in the thermodynamic potential. But the coupling between the $P_i$ and $\varphi$ in (10) gives rise to a contribution to $\chi$ of an essentially different nature. Let us first draw attention to the nonanalyticity of the dependence of $\chi$ on $\mathbf{q}$ at $\mathbf{q}\to 0$, as is the case for the scattered-light intensity. We point out that a nonanalyticity of similar nature is observed in normal piezoelectric crystals.[27]

But, as seen from (13), this nonanalyticity will be manifest only at low frequencies. It disappears at $\omega \gg \omega_0=\delta_i q^2/\nu$, at which point the contribution of the phason to the susceptibility becomes negligibly small. At low frequencies the electromagnetic waves are long; therefore, setting $q\sim 1/L$, where $L$ is the sample dimension, we find for the present case, in much the same way as (9) was found, the estimate

$$\omega_0\approx(1-10)\,\omega_a/(k_aL)^2.$$

For $L\sim 0.1$ cm we have $\omega_0\sim(0.1-1)$ Hz. Let us note that, in normal piezoelectric crystals, the analogous nonanalyticity is observed only at not very high frequencies: it disappears in these crystals at frequencies higher than the piezoresonance frequency, which is equal to $\sim c/L\sim\omega_a/k_aL$ ($c$ is the velocity of sound) and is $k_aL$ times higher than $\omega_0$.

The elastic susceptibilities $s_{iklm}$ can be treated in similar fashion. In contrast to the polarization vector, the strain tensor $u_{ik}$, like the tensor $\varepsilon_{ik}$, is always coupled to the quantities $\rho^2\partial\varphi/\partial x_i$. The expressions for $s_{iklm}(\mathbf{q},\omega)$ are extremely unwieldy, but the results have the same qualitative character as for $\chi$: in particular, there also exists at $\mathbf{q}\to 0$ a nonanalyticity that disappears at frequencies of the order of $\omega_0$. A distinctive feature of the present case is that the correction to $s_{iklm}(\mathbf{q},\omega)$ analogous to the second term in (13) becomes appreciable again at frequencies $\sim\omega_a$, when all the terms in the denominator are of the same order of magnitude; but it hardly makes sense to speak of a phason at such frequencies and at the corresponding $q\sim k_a$.

Note that a strain–phason interaction similar to the one considered by us is mentioned in Ref. 26, but its effect on the elastic-susceptibility anomalies is not discussed there. Perhaps that is why the assertion made in Ref. 26 that the interaction in question is proportional to $|\mathbf{q}|$ is incorrectly interpreted in Ref. 28 as evidence of the vanishing of the interaction between the phason and the acoustic modes in the long-wave limit. In fact, as indicated above, at $\omega=0$ the modulus of $\mathbf{q}$ cancels out in the expression for the correction to the elastic susceptibility.

## 2. NON-GOLDSTONE EXCITATIONS

Let us note first of all that the other characteristic excitation of the incommensurate phase—the amplitudon—manifests itself in light scattering and susceptibility dispersion as a normal, fully symmetric soft mode.[14,15] Light scattering involving the amplitudon has been observed in many experiments,[1,4,6-10,13] and its interpretation offers no difficulty.

We consider now the manifestation in the spectra of other excitations of the incommensurate phase, first for the case of light scattering. The Raman-scattering spectrum, in the case of any phase transition connected with a change in the translational symmetry of the crystal, is known to exhibit new lines, since the number of atoms in the unit cell changes in the transition. For transitions into the incommensurate phase, the entire crystal becomes a single unit cell, and the number of new lines, one would think, should be virtually infinite. We shall show, however, that in fact the number of new lines with observable intensities in the spectrum of the incommensurate phase is relatively small.

We consider first some normal coordinate $Q_1$ not belonging to the soft mode, but pertaining to the same point in the Brillouin zone. In Fig. 1 this coordinate corresponds to one of the $C$ points located above the point $A$. The quantity $QQ^*$ will also be invariant under translation, and, in particular, some elements of the tensor $\varepsilon_{ik}$ may turn out to be proportional to $QQ_1^*$. A spontaneous value of $Q=Q_e\neq 0$ appears below the phase-transition point, and therefore the change in $Q_1$ will give rise to proportional changes in $\varepsilon_{ik}$. This means that frequencies not occurring at $T>T_i$ will appear in the scattering spectrum, since the point $A$ is not located at the center of the Brillouin zone of the symmetric phase. The intensities of these lines are proportional to

$$|Q_e|^2=\rho_e^2\sim T_i-T.$$

The quantity $QQ_1{}^*$ may or may not be a full invariant. In the first case $QQ_1{}^*$ can be coupled to only the diagonal elements of $\varepsilon_{ik}$; in the second, to the off-diagonal elements, which are forbidden in the symmetric phase (we recall that we are dealing with the orthorhombic system). Therefore, the indicated lines can appear in the spectra of light scattered with or without a change in its polarization.

Let us now consider a normal coordinate $Q_2$ corresponding to the wave vector $2\mathbf{K}_0+\mathbf{b}$, where $\mathbf{b}$ is some reciprocal-lattice vector of the symmetric phase, which can be chosen, for example, such that the vector $2\mathbf{K}_0+\mathbf{b}$ lies in the first Brillouin zone. The normal coordinate $Q_2$ may belong to both a high-lying branch and the soft branch. In the present case we can form the translation invariant $Q^2Q_2{}^*$. Proceeding as above, we see that the $Q_2$ vibration can be active in the scattering, but that the intensity of the spectral line is proportional to $|Q_e|^4=\rho_e{}^4$. It is clear that the normal coordinate $Q_n$ corresponding to the wave vector $n\mathbf{K}_0+\mathbf{b}$ with arbitrary $n$ can also be active in the scattering, the intensity of the line being proportional to $\rho_e{}^{2n}$.

Thus, as expected, there will appear in the scattering spectrum of the incommensurate phase an infinite number of new lines (in view of the incommensurability of $\mathbf{K}_0$ and $\mathbf{b}$, no two frequencies will be exactly equal), but only a limited number will in practice be observed, since, as a rule, $\rho_e$ is small in the entire existence domain of the incommensurate phase, and therefore the intensity of the lines will decrease rapidly with increasing $n$. In each specific case, knowing the transformation properties of the order parameter and the normal coordinates $Q_n$, we can find out which element of the tensor $\varepsilon_{ik}$ is coupled to a particular $Q^nQ_n{}^*$, and, consequently, determine the scattering geometry in which the spectral line in question should be observed. Let us note that the appearance in the incommensurate phase of new lines with intensities $\sim\rho_e{}^{2n}$ ($n=2,3,\ldots$) has been considered by Dvořák and Petzelt,[14] but they analyzed only the non-Goldstone excitations belonging to the soft branch, failing to take into account the possibility of the appearance of more intense new lines connected with the $Q_1(\mathbf{K}_0)$ vibrations corresponding to other branches.

In the foregoing discussions we have neglected the frequency splitting due to the appearance of the superstructure. The origin of this splitting in the present case is easily explained as follows. Since there exists the translation invariant $Q(\mathbf{K}_0)Q_1{}^*(\mathbf{K}_0)$, the thermodynamic potential will contain the full invariant $Q^2(Q_1{}^*)^2$ + c.c. There will appear in the thermodynamic potential of the incommensurate phase, where the spontaneous value $Q=Q_e$ arises, $Q_1$-dependent off-diagonal terms, i.e., terms that do not reduce to $|Q_1|^2$, and this leads to the splitting of the frequencies associated with the wave vector $\mathbf{k}=\mathbf{K}_0$. Let us note that the frequency splitting is observed not only at $\mathbf{k}=\mathbf{K}_0$. An even larger splitting occurs at $\mathbf{k}=\mathbf{K}_0/2$ if the translation invariant $Q(\mathbf{K}_0)\,Q_0{}^*(\mathbf{K}_0/2)^2$ + c.c. is a full invariant, and will therefore be present in thermodynamic potential. But this splitting does not appear in optical spectra, since the point $\mathbf{k}=\mathbf{K}_0/2$ corresponds to the edge of the "Brillouin zone" of the superstructure. Let us note that there also occur at $\mathbf{k}=2\mathbf{K}_0+\mathbf{b}, 3\mathbf{K}_0+\mathbf{b}, \ldots$, weaker splittings, which can be explained in similar fashion.

Let us discuss the temperature dependence of the frequencies of the generated "non-Goldstone" lines. The frequency corresponding to the amplitudon increases with decreasing temperature, just as the frequency of a normal mode. The frequencies of the other new lines, which correspond to high-lying optical branches, in the general case depend weakly on temperature, largely as a result of the variation of $\mathbf{K}_0$. On the other hand, they may correspond to both the softest branch (at $\mathbf{k}=2\mathbf{K}_0+\mathbf{b}, 3\mathbf{K}_0+\mathbf{b}, \ldots$) and to a branch genetically related to it (if such a branch exists; see the branch 3 in Fig. 1), and then they may be low and quite strongly dependent on temperature. Let us discuss in greater detail the temperature dependence of the frequencies of such lines, noting that, since the damping constant in the present case is of the same order of magnitude as the damping constant for the other optical branches, the low-frequency lines may actually make a contribution to the central peak, and not be sideband components.

Let us first consider the case in which the end of the vector $\mathbf{K}_0$ lies near the boundary of the Brillouin zone of the symmetric phase, assuming for simplicity that the vector $\mathbf{K}_0$ is parallel to a fundamental vector $\mathbf{b}_1$ of the reciprocal lattice, i.e., that $K_0=b_1/2-k_0$, where $k_0$ is small. It often turns out that the softening optical branch is degenerate at the zone boundary, i.e., that there exists a branch genetically related to it. As can be seen from Fig. 1, the frequency $\omega(K_0)$ of this branch 3 at $k=K_0$ (we actually have two close frequencies to deal with at $T<T_i$), to which corresponds a line with intensity $\sim\rho_e{}^2$, is as small as $k_0$. Let us discuss the dependence of $k_0$ on temperature. At the considered position of $\mathbf{K}_0$ there is usually observed at low temperatures a commensurate phase whose order parameter is the normal coordinate corresponding to $\mathbf{k}=\mathbf{b}_1/2$. If the incommensurate–commensurate phase transition were of second order, $k_0$ together with one of the two frequencies $\omega(K_0)$ would vanish at the critical point of this transition. But in all the published experiments this phase transition is found to be of first order, and $k_0$ is found to change within the limits of existence of the incommensurate phase by several times at most; $\omega(K_0)$ will also change by roughly the same factor. Since in this case $k_0$ is roughly two orders of magnitude smaller than $b_1$, the frequency $\omega(K_0)$ is roughly two orders of magnitude lower than the characteristic frequencies of the crystal.

In the unified description of the transitions from the high-temperature phase into the incommensurate phase and from this phase into the commensurate low-temperature with a doubled lattice constant, the thermodynamic potential is represented in the form of a function of the normal coordinate corresponding to $\mathbf{k}=\mathbf{b}_1/2$, and not $\mathbf{k}=\mathbf{K}_0$, as in the present paper (see the first section). Precisely this approach is used in Ref. 15 to investigate light scattering, and the results of that paper agree with the observations made above (the activity of the Goldstone mode is neglected and the branches genetically unrelated to the soft branch are not considered in Ref. 15). The frequency $\omega(K_0)$ figuring above is referred to in Ref. 15 as the phason frequency associated with the wave vector $2k_0$, but it is not mentioned that it belongs to another branch (the sense in this designation is evident from Fig. 1, especially if $k$ is measured from the point $A'$). The above-discussed splitting of the branch at $\mathbf{k}=\mathbf{K}_0$ is in fact obtained in Ref. 15, since, besides the phason frequency, an amplitudon frequency is found there to appear at the same wave vector $2k_0$ (the points $C$ and $C'$ in Fig. 1).

In the case considered in Ref. 15 the normal coordinate corresponding to the branch 3 and the coordinate pertaining to the soft branch transform according to different irreducible representations, whence it follows that the quantity $QQ^*$ (see above) is not a full invariant. Therefore, the frequencies in question should, in accordance with the foregoing, be observed in polarization-nonpreserving light scattering, as was likewise found in Ref. 15. We note that the estimates given in Ref. 15 show that the modes corresponding to these frequencies may be either overdamped or underdamped; also given in that paper are formulas for the corresponding scattered-light intensity.

Let us now consider the case in which the end of the vector $\mathbf{K}_0$ again lies near the Brillouin-zone boundary, but no degeneracy exists at the zone boundary. Since there are then no branches genetically related to the soft branch, the new spectral lines with intensities $\sim\rho_e^2$ will correspond to only high-lying branches whose frequencies depend weakly on temperature. As pointed out above, the vibrations with normal coordinates $Q_n(n\mathbf{K}_0)$ can also manifest themselves in light scattering. In the case of the normal coordinate $Q_2(2K_0)$ the spectral-line intensity is proportional to $\rho_e^4$, and the frequency $\omega(2K_0)$ of the soft branch is not low and depends weakly on temperature, since the point $2K_0=b_1-2k_0$ lies in the neighborhood of the center of the Brillouin zone. The most intense line, which corresponds to a low frequency, and depends fairly strongly on temperature, appears because the point $3K_0=3b_1/2-3k_0$, when referred to the first zone, lies near $K_0$; but the intensity of this line is $\sim\rho_e^6$ (in more exact estimates we should, of course, take into account the fact that the low frequency gives a small term—the square of the frequency—in the denominator, and this increases the intensity [cf. Ref. 15)]. Naturally, all the lines discussed here occur in the case in which the soft mode is degenerate at the zone boundary.

If the lattice constant of the low-temperature commensurate phase is three times that of the symmetric phase, then the quantity $K_0$ is close to $b_1/3$, i.e., $K_0=b_1/3-k_0$. Now even the frequency $\omega(2K_0)$ will be low, since, modulo a reciprocal-lattice vector, the difference between $2K_0$ and $-K_0$ is equal to $-3k_0$. The intensity of the corresponding line will be proportional to $\rho_e^4$. This case is considered in Ref. 14. Using the foregoing arguments, we can easily determine the other low frequencies that manifest themselves in the scattering spectrum of the incommensurate phase; but the intensities of the lines will be even lower than the intensities in the cases considered. As to the formulas for the spectral distribution of the intensities of the generated non-Goldstone lines, they can be derived in standard fashion; they have the usual form, and the specific nature of the incommensurate phase in no way manifests itself in them. This latter fact can be seen from the particular case of the formulas obtained in Ref. 15.

The same normal modes that are active in light scattering should manifest themselves also in the dispersion of the elastic susceptibilities, since the tensor $u_{ik}$ possesses the same transformation properties as $\varepsilon_{ik}$. Naturally, such dispersion can be observed by acoustic methods only if the frequencies of the corresponding vibrations are sufficiently low. In the light of what has been said in the present section, we have reason to believe that such frequencies exist in the incommensurable phase.

In order to ascertain the non-Goldstone-excitation-related characteristics of the dielectric dispersion in the incommensurate phase, we must determine which of the translation invariants $Q^nQ_n^*$ transform like the components of the vector $\mathbf{P}$. Here also the number of new lines that appear in the spectrum is relatively small, since the oscillator strength is proportional to $\rho_e^{2n}$. Let us note that the contribution of the soft branch to the dielectric dispersion is analyzed in Ref. 14.

## 3. FOUR-COMPONENT ORDER PARAMETER

Let us now discuss those new features which appear in the considered problems when the number of the order-parameter components is greater than two. As far as we know, except for some assertions, often of an erroneous nature, in some experimental papers, the light-scattering and susceptibility anomalies occurring in the incommensurate phase in this case have not been discussed in the literature at all. Let us perform the analysis for the particular case of a four-component order parameter, when the star of the physically irreducible representation contains four nonequivalent vectors: $\mathbf{K}_1, -\mathbf{K}_1, \mathbf{K}_2, -\mathbf{K}_2$. As the order-parameter components, we can take the corresponding normal coordinates $Q_1(\mathbf{K}_1)$, $Q_1^*$, $Q_2(\mathbf{K}_2)$, and $Q_2^*$.

If the order parameter is a two-component one, then the point symmetry of the incommensurable phase is always the same as that of the symmetric phase. This is due to the fact that in the present case the only translation invariants that depend on the order parameter, but do not contain its derivatives are the quantity $QQ^*=|Q|^2$ and its powers, which are also full invariants. Therefore, no tensor quantities forbidden in the symmetric phase arise in the transition into the incommensurate phase, when $Q$ acquires a spontaneous value, i.e., the point symmetry does not change (see also Ref. 29).

A different situation arises when the number of order-parameter components is greater than two. In the case of four components, besides $|Q_1|^2+|Q_2|^2$, the quantity $|Q_1|^2-|Q_2|^2$, for example, will also be a translation invariant. This invariant cannot be a full one, since there is only one second-order full invariant: $|Q_1|^2+|Q_2|^2$. Therefore, if the phase with $|Q_{1e}|^2 \neq |Q_{2e}|^2$ corresponds to a state of thermodynamic equilibrium, then the spontaneous tensor quantity $|Q_{1e}|^2 - |Q_{2e}|^2$, which is forbidden by the symmetry of the high-temperature phase, arises, i.e., the point symmetry changes. As will be shown below, such a symmetry change can manifest itself in, for example, light scattering.

In the case of the four-component order parameter, the soft mode in the symmetric phase is formally fourfold degenerate. This degeneracy is partially or completely lifted below the phase-transition point, i.e., there can arise below this point up to four different modes, although, as we shall see below, not every one of these modes has to contribute to light scattering or the susceptibility dispersion. When the order parameter possesses four components, there can be realized phases with two-dimensional incommensurability and, consequently, with two Goldstone modes,[30,31] and we shall show below that both are active in scattering.

We must, in considering incommensurate phase transitions with a four-component order parameter, distinguish two cases.[12] In the first, some combination of the vectors $K_1$ and $K_2$ coincides with one of the reciprocal-lattice vectors, and incommensurability is possible only along one direction. In the second case no combination of the vectors $K_1$ and $K_2$ yields a reciprocal-lattice vector, and two-dimensional incommensurability is possible here. Let us, for concreteness, discuss the first case, using as an example barium manganese fluoride $BaMnF_4$, in which this case is realized,[32] and which has been the subject of a relatively large number of experimental investigations. Let us limit ourselves to light scattering by the order-parameter oscillations. It is not difficult to take the other oscillations into account in the investigation, and also to analyze the susceptibility anomalies in much the same way as this was done for the two-component order parameter.

If we set

$$Q_1=\rho_1\exp(i\varphi_1),\quad Q_2=\rho_2\exp(i\varphi_2),$$

then the thermodynamic potential density has, after we have allowed for the transformation properties of the order parameter in the $BaMnF_4$ case,[32,33] the form

$$\Phi=\frac{\alpha}{2}(\rho_1^2+\rho_2^2)+\frac{\beta_1}{4}(\rho_1^4+\rho_2^4)+\frac{\beta_2}{2}\rho_1^2\rho_2^2+\frac{\beta_3}{2}\rho_1^2\rho_2^2\cos 2(\varphi_1-\varphi_2)+\frac{1}{2}\sum_{i=1}^{3}\delta_i\left[\left(\frac{\partial\rho_1}{\partial x_i}\right)^2+\rho_1^2\left(\frac{\partial\varphi_1}{\partial x_i}\right)^2+\left(\frac{\partial\rho_2}{\partial x_i}\right)^2+\rho_2^2\left(\frac{\partial\varphi_2}{\partial x_i}\right)^2\right]+\frac{\delta_4}{2}\left(\frac{\partial\rho_1}{\partial y}\frac{\partial\rho_1}{\partial z}+\rho_1^2\frac{\partial\varphi_1}{\partial y}\frac{\partial\varphi_1}{\partial z}-\frac{\partial\rho_2}{\partial y}\frac{\partial\rho_2}{\partial z}-\rho_2^2\frac{\partial\varphi_2}{\partial y}\frac{\partial\varphi_2}{\partial z}\right), \tag{14}$$

where of the invariants that are quadratic in the derivatives we have written out only those which are essential to our problem. In $BaMnF_4$ the incommensurability is observed along the polar axis $x$, and the Lifshitz invariant has the form

$$\frac{\rho_1^2\partial\varphi_1}{\partial x}+\frac{\rho_2^2\partial\varphi_2}{\partial x}.$$

The diagonal elements of the tensor $\varepsilon_{ik}$ are linearly coupled to $\rho_i$ and $\varphi_i$ combinations of the type of those which enter into (14) and to the Lifshitz invariant; the element $\varepsilon_{yz}$ is linearly coupled to

$$\rho_1^2-\rho_2^2,\quad \rho_1^2\rho_2^2\sin 2(\varphi_1-\varphi_2),\quad \rho_1^2\frac{\partial\varphi_1}{\partial x}-\rho_2^2\frac{\partial\varphi_2}{\partial x};$$

and the elements $\varepsilon_{xy}$ and $\varepsilon_{xz}$ are coupled respectively to

$$a_1\left(\rho_1^2\frac{\partial\varphi_1}{\partial y}+\rho_2^2\frac{\partial\varphi_2}{\partial y}\right)+a_2\left(\rho_1^2\frac{\partial\varphi_1}{\partial z}-\rho_2^2\frac{\partial\varphi_2}{\partial z}\right),$$

$$a_3\left(\rho_1^2\frac{\partial\varphi_1}{\partial z}+\rho_2^2\frac{\partial\varphi_2}{\partial z}\right)+a_4\left(\rho_1^2\frac{\partial\varphi_1}{\partial y}-\rho_2^2\frac{\partial\varphi_2}{\partial y}\right).$$

Depending on the coefficients in $\Phi$ [Eq. (14)], there can be two equilibrium solutions[3)] corresponding to different incommensurate phases[32]:

$$1)\ \rho_1=\rho_{1e}\neq 0,\ \rho_2=0\ (\text{or}\ \rho_1=0,\ \rho_2\neq 0),\quad 0<\beta_1<\beta_2-|\beta_3|; \tag{15}$$

$$2)\ \rho_1=\rho_2=\rho_e,\ \sin 2(\varphi_1-\varphi_2)=0,\quad \beta_1>|\beta_2-|\beta_3||. \tag{16}$$

In the case of (15) the point symmetry of the incommensurate phase is different from that of the symmetric phase ($\varepsilon_{yz}^{(e)}\neq 0$); in the case of (16) the point symmetries of the two phases are the same ($\varepsilon_{yz}^{(e)}=0$).

In both cases four soft branches arise below the transition point.[32] In the case of (15) these branches are connected with the oscillations associated with $\rho_1$, $\varphi_1$, and the two linear combinations of $Q_2$ and $Q_2^*$. Corresponding to the Goldstone mode are the $\varphi_1$ "vibrations" with the dispersion law

$$\mu\omega^2+i\nu\omega=\delta_1q_x^2+\delta_2q_y^2+\delta_3q_z^2+\delta_4q_yq_z,$$

for the complex frequency, vibrations which are clearly overdamped (see the first section). The $Q_2$ and $Q_2^*$ vibrations do not manifest themselves in either first-order light scattering or susceptibility dispersions, since any translation invariant is quadratic in these quantities, and the spontaneous value of $Q_2$ is equal to zero. The amplitudon contributes not only to the fluctuations of the diagonal elements of $\varepsilon_{ik}$, as in the case of the two-component order parameter, but also to the $\varepsilon_{yz}$ fluctuations.

The $\varphi_1$ fluctuations will manifest themselves in light scattering owing to the presence of the quantities $\rho_{1e}^2\partial\Delta\varphi_1/\partial x_i$, which make contributions to the fluctuations of all the elements of $\varepsilon_{ik}$. Moreover, the character of the scattering by the phasons is the same as in the case of the two-component order parameter, except that the angular dependence is different, since for the fluctuations of the Fourier component $\varphi_1(\mathbf{q})$ we have

$$\langle|\varphi_1(\mathbf{q})|^2\rangle=k_BT/V\rho_{1e}^2(\delta_1q_x^2+\delta_2q_y^2+\delta_3q_z^2+\delta_4q_yq_z). \tag{17}$$

This quantity changes when the sign of $q_y$ or $q_z$ is changed, i.e., the directivity pattern for the scattering does not, in contrast to the case of the two-component order parameter, possess the symmetry of the high-temperature phase. In the final analysis, this is due to the lowering of the point symmetry in the transition into the incommensurate phase. Naturally, this effect is weaker in a multidomain sample. Let us note that now the mean square fluctuations $\langle|\Delta\varepsilon_{ik}(\mathbf{q})|^2\rangle$ may contain in the numerator not only the squares of the individual components of the vector $\mathbf{q}$, as in (5), but also combinations of the form $(aq_y + bq_z)^2$.

In the case (16) the Goldstone mode corresponds to the $\varphi^{(+)} = \varphi_1 + \varphi_2$ vibrations with the dispersion law

$$\mu\omega^2+i\nu\omega=\delta_1 q_x^2+\delta_2 q_y^2+\delta_3 q_z^2,$$

for the complex frequency; three other soft modes are connected with the $\varphi^{(-)} = \varphi_1 - \varphi_2$, $\rho^{(+)} = \rho_1 + \rho_2$, and $\rho^{(-)} = \rho_1 - \rho_2$ vibrations.[32] The Goldstone mode manifests itself in the fluctuations of all the elements of $\varepsilon_{ik}$, except the element $\varepsilon_{yz}$, since the fluctuations of these elements are, when allowance is made for (16), coupled to the $\rho_e^2\partial\Delta\varphi^{(+)}/\partial x_i$. The mean square fluctuation of the Fourier transform of $\Delta\varphi^{(+)}$ here is, for small values of $|\mathbf{q}|$, given by a formula coinciding up to a factor of 2 with (4), and does not, in contrast to (17), change when the sign of $q_y$ or $q_z$ is changed. The $\varepsilon_{ik}$ fluctuations coupled to $\Delta\varphi^{(+)}$ are given by formulas of the type (5).

For the non-Goldstone excitations the specific nature of the incommensurate phase practically does not manifest itself. Cn account of (16), the $\varphi^{(-)}$-phase oscillation make in the limit $\mathbf{q}\to 0$ a contribution only to the $\varepsilon_{yz}$ fluctuations, with

$$\langle|\Delta\varepsilon_{yz}(\mathbf{q}\approx 0)|^2\rangle\sim\rho_e^8\langle|\Delta\varphi^{(-)}(\mathbf{q}\approx 0)|^2\rangle\sim\rho_e^4\sim(T_i-T)^2.$$

The diagonal elements of $\varepsilon_{ik}$ are coupled to $\rho_1^2 + \rho_2^2$, and the $\rho^{(+)}$ oscillations manifest themselves in their fluctuations; the $\rho^{(-)}$ vibrations, on the other hand, make a contribution to the $\varepsilon_{yz}$ fluctuations, since $\varepsilon_{yz}$ depends on $\rho_1^2 - \rho_2^2$. As is usually the case for soft modes,[16] the intensity of light scattered by the $\rho^{(+)}$ or $\rho^{(-)}$ oscillations undergoes a jump at the critical point of the transition into the incommensurate phase.

Proceeding to the analysis of the case in which no combination of the vectors $\mathbf{K}_1$ and $\mathbf{K}_2$ forms a reciprocal-lattice vector, let us, for convenience of comparison with the preceding case, perform the analysis for crystals possessing the same space group in the symmetric phase ($A2_1am-C_{2v}^{12}$) as $BaMnF_4$. Although we do not know any experimental examples of this incommensurate phase transition in crystals with precisely the indicated space group, such a transition is observed[34] in barium sodium niobate $Ba_2NaNb_5O_{15}$, which belongs to another system. A special analysis shows that the results presented below are qualitatively valid for crystals of the type $Ba_2NaNb_5O_{15}$ as well.

Let us take as the star of the irreducible representation a star of the general form in the $k_yk_z$ plane inside the Brillouin zone. If we retain only the terms of importance to us, then the thermodynamic potential will also have the form (14), except that the term with the coefficient $\beta_3$ will be absent; the analogous terms in the other translation invariants will also vanish. In the present case there are two Lifshitz invariants:

$$\rho_1^2\frac{\partial\varphi_1}{\partial y}+\rho_2^2\frac{\partial\varphi_2}{\partial y},\quad \rho_1^2\frac{\partial\varphi_1}{\partial z}-\rho_2^2\frac{\partial\varphi_2}{\partial z},$$

which reflects the absence of a symmetry-related extremum of the dispersion surface along both the $k_y$ and the $k_z$ axes at an arbitrary point in the $k_yk_z$ plane. The dependence of $\varepsilon_{ik}$ on $\varphi_1$ and $\varphi_2$ is given by the Lifshitz invariants for the diagonal components and by analogous quantities, but with derivatives with respect to other coordinates, for the off-diagonal components.

Depending on the values of the coefficients in $\Phi$, one of two incommensurate phases is realized (we shall not discuss the third solution, which arises when the higher-order invariants are taken into account):

$$1)\ \rho_1=\rho_{1e}\neq 0,\quad \rho_2=0\quad (\text{or } \rho_1=0,\ \rho_2\neq 0),\quad 0<\beta_1<\beta_2; \tag{18}$$

$$2)\ \rho_1=\rho_2=\rho_e,\quad \beta_1>|\beta_2|. \tag{19}$$

In the case of (18) there arises a one-dimensional incommensurability (the wave with vector $\mathbf{K}_1$ or $\mathbf{K}_2$ is frozen), and the point symmetry decreases in the transition ($\varepsilon_{yz}^{(e)} \sim (\rho_1^2 - \rho_2^2) \neq 0$); in the case (19) the incommensurability is two-dimensional (being characterized by both vectors: $\mathbf{K}_1$ and $\mathbf{K}_2$), and the point symmetry remains unchanged in the transition ($\varepsilon_{yz}^{(e)} = 0$).

The laws governing light scattering in the case of the phase corresponding to (18) are similar to those discussed above for the solution (15), but instead of two inactive soft modes, we have here a single doubly degenerate one.

Some new features appear in the case of the two-dimensional incommensurability. Naturally, here there are two Goldstone modes corresponding to the $\varphi_1$- and $\varphi_2$-phase oscillations, the dispersion law for the complex frequencies having the form

$$\mu\omega^2+i\nu\omega=\delta_1 q_x^2+\delta_2 q_y^2+\delta_3 q_z^2\pm\delta_4 q_y q_z.$$

The two modes make a contribution to the fluctuations of all the elements of $\varepsilon_{ik}$. The angular dependence of the intensity of the scattered light connected with this contribution will be quite complicated here, which can be seen from the particular case of the mean square fluctuations of the diagonal elements of the $\varepsilon_{ik}$ tensor:

$$\langle|\Delta\varepsilon_{ii}(\mathbf{q})|^2\rangle=\frac{2k_BT\rho_e^2[(a^2q_y^2+b^2q_z^2)(\delta_1q_x^2+\delta_2q_y^2+\delta_3q_z^2)-2ab\delta_4q_y^2q_z^2]}{V[(\delta_1q_x^2+\delta_2q_y^2+\delta_3q_z^2)^2-\delta_4^2q_y^2q_z^2]}, \tag{20}$$

where $a$ and $b$ are coefficients in the expansion of $\varepsilon_{ii}$ in powers of the order parameter. In spite of the fact that the $\varphi_1$ and $\varphi_2$ fluctuations are not invariant under a change of the sign of $q_y$ or $q_z$ [the quantity $\langle|\varphi_1(\mathbf{q})|^2\rangle$ is also given by an expression of the type (17), while $\langle|\varphi_2(\mathbf{q})|^2\rangle$ is given by the analogous expression with the sign in front of $\delta_4$ changed], the contribution of the two Goldstone modes to scattering results in the invariance of (20) under a change of the sign of $q_y$ or $q_z$, which also accounts for the complicated form of this expression.

## 4. EXPERIMENTAL SEARCH FOR THE PHASON

In our opinion, it cannot at present be said that the Goldstone phason has been experimentally detected. In a number of papers on light scattering and dielectric dispersion[7,5] where the observation of the phason is claimed, the authors actually meant, as can be concluded from the texts of their papers, non-Goldstone low-frequency modes (the terminology ambiguity noted in the first footnote shows up here).

Lyons *et al.*[2,3] have reported the observation in $BaMnF_4$ of a central peak whose width is proportional to the square of the vector **q**, as it should be in the case of scattering by the Goldstone phason (see above). They detected this peak in experimental investigations of polarization-preserving light scattering, i.e., light scattering connected with the fluctuations of the diagonal elements of $\varepsilon_{ik}$. Above we asserted that in the case of $BaMnF_4$ the diagonal elements of $\varepsilon_{ik}$ depend on $\varphi_1$ and $\varphi_2$ because of the Lifshitz invariant

$$\rho_1^2\frac{\partial\varphi_1}{\partial x}+\rho_2^2\frac{\partial\varphi_2}{\partial x}$$

and, consequently, their mean square fluctuations are proportional to $q_x^2$. But Lyons *et al.*[2,3] report that the central peak under discussion disappears precisely when q is oriented along the $x$ axis, and, moreover, that its maximum width $\Delta\Omega$ is roughly $10^{10}$ Hz, which is two-to-three orders of magnitude greater than the width (9). Thus, their claim that they have detected scattering by phasons is clearly not supported by theory. Let us also note that Lockwood *et al.*[11] have expressed doubt about the correctness of the interpretation of the experimental data in Refs. 2 and 3.

In their turn, Lockwood *et al.* report in Ref. 11 the detection of light scattering by phasons, to which they attribute the contribution to the Mandel'shtam–Brillouin components. But, as asserted above, the Goldstone phason should contribute to the cental peak.

It is already evident from the foregoing that the detection of the manifestations of the Goldstone phason is not a simple experimental problem. First, the width of the phason central peak is quite small. Secondly, this peak should have a relatively low intensity. As has already been noted, the intensity of the phason line is proportional to $\rho_e^2$, i.e., it vanishes at the critical point ($T = T_i$) of the transition into the incommensurate phase.

In conclusion, the authors express their gratitude to A.A. Sobyanin, J. Petzelt, and R.A. Suris for a discussion of the results of the present paper and for useful comments.

1) Let us note that the terminology in this field is not standardized. Sometimes by the word "phasons" is meant not only the Goldstone excitations, i.e., the excitations corresponding to a small neighborhood of the point $A$ (see Fig. 1), but also the higher excitations of the branch 1, which are called phasons with wave vectors $2k_0$, $3k_0$, etc. In the present section we consider only the Goldstone excitations; the activity of the higher-lying excitations will be discussed below.

2) Note that such a temperature dependence obtains only in the region of applicability of the Landau theory, in which $\rho_e^2 \sim (T_i - T)$; in the scaling region the quantity $\rho_e^2$ is proportional to $(T_i - T)^{2\beta}$, where $\beta$ is one of the critical exponents (see, for example, Ref. 16 and the literature cited there). Below, for brevity, we limit ourselves everywhere to the region of applicability of the Landau theory; the generalization of the corresponding formulas to the scaling region is trivial, since the important temperature dependences in them are determined only by the quantity $\rho_e$.

3) Allowance for higher-order invariance shows that a third solution is also possible.[33] ($0\neq\rho_1\neq\rho_2\neq 0$); it will not be analyzed here. Nor shall we identify the case actually realized in $BaMnF_4$.


1. P. A. Fleury, S. Chiang, and K. B. Lyons, Solid State Commun. **31**, 279 (1979).
2. K. B. Lyons and H. J. Guggenheim, Solid State Commun. **31**, 285 (1979).
3. K. B. Lyons, T. J. Negran, and H. J. Guggenheim, J. Phys. C **13**, L415 (1980).
4. H.-G. Unruh, W. Eller, and G. Kirf, Phys. Status Solidi A **55**, 173 (1979).
5. A. A. Volkov, Y. Ishibashi, G. V. Kozlov, and J. Petzelt, Fiz. Tverd. Tela (Leningrad) **22**, 1424 (1980) [Sov. Phys. Solid State **22**, 831 (1980)].
6. M. Takashige and T. Nakamura, Ferroelectrics **24**, 143 (1980).
7. K. Inoue, S. Koiwai, and Y. Ishibashi, J. Phys. Soc. Jpn. **48**, 1785 (1980).
8. V. Winterfeldt and G. Schaack, Z. Phys. B **36**, 303 (1980).
9. E. Francke, M. Le Postollec, J. P. Mathieu, and H. Poulet, Solid State Commun. **35**, 183 (1980).
10. D. I. Siapkas, Ferroelectrics **29**, 29 (1980).
11. D. J. Lockwood, A. F. Murray, and N. L. Rowell, J. Phys. C **14**, 753 (1981).
12. R. A. Cowley, Adv. Phys. **29**, 1 (1980).
13. M. Wada, H. Uwe, A. Sawada, Y. Ishibashi, Y. Takagi, and T. Sakudo, J. Phys. Soc. Jpn. **43**, 544 (1977).
14. V. Dvořák and J. Petzelt, J. Phys. C **11**, 4827 (1978).
15. V. A. Golovko and A. P. Levanyuk, Pis'ma Zh. Eksp. Teor. Fiz. **32**, 104 (1980) [JETP Lett. **32**, 93 (1980)].
16. V. L. Ginzburg, A. P. Levanyuk, and A. A. Sobyanin, Usp. Fiz. Nauk **130**, 615 (1980); Phys. Rep. **57**, 151 (1980).
17. A. Janner and T. Janssen, Acta Crystallogr. Sect. A **36**, 399, 408 (1980).
18. I. E. Dzyaloshinskiĭ, Zh. Eksp. Teor. Fiz. **46**, 1420 (1964); **47**, 336, 992 (1964) [Sov. Phys. JETP **19**, 960 (1964); **20**, 223, 665 (1965)].
19. A. P. Levanyuk and D. G. Sannikov, Fiz. Tverd. Tela (Leningrad) **18**, 423 (1976) [Sov. Phys. Solid State **18**, 245 (1976)].
20. M. Iizumi, J. D. Axe, G. Shirane, and K. Shimaoka, Phys. Rev. B **15**, 4392 (1977).
21. V. Dvořák and Y. Ishibashi, J. Phys. Soc. Jpn. **45**, 775 (1978).
22. Y. Ishibashi, Ferroelectrics **24**, 119 (1980).
23. M. Iizumi and K. Gesi, J. Phys. Soc. Jpn. **49**, Suppl. B, 72 (1980).
24. A. W. Overhauser, Phys. Rev. B **3**, 3173 (1971).
25. P. A. Lee, T. M. Rice, and P. W. Anderson, Solid State Commun. **14**, 703 (1974).
26. A. D. Bruce and R. A. Cowley, J. Phys. C **11**, 3609 (1978).
27. L. D. Landau and E. M. Lifshitz, Élektrodinamika sploshnykh sred (Electrodynamics of Continuous Media), Fizmatgiz, Moscow, 1959 (Eng. Transl., Pergamon Press, Oxford, 1960).
28. I. Hatta, M. Hanami, and K. Hamano, J. Phys. Soc. Jpn. **48**, 160 (1980).
29. V. A. Golovko and A. P. Levanyuk, Zh. Eksp. Teor. Fiz. **77**, 1556 (1979) [Sov. Phys. JETP **50**, 780 (1979)].
30. T. M. Rice, Solid State Commun. **17**, 1055 (1975).
31. W. L. McMillan, Phys. Rev. B **12**, 1187 (1975).
32. D. E. Cox, S. M. Shapiro, R. A. Cowley, M. Eibschütz, and H. J. Guggenheim, Phys. Rev. B **19**, 5754 (1979).
33. V. Dvořák and J. Fousek, Phys. Status Solidi A **61**, 99 (1980).
34. J. Schneck and F. Denoyer, Phys. Rev. B **23**, 383 (1981).


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