# Spin-lattice relaxation-rate anomaly at structural phase transitions

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The theory of spin-lattice relaxation (SLR)-rate anomaly at structural phase transitions proposed about 30 years ago is reconsidered taking into account that knowledge about the relevant lattice response functions has changed considerably. We use both the results of previous authors and perform original calculations of the response functions when it is necessary. We consider displacive systems and use the perturbation theory to treat the lattice anharmonicities in a broad temperature region whenever possible. Some comments about the order-disorder systems are made as well. The possibility of linear coupling of the order parameter and the resonance frequency is always assumed. It is found that in the symmetrical phase the anomaly is due to the one-phonon processes, the anomalous part being proportional to either $(T-T_c)^{-1}$ or $(T-T_c)^{-1/2}$ depending on some condition on the soft-mode dispersion. In both cases the value of the SLR rate at the boundary of applicabity of the theory (close to the phase transition) is estimated to be $10^2-10^3$ times more than the typical value of the SLR rate in an ideal crystal. An essential specific feature of the nonsymmetrical phase is appearance of third-order anharmonicities that are well known to lead to a low-frequency dispersion of the order-parameter damping constant. We have found that this constant exhibits, in addition, a strong wave-vector dispersion, so that the damping constant determing the SLR rate is quite different from that at zero wave vector. In the case of two-component order parameter the damping constant for the component with nonzero equilibrium value is different from that for the other component, the difference is of the same order of magnitude as the damping constants themselves. In the case of the incommensurate phase a part of the mentioned third-order anharmonicity is responsible for longitudinal-transversal interaction that is well known to influence the static longitudinal response function. We calculate as well the dynamic response function to find that for the SLR calculations the imaginary part is of main importance. Due to this interaction the longitudinal SLR rate acquires a dependence on the Larmor frequency. This dependence is however, fairly weak: a logarithmic one. The implications of the obtained results for interpretation of the experimental data on SLR in incommensurate phase are discussed as well. [S0163-1829(97)08545-7]

## I. INTRODUCTION

The study of spin-lattice relaxation (SLR) rate ($T_1^{-1}$) anomalies near structural phase transitions began 30 years ago[1] with observation of a $T_1^{-1}$ maximum at the ferroelectric phase transition in NMR experiments on $NaNO_2$. Similar anomalies have been observed since then[2] in $KH_2PO_4$ and at many other phase transitions both in NMR and NQR experiments (for a recent review, see Ref. 3). The theory used to interpret these anomalies is basically the same that was developed about the same time,[1,2,4,5] and that is presented in the most exhaustive form in Ref. 5 which we refer to below.

The conclusions of SLR theory depend essentially on the results or assumptions about the relevant lattice-dynamic response functions, in our case about the dynamic response function of the order parameter. It should be emphasized that the response functions probed in a SLR experiment are fairly special: for low frequencies and, in principle, any wave vectors, such functions are not probed directly, either in scattering experiments or in low-frequency macroscopic ones (e.g., in the measurements of dielectric losses, for ferroelectrics). So it was natural and inevitable that in the first theory of the spin-lattice relaxation anomalies some assumptions were initially made about the response functions. Specifically, it was assumed that the order-parameter response function is that of an oscillator with damping with all the parameters being frequency independent and only one of them having a temperature and wave-vector dependence: the eigenfrequency of the oscillator (the soft-mode frequency). Neither were the orders of magnitude of the parameters involved discussed: such estimations were in their initial stage at that time. Similar comments can be addressed to an important later development of the theory: its application to incommensurate (IC) phases.[6]

Since then considerable progress has been made in understanding the character of the order-parameter response functions for structural displacive transitions or, rather, for the so-called displacive limit. Let us recall that in the displacive limit it is supposed that the phase transition occurs in a

 

weakly anharmonic crystal[7–12] or, rather in a crystal that can be considered as a weakly anharmonic one in a broad temperature region ($T \ll T_{at}$, $T_{at}$ is the atomic temperature, $T_{at} \sim 10^4 - 10$ K) excluding a fairly narrow vicinity of the phase transition (as long as the phase-transition temperature $T_c \ll T_{at}$). The condition for this vicinity reads[10]

$$\frac{|T-T_c|}{T_c} < \frac{T_c}{T_{at}}, \tag{1}$$

and beyond it one can use a standard perturbation theory to take into account the effects of the anharmonicity. That is why it is possible to use a regular theoretical approach to treat dynamic properties of the displacive systems, beyond the region defined by Eq. (1), and it is in difference with the order-disorder systems where the anharmonicity is always large and should be modeled in some way (kinetic Ising model or something of the kind).

The main qualitative conclusion that can be made as a result of this progress is that the order-parameter response function is far more complicated that it was assumed in Refs. 5,6. For ''ordinary'' (not incommensurate) phase transitions it reduces to the statement that the low-frequency damping coefficient can be quite different from the high-frequency one probed in neutron or light-scattering experiments. For some special cases it was already indicated in Refs. 13,14 but a decisive step was made by Cowley and Coombs[15] who argued that in the nonsymmetrical phase, due to a new third-order anharmonicity, the damping constant acquires strong frequency dispersion at frequencies much lower than the soft-mode one. They proposed a semiphenomenological low-frequency response function with temperature dependence, not only of the soft-mode frequency but of the other parameters as well. Later some of these dependences have been corrected[16] but the qualitative result of Cowley and Coombs remains intact. In addition, the temperature anomaly of the damping constant in the symmetrical phase was predicted long ago.[14] One sees that there are enough reasons to reconsider the theory of Ref. 5. However, this reconsideration cannot only consist of application of the known results for the order-parameter response function to the calculation of the SLR anomaly. The problem is that the known results refer to low frequencies and *small wave vectors* and no study of the wave-vector dependence has been made, to the best of our knowledge. It may be due to the idea that in the scaling region the wave-vector dependences have only one scale: the reciprocal correlation radius ($r_c^{-1}$). But we are interested in temperatures beyond the critical region and the scaling hypothesis is not applicable here. We will see that the wave-vector dependence at a scale which is less than $r_c^{-1}$ proves to be essential and important in the treatment of SLR.

For IC systems the importance of the third-order anharmonicity specific for the nonsymmetrical (IC) phase is well understood. Here one of the aspects of this anharmonicity is interaction between the logitudinal and the transversal order-parameter fluctuations which correspond, in the harmonic approximation, to amplitudon and phason. An IC phase in an ideal crystal is soft: there is no restoring force to the inhomogeneous shift of the IC modulation, i.e., the fluctuation of the phase of IC modulation, the transversal fluctuations, diverge when the fluctuation vector goes to zero. This property is general for degenerated systems including Heisenberg magnetics and nematic liquid crystals[17–21] where the effects of these fluctuations were discussed long ago. The main effect of the third-order anharmonicity is that not only the transversal susceptibility is infinite at $\mathbf{q} \to 0$ ($\mathbf{q}$ is the wave vector) but also the longitudinal one. For structural IC systems the divergence of the longitudinal susceptibility was discussed by Bruce and Cowley in their frequently cited paper.[22] They did not calculate, however, the dynamic longitudinal response function which is necessary to treat the contribution of the longitudinal fluctuations to the SLR rate. Surprisingly, these calculation have not been made, despite the experimentalists repeatedly emphasizing their necessity (see, e.g., Refs. 23,24). To perform these calculations is one of the aims of the present paper.

To calculate the response functions of interest, we shall start from classical equations of motion for the order parameter in the symmetrical phase treating then the effects of some of the anharmonic terms within first-order perturbation theory. A part of the crystal anharmonicity will be taken into account (phenomenologically) in the initial equations of motion by a phenomenological viscosity coefficient which we will estimate according to the results of previous authors. This method allows us to avoid the partial summation of the infinite perturbation theory series that is required when one calculates the low-frequency response function and assumes, as the zero approximation, the response function of the harmonic crystal. As temperature plays the role of a parameter in the equations, we do not take into account, by this method, the part analogous to the thermoelastic losses for longitudinal-acoustic waves but calculate them separately within a standard macroscopic treatment.[25]

The paper is organized as follows. In Sec II we present the formulas which will be used to calculate $T_1^{-1}$. In Sec. III we discuss the case of one-component order parameter. This case proves to be the simplest one. In Sec. IV the case of many-component order parameter is discussed with a special attention to incommensurate systems. In Sec. V we make some comments about order-disorder systems. Finally, Sec. VI contains a brief summary and a discussion of implications of the theoretical results of the paper for interpretation of the experimental data.

## II. SPIN-LATTICE RELAXATION TIME AND THE ORDER-PARAMETER CORRELATION FUNCTIONS

The spin-lattice relaxation is determined by the probabilities of transitions between the states of the Zeeman Hamiltonian due to perturbations caused by the lattice fluctuations.[26] In general, the return of the nuclear magnetization back to its thermal equilibrium value cannot be described by one exponent but in any case the temporal equations contain probabilities that are proportional to some combinations of the spectral densities of local fluctuations of the electric-field-gradient tensor. These spectral densities are proportional to the spectral densities of fluctuations of the lattice variables among which we will single out those corresponding to the order parameter. It is, in effect, the local spectral density of the order-parameter fluctuations that will be discussed in the present paper. To demonstrate its specific features is enough to consider the simplest case where one

can write the perturbation Hamiltonian as a product $AF(t)$, where $A$ is an operator acting on the spin variables and $F(t)$ is a function depending on the lattice variables. The generalization is straightforward. In this simplest case[26]

$$T_1^{-1}=gJ(\Omega_L), \tag{2}$$

where $g$ is a constant, $\Omega_L$ is the Larmor frequency,

$$J(\omega)=\int_{-\infty}^{\infty}\langle F(t)F(t+\tau)\rangle e^{-i\omega\tau}d\tau, \tag{3}$$

and $\langle\ \rangle$ designates the statistical average. Being interested in the phase-transition anomaly we present $F$ as

$$F=F_0+a_1\eta+a_2\eta^2+\cdots, \tag{4}$$

where $\eta$ is the order parameter which, for the moment, we assume to have only one (real) component.

Using Eqs. (3) and (4) one obtains

$$\begin{aligned}J(\Omega_L)=a_1^2\int D^3\mathbf{q}\langle|\eta(\mathbf{q},\Omega_L)|^2\rangle+a_2^2\int D^3\mathbf{q}\langle|\eta^2(\mathbf{q},\Omega_L)|^2\rangle\\ +a_1a_2\int D^3\mathbf{q}[\langle\eta(\mathbf{q},\Omega_L)\eta^2(-\mathbf{q},-\Omega_L)\rangle\\ +\langle\eta(\mathbf{q},-\Omega_L)\eta^2(-\mathbf{q},\Omega_L)\rangle]\\ \equiv J_1+J_2+J_{12},\end{aligned} \tag{5}$$

where $\eta(\mathbf{q},\Omega_L)$, $\eta^2(\mathbf{q},\Omega_L)$ are time and space Fourier transforms of $\eta(\mathbf{r},t)$ and $\eta^2(\mathbf{r},t)$, $D^3\mathbf{q}=d^3\mathbf{q}/(2\pi)^3$, and $\langle\ \rangle$ means statistical average. The first term in the right-hand side (rhs) of Eq. (5) corresponds to the one-phonon process, the second to the two-phonon (Raman) process, and the third to what can be called the ''mixed'' contribution. This contribution is absent, of course, for a harmonic lattice but we will be interested in anharmonic effects as well and there is no reason to neglect this contribution *ad hoc*.

The correlation functions are related to the dynamic response function for the order parameter determined by the formula:

$$\eta(\mathbf{q},\Omega_L)=\chi(\mathbf{q},\Omega_L)h(\mathbf{q},\Omega_L), \tag{6}$$

where $h$ is the generalized force conjugated to $\eta$. One has[27]

$$\langle|\eta(\mathbf{q},\Omega_L)|^2\rangle=\frac{T}{\pi\Omega_L}\ \mathrm{Im}\ \chi(\mathbf{q},\Omega_L). \tag{7}$$

The dynamic response function of the order parameter can be calculated from the equation of motion for the order parameter which is obtainable from the continuous medium effective potential energy which is the Landau free energy:

$$\Phi(\eta)=\frac{A}{2}\eta^2+\frac{B}{4}\eta^4+\frac{D}{2}(\nabla\eta)^2 \tag{8}$$

with addition of the inertial and the viscosity terms:

$$m\ddot{\eta}+\gamma\dot{\eta}+A\eta-D\Delta\eta+B\eta^3=h. \tag{9}$$

This equation is supposed to be obtained as a result of integration over all the degrees of freedom of the system but are those corresponding to the long-wave order-parameter fluctuations ($kd\ll 1$, $k$ is the wave vector, $d$ is the interatomic distance). As usual, the anharmonic interaction of the order parameter and other degrees of freedom is taken into account by the temperature dependence of the coefficient $A=A'(T-T_c)$ and by the viscosity term. In fact, even for a weakly anharmonic crystal, the exact equation is more complicated than Eq. (9). In particular, the coefficient $\gamma$ proves to depend essentially on the frequency (see below). However, within this paper we are interested in the response function for low frequency ($\Omega_L$ is lower than any characteristic frequency of the system) and the wave vectors comparable with the order-parameter correlation radius which, for the displacive limit systems, is much larger than the interatomic distance even far from the phase transition.[10] Under such conditions the coefficient $\gamma$ in Eq. (9) can be considered as a constant but, in general, quite different from that obtainable from study of the form of the soft-mode line in neutron or Raman-scattering spectra (see below). Also the anharmonic coefficient $B$ can be considered as independent of the wave vectors.

Our study will be centered on taking into account consistently the effects of the last term in the lhs of Eq. (9) in the response function. Explicitly we shall make it only for the cases where the results needed cannot be found in literature referring otherwise to other authors. We will see below that despite that a good part of the effects of the last term in the lhs of Eq. (9) has been discussed already, a part of the study that is important for the theory of the SLR anomaly was not made.

As it has been already mentioned in the displacive limit the calculations can be made within the perturbation theory if $|T-T_c|$ is not too small. In terms of the coefficients in Eqs. (8),(9) this condition reads

$$\xi=\frac{BT}{D^{3/2}|A|^{1/2}}\ll 1. \tag{10}$$

This is the condition of applicability of the Landau theory as well.[27] Taking into account the estimations of the Landau coefficients in the displacive limit due to Vaks:[10] $A'\sim d^{-5}$, $B\sim T_{\mathrm{at}}d^{-7}$, $D\sim T_{\mathrm{at}}d^{-3}$, where $T_{\mathrm{at}}\sim 10^4$–$10^5$ K and the ''atomic'' (maximum) value of the order parameter is estimated as $d$, one comes to Eq. (1).

## III. ONE-COMPONENT ORDER PARAMETER

### A. Symmetrical phase

To the zero approximation the response function is

$$\chi_0(\mathbf{k},\omega)=\frac{1}{-m\omega^2+i\omega\gamma+A+Dk^2}. \tag{10a}$$

Let us discuss the order of magnitude of the order-parameter viscosity constant $\gamma$ or, rather, the soft-mode damping constant $\Gamma=\gamma/m$. The calculations of this constant have been made by many authors, for recent reviews see, Ref. 28. It is convenient to discriminate the contributions due to the coupling of the soft mode to other ''hard'' optical modes ($\Gamma_h$), due to the coupling to acoustic modes ($\Gamma_a$) and due to the anharmonicity within the soft-mode branch, i.e, the effect of the last term in the lhs of Eq. (9), ($\Gamma_c$):

$$\Gamma=\Gamma_h+\Gamma_a+\Gamma_c. \tag{11}$$

It was estimated by Stolen and Dransfeld[29] that at $T>T_D$ ($T_D$ is the Debye temperature)

$$\Gamma_h\sim\Omega_D\left(\frac{T}{T_{\rm at}}\right)^2, \tag{12}$$

where $\Omega_D$ is the Debye frequency. At $T\ll T_D$ this contribution is exponentially small at frequencies that are much smaller than the hard optical mode frequencies.

The coupling with the acoustic mode (the coupling term[30] is the ''striction'' one, $\eta^2 u_{ii}$, $u_{ik}$ is the strain tensor) gives the contribution[14,31]

$$\Gamma_a\sim\Omega_D\frac{T}{T_{\rm at}}, \tag{13}$$

if the frequency of the $\eta$ vibrations is close to the frequency of the soft mode[16] as it takes place, e.g., in neutron experiments. For small frequencies [$\omega\ll(A/m)^{1/2}\equiv\omega_0$] the situation is more complicated.[16] For the weakly dispersive soft mode (WDSM) $(D/m)^{1/2}\ll\nu$, where $\nu$ is the sound speed, the estimation (13) remains valid for low frequencies as well, in the opposite case, for a strongly dispersive soft mode (SDSM), the contribution of the acoustic mode is negligible. An intermediate situation is possible as well, of course. We shall discuss below the two opposite cases.

The contribution of the anharmonic coupling within the soft branch can be estimated as[14]

$$\Gamma_c\sim\Omega_D\left(\frac{T}{T_{\rm at}}\right)^2\left(\frac{T_{\rm at}}{T-T_c}\right)^{1/2}. \tag{14}$$

This contribution is due to the fourth-order anharmonism [term $\propto\eta^4$ in Eq. (8)] and does not exhibit essential frequency dependence. Comparing Eqs. (13) and (14) one sees that outside the critical region [see Eq. (1)] $\Gamma_c<\Gamma_a$ and they become comparable at the boundary of the critical region.

We see that in the SDSM case the temperature dependences of the high-frequency and the low-frequency soft-mode damping coefficients are quite different. The main contribution to the high-frequency damping coefficient is due to the interaction with the acoustic modes [Eq. (13)] and, therefore, only a small anomaly is expected for this coefficient beyond the critical region. At the same time the main contribution to the low-frequency coefficient is due to the anharmonicity within the soft branch [compare Eqs. (14) and (12)] and the anomaly is very strong.

With all the anharmonicities taken into account a good approximation for the order-parameter low-frequency response function is to write it in the same form as in Eq. (10a) with $\gamma$ substituted for $m\Gamma_c$ given by Eq. (14). The renormalization due to the anharmonicity of the coefficient $A$ reduces to a redefinition of the phase-transition temperature and it is not reasonable to take into account explicitly. The only difference of this response function from that used in Ref. 5 is the temperature dependence of the damping constant [see Eq. (14)]. Using the result of Ref. 5 or Eqs. (7) and (5), one obtains for the contribution of the direct processes [the first term in the rhs of Eq. (5)]:

$$J_1(\Omega_L)=a_1^2\frac{mT\Gamma_c}{8\pi^2D^{3/2}}\frac{1}{A^{1/2}}\sim a_1^2\frac{d^2}{\Omega_D}\left(\frac{T_c}{T_{\rm at}}\right)^2\left(\frac{T_c}{T-T_c}\right), \tag{15}$$

where Eq. (14) has been used, it was estimated that $(D/m)d^{-2}\sim\Omega_D^2$, as $(D/m)^{1/2}=c$ is of order of magnitude of the sound speed, and for $D$, $A'$ the same estimation has been used following Eq. (10)

To calculate the contribution of the two-phonon processes it is enough to use the harmonic approximation neglecting the damping. As a result one obtains

$$J_2(\Omega_L)\approx a_2^2\frac{1}{16\pi}\frac{m^{1/2}T^2}{D^{5/2}}[q_{\rm max}-2(A/D)^{1/2}]. \tag{16}$$

It is the same temperature dependence as in Ref. 5 but we have calculated the coefficients as well. Estimating $q_{\rm max}$ as $d^{-1}$ one can see that at $A=0$ the value of $J_2(\Omega_L)$ given by Eq. (16) corresponds to a standard estimation of the contribution of the two-phonon processes to the SLR rate irrespective of a phase transition.[26] That is, this value is of the same order of magitude as the ''background value'' of the SLR, $J_0$, for an ideal crystal and without interaction between the relaxing centers. Using the same estimation as after Eq. (15) and estimating as well $a_1\sim a_2 d$, one sees that

$$J_0\sim a_1^2\frac{d^2}{\Omega_D}\frac{T^2}{T_{\rm at}^2}, \tag{17}$$

where a numerical factor is ignored. Comparing with Eq. (15) one sees that in the SDSM case the anomalous part of the SLR rate is more than its ''background'' value in a very broad temperature region ($T-T_c<T_c$), and at the boundary of applicability of the perturbation theory (i.e., at the boundary of the critical region), it is more than the background value by a factor of the order of value of $(T_{\rm at}/T_c)$, i.e., $10^2-10^3$ times.

The anomalous part of the SLR rate is even more in the case of WDSM. Substituting in Eq. (15) $\Gamma_a$ given by Eq. (13) for $\Gamma_c$ one finds that in this case

$$J_1(\Omega_L)\sim a_1^2\frac{d^2}{\Omega_D}\left(\frac{T_c}{T_{\rm at}}\right)^{3/2}\left(\frac{T_c}{T-T_c}\right)^{1/2}. \tag{17a}$$

One sees that the value of $J_1$ given by Eq. (17a) is larger than that given by Eq. (15) in the whole region of applicability of the perturbation theory [Eq. (1)]. The ''mixed'' contribution [the third term in the rhs of Eq. (5)] is absent in the symmetrical phase within our treatment.

### B. Nonsymmetrical phase

One has to take into account now that the order parameter has a nonzero equlibrium value. To the zero approximation, i.e., neglecting the $\eta$ fluctuations one has

$$\eta_e^2=-A/B. \tag{18}$$

To the approximation used in this paper this formula remains valid with the fluctuations taken into account as well, their effect reduces to a renormalization of the phase-transition

temperature. In Eq. (9) it is convenient now to single out $\eta_e$ and $\eta(\mathbf{r},t)-\eta_e$. The latter function we represent in the Fourier form:

$$\eta(\mathbf{r},t)-\eta_e=\sum_{\mathbf{k}}\ \eta(\mathbf{k},t)\exp(i\mathbf{kr}), \tag{19}$$

and write the Eq. (9) as

$$m\,\ddot{\eta}(\mathbf{q})+\gamma\dot{\eta}(\mathbf{q})+(-2A+Dq^2)\,\eta(\mathbf{q})$$
$$+3B\,\eta_e\sum_{\mathbf{k}}\ \eta(\mathbf{k})\,\eta(\mathbf{q}-\mathbf{k})=h(\mathbf{q},t), \tag{20}$$

where we have omitted the term of the third order in $\eta(\mathbf{k})$, i.e., the fourth-order anharmonocity. The effect of this anharmonicity has been discussed already for the symmetrical phase, it is basically the same for the nonsymmetrical one and we shall use the above results at the proper moment. Our aim now is to calculate the order-parameter response function taking into account the third-order anharmonicity described by the last term in the lhs of Eq. (20), i.e., to calculate $\eta(\mathbf{q},\Omega)$ in the presense of the force $h(\mathbf{q},\Omega)$ averaging over the fluctuations of $\eta(\mathbf{k})$ with $\mathbf{k}\neq\mathbf{q}$.

To this end we write Eq. (20) in the form

$$(-m\Omega^2+i\gamma\Omega-2A+Dq^2)\,\eta(\mathbf{q},\Omega)$$
$$=h(\mathbf{q},\Omega)-3B\,\eta_e\sum_{\mathbf{k}}\ \int\ \eta(\mathbf{k},\omega)\,\eta(\mathbf{q}-\mathbf{k},\Omega-\omega)\,d\omega. \tag{21}$$

The second term in the rhs plays the role of an additional force conjugated to $\eta(\mathbf{q})$. A part of this force is independent of $h(\mathbf{q},t)$ and describes interaction between the fluctuations of $\eta(\mathbf{q})$ and the rest of the $\eta$ fluctuations in the absence of the force $h$. This part is of no importance for us because we are interested just in the response of the system to the action of the force. Therefore, the second term in the rhs of Eq. (21) should be replaced, to the first approximation, by

$$-6B\,\eta_e\sum_{\mathbf{k}}\ \int\ \eta'(\mathbf{k},\omega)\,\eta^{(0)}(\mathbf{q}-\mathbf{k},\Omega-\omega)\,d\omega, \tag{22}$$

where $\eta'(\mathbf{k},\omega)$ is the change in the random variable $\eta(\mathbf{k},\omega)$ due to action of the force $h(\mathbf{q},\Omega)$, $\eta^{(0)}(\mathbf{k},\omega)$ represents the fluctuations in the absence of the force. One sees from Eq. (20) that, to the first approximation,

$$\eta'(\mathbf{k},\omega)=\frac{-6B\,\eta_e\,\eta^0(\mathbf{q},\Omega)\,\eta^{(0)}(-\mathbf{q}+\mathbf{k},-\Omega+\omega)}{-m\omega^2+i\gamma\omega-2A+Dk^2}, \tag{23}$$

where $\eta^0(\mathbf{q},\Omega)=\chi^{(0)}(\mathbf{q},\Omega)h(\mathbf{q},\Omega)$ and, for the nonsymmetrical phase,

$$\chi^{(0)}(\mathbf{q},\Omega)=\frac{1}{-m\Omega^2-2A+Dq^2+i\gamma\Omega}$$
$$\equiv\frac{1}{m[-\Omega^2+\omega_0^2(\mathbf{q})+i\Gamma\Omega]}, \tag{24}$$

where $\omega_0(\mathbf{q})$ is the soft-mode frequency. Thus, to first approximation, the response function

$$\chi^{-1(1)}(\mathbf{q},\Omega)=\chi^{-1(0)}(\mathbf{q},\Omega)-(6B\,\eta_e)^2$$
$$\times\sum_{\mathbf{k}}\ \int\ \frac{\langle|\,\eta^{(0)}(\mathbf{q}-\mathbf{k},\Omega-\omega|^2\rangle}{m[-\omega^2+\omega_0^2(\mathbf{k})+i\Gamma\omega]}\,d\omega. \tag{25}$$

Designating the second term in the rhs of Eq. (25) as $\Sigma(\mathbf{q},\Omega)=-(6B\,\eta_e)^2\Pi(\mathbf{q},\Omega)$ one obtains

$$\Pi(\mathbf{q},\Omega)=\frac{T}{8\pi^4m^2}\int\int\ \frac{\Gamma\,d\mathbf{k}\,d\omega}{\{[(\omega-\Omega)^2-\omega_0^2(\mathbf{k}-\mathbf{q})]^2+\Gamma^2(\omega-\Omega)^2\}[-\omega^2+\omega_0^2(\mathbf{k})+i\Gamma\omega]}, \tag{26}$$

where we have made use of Eqs. (7) and (24) to calculate $\langle|\,\eta^{(0)}(\mathbf{k},\omega)|^2\rangle$. It is important that the integrand is maximum at $\omega$ in the region of the soft-mode frequencies. This means that the damping constant in the integral is, in effect, the high-frequency one.

The real part of $\Pi(\mathbf{q},\Omega)=\Pi_1(\mathbf{q},\Omega)+i\Omega\Pi_2(\mathbf{q},\Omega)$ is of no interest for us: as $\Omega_L$ is smaller than any of the characteristic frequencies of the system it provides, in effect, the first fluctuation correction to the static response function that had been calculated earlier (see, e.g., Refs. 9,10). The function $\Pi_2(\mathbf{q}=0,\Omega)$ has already been calculated.[16] One finds

$$\Pi_2(\mathbf{q}=0,\Omega)=\frac{Tr_c}{8\pi D^2}\,\frac{\Gamma}{\Gamma^2+\Omega^2}. \tag{27}$$

One sees that the contribution of the considered third-order anharmonicity to the damping constant decreases rapidly with $\Omega$ at $\Omega>\Gamma$ and, as $\omega_0\gg\Gamma$ in the temperature region under discussion, the high-frequency damping constant $[\Gamma(\omega_0)\equiv\Gamma_0]$ is almost the same as in the symmetrical phase and can be estimated according to Eq. (13).

It is not the case, however, for the low-frequency damping constant. Using Eq. (27) and omitting a numerical factor which is close to unity one gets

$$\Omega^{-1}\ \mathrm{Im}\ \Sigma(\mathbf{q},\Omega=0)\equiv m\,\delta\Gamma(0)\approx\frac{TB}{D^{3/2}}\,\frac{|A|^{1/2}}{\Gamma_0}, \tag{28}$$

where $\delta\Gamma(0)$ is the increase of the low-frequency order-parameter damping constant in the nonsymmetrical phase. One sees that $\delta\Gamma(0)\propto(T_c-T)^{1/2}$. It is important that $\delta\Gamma(0)>\Gamma_0$ in all the region of applicability of the perturbation theory. Indeed, close enough to the phase transition, just at the boundary of applicability of the Landau (and the per-

turbation) theory the first factor in the rhs of Eq. (30) is of the order of magnitude of unity [see Eq. (10)] and the second factor can be presented as $\omega_0(\omega_0/\Gamma_0)$. At the boundary of applicability of the Landau theory $\omega_0 \sim \Gamma_0$, as one sees from Eq. (13), the estimation[10] $\omega_0 \sim \Omega_D |(T-T_c)/T_{\rm at}|^{1/2}$, and Eq. (1). Therefore at the boundary of applicability of the Landau theory $\delta\Gamma(0) \approx \Gamma_0$. As $\delta\Gamma(0) \propto (T_c - T)^{1/2}$, while $\Gamma_0$ changes slowly with temperature, one concludes that $\delta\Gamma(0)$ exceeds $\Gamma_0$ in all the regions of applicability of the perturbation theory.

Thus the relation between the low- and high-frequency order-parameter damping constants is quite different (in the region of applicability of the perturbation theory) in the symmetrical and nonsymmetrical phase. In the symmetrical phase, the low-frequency damping constant is *much smaller* than $\Gamma_0$ in the SDSM case and is of the same order of magnitude in the WDSM case, while in the nonsymmetrical phase it is *much bigger*. Using Eq. (13) and the above estimations of the coefficients one can present Eq. (28) in the form

$$\delta\Gamma(0) \sim \Omega_D \left( \frac{T_c - T}{T_{\rm at}} \right)^{1/2} \sim \omega_0 . \tag{29}$$

Let us suppose for a moment that the wave-vector dependence of the low-frequency damping constant can be negected. Using Eq. (15) with substitution $\Gamma_c$ by $\delta\Gamma(0)$ given by Eq. (29) one comes to the conclusion that the contribution of the direct processes does not depend on temperature in the nonsymmetrical phase and, comparing with Eq. (16) one concludes that the one-phonon contribution is larger than the two-phonon one by a factor of about $(T_{\rm at}/T_c)$, i.e., by 2–3 orders of magnitude. We shall see that the both conclusions are not correct due to the wave-vector dependence of $\mathrm{Im}\,\Sigma(\mathbf{q},\Omega \approx 0)$ which we will now study.

The function $\Pi_2(\mathbf{q},\Omega=0)$ is calculated and discussed in the Appendix. The value of the spin-lattice relaxation rate is determined by this function in the region $q \sim r_c^{-1}$ which is different from the function for $q \approx 0$ due to a dispersion in the region $q \sim \Gamma_0/c$, $c=(D/m)^{1/2}$. Taking this into account and using Eq. (47) one obtains, to a reasonable approximation for this region:

$$\Omega^{-1}\ \mathrm{Im}\ \Sigma(q \approx r_c^{-1}, \Omega=0) \equiv m\,\delta\Gamma(q \approx r_c^{-1}) \sim \frac{TB}{Dc}. \tag{30}$$

Now we will take into account that the changes of the order parameter in the nonsymmetrical phase lead to changes in temperature. The influence of this coupling on the order-parameter response function has been discussed by several authors.[32–35] For the corresponding contribution $\chi^{-1}(\mathbf{q},\Omega) - \chi^{-1(0)}(\mathbf{q},\Omega) = \Sigma_T(\mathbf{q},\Omega)$ one has

$$\Sigma_T(\mathbf{q},\Omega) = i\Omega T \frac{A'^2 \eta_e^2}{i\Omega C + \kappa q^2}, \tag{31}$$

where $\kappa$ is the thermal conductivity coefficient, $C$ is the specific heat per unit volume. Let us compare the two terms in the denominator in the rhs of Eq. (31) for $q \sim r_c^{-1}$ because we are interested just in this region. For $T \geqslant T_D$ the thermal conductivity coefficient in an ideal dielectric crystal can be estimated as[28] $(T_{\rm at}/T)\Omega_D d^{-1}$. Estimating $C \sim d^{-3}$ and taking into account that $r_c^{-2} \sim d^{-2}|T-T_c|/T_{\rm at}$ (one has to use the estimations of the coefficients in the end of Sec. III) one finds that $\kappa r_c^{-2} C^{-1} \sim \Omega_D |T-T_c|/T_c$. Therefore, even at the boundary of the applicability of the perturbation theory [Eq. (1)] the Larmor frequency can be considered as quite small. Thus one obtains

$$\Omega^{-1}\ \mathrm{Im}\ \Sigma_T \approx T \frac{A'^2 D}{B\kappa}. \tag{32}$$

Comparing Eqs. (32) and (30) and using the above estimations one sees that the rhs of Eq. (32) is smaller than the rhs of Eq. (30) about $T_{\rm at}/T_c$ times. This difference can be less in real crystals because the defects can strongly diminish the thermal conductivity at not so high temperatures, while Eq. (30) contains, at the end, thermodynamic coefficients which are less sensitive to the defects than the kinetic ones.

Now we will calculate the contribution of the ''mixed'' term, i.e., of the last term in the rhs of Eq. (5). The correlation function

$$\langle \eta(\mathbf{q},\Omega)\,\eta^2(-\mathbf{q},-\Omega)\rangle = \sum_{\mathbf{k}} \int \langle \eta(\mathbf{q},\Omega)\,\eta(\mathbf{k},\omega) \times \eta(-\mathbf{q}-\mathbf{k},-\Omega-\omega)\rangle\, d\omega \tag{33}$$

is zero if the anharmonicity is neglected. To take it into account the first approximation, one can use Eq. (23) considering $\eta^{(0)}(\mathbf{q},\Omega)$ as the fluctuation in the harmonic approximation. One finds

$$\langle \eta(\mathbf{q},\Omega)\,\eta^2(-\mathbf{q};-\Omega)\rangle = -18B\,\eta_e \langle |\eta^{(0)}(\mathbf{q},\Omega)|^2\rangle \times \sum_{\mathbf{k}} \int \frac{|\eta^{(0)}(\mathbf{q}-\mathbf{k},\Omega-\omega)|^2\rangle}{m[-\omega^2+\omega_0^2(\mathbf{k})+i\Gamma\omega]}\, d\omega. \tag{34}$$

Comparing Eqs. (34) and (25) one sees that

$$\langle \eta(\mathbf{q},\Omega)\,\eta^2(-\mathbf{q},-\Omega)\rangle + \langle \eta(\mathbf{q},-\Omega)\,\eta^2(-\mathbf{q},\Omega)\rangle = \langle |\eta^{(0)}(\mathbf{q},\Omega)|^2\rangle \frac{\Sigma_1(\mathbf{q},\Omega)}{B\,\eta_e}, \tag{35}$$

where it is taken into account that $\Sigma_1(\mathbf{q},\Omega)$ and $\Sigma_2(\mathbf{q},\Omega)$ are, correspondingly, even and odd functions of $\Omega$). One sees that it is easy to compare the ''mixed'' contribution with the harmonic approximation for the first-order one [the first term in the rhs of Eq. (5)], one has just to compare the fraction in the rhs of Eq. (35) with unity. One can well use the static value $\Sigma_1(\mathbf{q},\Omega=0)$ that has already been calculated:[10]

$$\Sigma_1(\mathbf{q}) = \frac{18}{2\pi} \frac{B^2 \eta_e^2 T}{D^2 q} \tan^{-1} \frac{q r_c}{2}. \tag{36}$$

Using the estimations of the coefficients (Sec. II) one finds that for $q \sim r_c^{-1}$ the fraction is equal to $T_c/T_{\rm at}$ by the order of magnitude and consequently the ''mixed'' contribution is small.

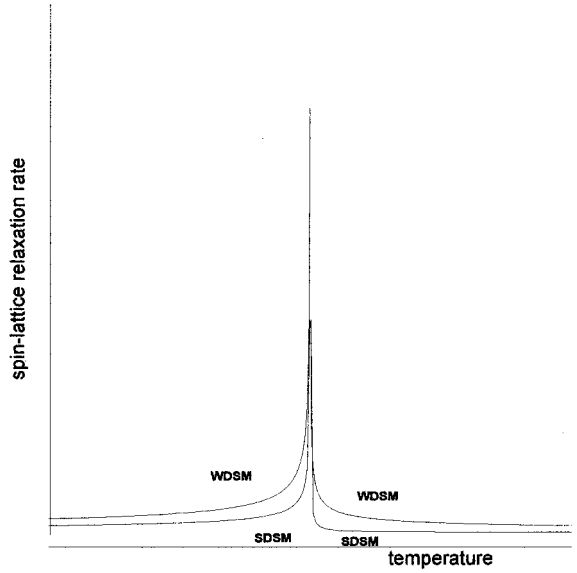


FIG. 1. Qualitative form of the spin-lattice relaxation-rate anomaly at phase transition in an ideal displacive system (see text).

Therefore, to estimate the contribution of the third-order anharmonicity arising in the nonsymmetrical phase to the spin-lattice relaxation rate anomaly ($J_{1,3}$), one can substitute the last term in Eq. (30) for $\Gamma_c$ in Eq. (15) to obtain

$$J_{1,3}\approx a_1^2\,\frac{d^2}{\Omega_D}\left(\frac{T_c}{T_{\rm at}}\right)^{3/2}\left(\frac{T_c}{|T-T_c|}\right)^{1/2}. \tag{37}$$

The contribution of the fourth-order anharmonicity is still given by Eq. (15) and the contribution due to coupling to the acoustic mode by Eq. (17a) in the WDSM case. One sees that the SLR anomaly is more symmetric in the WDSM case than in the SDSM case (Fig. 1): Eq. (37) is basically the same as Eq. (17a).

## IV. TWO-COMPONENT ORDER PARAMETER

There is nothing special in the case for the symmetrical phase but there are some specific features for the nonsymmetrical one. To illustrate them we shall consider separately, the cases of the commensurate and incommensurate nonsymmetrical phase.

### A. Commensurate phase

In this case we will restrict ourselves to the Landau free energy of the form

$$\Phi(\eta_1,\eta_2)=\frac{A}{2}(\eta_1^2+\eta_2^2)+\frac{B_1}{4}(\eta_1^2+\eta_2^2)^2+\frac{B_2}{2}\eta_1^2\eta_2^2 + \frac{D}{2}[\nabla\eta_1)^2+(\nabla\eta_2)^2] \tag{38}$$

with $B_2>0$. The equlibrium values of the order-parameter components in the nonsymmetrical phase are: $\eta_{1e}^2=-A/B_1$, $\eta_{2e}=0$. Analogously to Eq. (20) one obtains for the Fourier components of $\eta_1(\mathbf{r},t)-\eta_{1e}$ and $\eta_2(\mathbf{r},t)$

$$m\ddot{\eta}_1(\mathbf{q})+\gamma\dot{\eta}_1(\mathbf{q})+(2B_1\eta_{1e}^2+Dq^2)\eta_1(\mathbf{q}) +3B_1\eta_{1e}\sum_{\mathbf{k}}\eta_1(\mathbf{q}-\mathbf{k})+(B_1+B_2)\eta_{1e} \times\sum_{\mathbf{k}}\eta_2(\mathbf{k})\eta_2(\mathbf{q}-\mathbf{k})=h_1(\mathbf{q},t), \tag{39a}$$

$$m\ddot{\eta}_2(\mathbf{q})+\gamma\dot{\eta}_2(\mathbf{q})+(B_2\eta_{1e}^2+Dq^2)\eta_2(\mathbf{q}) +2(B_1+B_2)\eta_{1e}\sum_{\mathbf{k}}\eta_2(\mathbf{k})\eta_1(\mathbf{q}-\mathbf{k})=h_2(\mathbf{q},t), \tag{39b}$$

In a similar way as in Sec. III one finds

$$\Sigma_1(\mathbf{q},\Omega)=-36B_1^2\eta_{1e}^2\sum_{\mathbf{k}}\int\frac{\langle|\eta_1^{(0)}(\mathbf{q}-\mathbf{k},\Omega-\omega)|^2\rangle}{m[-\omega^2+\omega_{01}^2(\mathbf{k})+i\Gamma\omega]}\,d\omega -4(B_1+B_2)^2\eta_{1e}^2 \times\sum_{\mathbf{k}}\int\frac{\langle|\eta_2^{(0)}(\mathbf{q}-\mathbf{k},\Omega-\omega)|^2\rangle}{m[-\omega^2+\omega_{02}^2(\mathbf{k})+i\Gamma\omega]}\,d\omega, \tag{40a}$$

$$\Sigma_2(\mathbf{q},\Omega)=-4(B_1+B_2)^2\eta_{1e}^2 \times\sum_{\mathbf{k}}\int\frac{\langle|\eta_1^{(0)}(\mathbf{q}-\mathbf{k},\Omega-\omega)|^2\rangle}{m[-\omega^2+\omega_{02}^2(\mathbf{k})+i\Gamma\omega]}\,d\omega -4(B_1+B_2)^2\eta_{1e}^2 \times\sum_{\mathbf{k}}\int\frac{\langle|\eta_2^{(0)}(\mathbf{q}-\mathbf{k},\Omega-\omega)|^2\rangle}{m[-\omega^2+\omega_{01}^2(\mathbf{k})+i\Gamma\omega]}\,d\omega, \tag{40b}$$

where $\omega_{01}^2(\mathbf{k})=(2B_1\eta_{1e}^2+Dk^2)/m$, $\omega_{02}^2(\mathbf{k})=(B_2\eta_{1e}^2+Dk^2)/m$. One sees the similarity to Eq. (25) which allows us to use the results of the Appendix to estimate Im $\Sigma_1$, Im $\Sigma_2$. If $B_1$ and $B_2$ are of the same order of magnitude the discussion for the case of one-component order parameter (Sec. III) is applicable for $\Sigma_{11}$ as well with the only difference being that there are now two different correlation radii for the two terms in the rhs of Eq. (40a). As long as they are of the same order of magnitude it is not important for the order-by-magnitude estimations. To discuss Im $\Sigma_2(\mathbf{q},\Omega\approx0)$ one can use as well Eq. (A1) substituting $A_1(k)$ for $A(k)$ and $A_2(\mathbf{q}-\mathbf{k})$ for $A(\mathbf{q}-\mathbf{k})$ $[A_{1,2}(k)=m\omega_{01,2}^2(k)]$ or vice versa. Realizing that both for $q=0$ and for $q\sim r_{c1,2}$ the integral is determined by $k\sim A/\gamma c$ and taking into account Eq. (13) one comes to the conclusion that the estimation of Eq. (30) is valid for this case as well, both for $q=0$ and for $q\sim r_{c1,2}$. Thus the $q$ dispersion at $q\sim\Gamma/c$ is absent for $\Sigma_2(\mathbf{q},\Omega\approx0)$.

Let us emphasize that, while the high-frequency damping constant is the same for the two components of the order parameter (there is, of course, some difference proprtional to $T_c-T$ but it can be neglected), the low-frequency damping constants are different for the two components. They are of the same order of magnitude in the $k$ region essential for the SLR but even in this region the difference between them is of the same order of magnitude as the constants themselves (or even more, taking into account that order-of-magnitude

estimations are quite rough) and in general they have different $q$ and temperature dependence.

To calculate the SLR rate one has to generalize Eq. (4) as

$$F=F_0+a_1\eta_1+a_2\eta_1^2+b_1\eta_2+b_2\eta_2^2\ldots\ . \tag{41}$$

The coefficients $a_n$, $b_n$ are, in general, different for different nuclei in the unit cell, and it can happen, in particular, that $a_n$ or $b_n$ are zero for the nucleus in question. For the case $b_n=0$ the estimations repeat those for the one-component order parameter. For the case $a_n=0$ there are no losses due to thermal conductivity and the ''mixed'' contribution is absent, to the first approximation. As these two contributions are negligible anyway and, for $B_1\sim B_2$, the damping constants for $q\sim r_c^{-1}$ are of the same order of magnitude. The same can be said about the two SLR rates. In the general case the ''mixed'' contribution contains additional terms of the type $\langle\eta_1(\mathbf{q},\Omega_L)\eta_2(-\mathbf{q},-\Omega_L)\rangle$ and $\langle\eta_1(\mathbf{q},\Omega_L)\eta_2^2(-\mathbf{q},-\Omega_L)\rangle$ that are nonzero because of the anharmonicity but can be shown to be negligible as well.

### B. Incommensurate phase

The simplest case of an IC system corresponds to the Landau free energy Eq. (38) with $B_2=0$ (and $B_1\equiv B$). Now $\eta_1(\mathbf{r},t)-\eta_{1e}$ corresponds to longitudinal fluctuation (amplitudon, to the harmonic approximation) and $\eta_2(\mathbf{r},t)$ is the transversal one (phason, to the harmonic approximation).

To find the transversal response function one can set $B_2=0$ in Eq. (40b) (the integrals remain finite) and taking into account, of course, that $B_2=0$ as well in the nonrenormalized transversal response function [three first terms in Eq. (39b)]. Therefore for the transversal damping constant the above estimations are valid as well. The result of Ref. 6 for the transversal (phason) contribution to the SLR rate remains valid.

The above method is not valid for the longitudinal response function: the second term in the rhs of Eq. (40a) becomes divergent at $q\to 0$ and the perturbation procedure used is no longer applicable. It is possible, nevertheless to reformulate the pertubation theory in such a way that the first approximation remains valid even at $B_2=0$. The trick is to develop the perturbation theory not for the reciprocal response functions as in Sec. IV but for the response functions themselves. The result of this section for the response function of $\eta_1(\mathbf{q})$ reads

$$\chi_{11}(\mathbf{q},\Omega)=\frac{1}{-m\omega^2+2B_1\eta_{1e}^2+Dk^2+i\Gamma_1\Omega}-\frac{\Sigma_{11}(\mathbf{q},\Omega)}{(-m\omega^2+2B_1\eta_{1e}^2+Dk^2+i\Gamma_1\Omega)^2}, \tag{42}$$

where $\Sigma_{11}(\mathbf{q},\Omega)$ is given by Eq. (40a). Now one can safely set $B_2=0$ and the divergencies arising are the real divergencies of the response function at $\mathbf{q}\to 0$, $\Omega\to 0$, because from the rest of the perturbation series no stronger divergencies appear if the conditions (1), (10) are fulfilled. This statement is the classical version of the result by Vaks, Larkin, and Pikin[17] who considered Heisenberg ferromagnet. The reason for the effectiveness of this version of the perturbation theory is mutual compensation of the divergencies in the higher-order terms which is quite typical for an IC phase (see, e.g., Refs. 36–37). Let us mention as well that the static longitudinal response function $\chi_{11}(\mathbf{q})$ obtained by this method coincides with that obtained by other authors[19–21] but we are not aware of calculations of the dynamic response function for structural IC systems with the divergencies that are due to the anharmonic longitudinal-transversal interaction taken into account.

Designating the limit $B_2=0$ of the second term in the rhs of Eq. (40a) as $-4(B\eta_{1e})^2\Pi_t(\mathbf{q},\Omega)$ one has

$$\Pi_t(\mathbf{q},\Omega)=\frac{T}{8\pi^4m^2}\int\int\frac{\Gamma_2 d\mathbf{k}\,d\omega}{\{[(\omega-\Omega)^2-c^2(\mathbf{k}-\mathbf{q})^2]^2+\Gamma_2^2(\omega-\Omega)^2\}(-\omega^2+c^2k^2+i\Gamma_2\omega)}, \tag{43}$$

where we have the possibility of improving the first-order perturbation formula substituting $\Gamma_2$ for $\Gamma$, where $\Gamma_2$ takes into account the renormalization described by Eq. (40b) at $B_2=0$. This substitution corresponds to a partial summation of the perturbation series. We have mentioned already that $\Gamma_2$ has no essential $k$ dependence in the region of interest and can be estimated according to Eq. (30).

To reveal the main divergencies at $\Omega\to 0$, $q\to 0$, one can set $m=0$ in Eq. (43). Integrating over $\omega$ one obtains

$$\Pi_t(\mathbf{q},\Omega)=\frac{T}{8\pi^3}\int\frac{[(\mathbf{q}-\mathbf{k})^2+k^2-i\Omega\gamma_2]d\mathbf{k}}{(\mathbf{q}-\mathbf{k})^2[[D(\mathbf{q}-\mathbf{k})^2+Dk^2]^2+\gamma_2^2\Omega^2]}, \tag{44}$$

where $\gamma_2=m\Gamma_2$. For $\gamma_2\Omega\ll Dq^2$ one can set $\Omega=0$ in the denominator of the integral in Eq. (44). As a result one has under this condition

$$\Pi_t(\mathbf{q},\Omega)=\frac{T}{8\pi D^2q}\left(\frac{\pi}{2}-i\Omega\,\frac{\Gamma_2}{c^2q^2}\right). \tag{45}$$

In the opposite limit $\gamma_2\Omega\gg Dq^2$ one can set $q=0$ in Eq. (44) to obtain

$$\Pi_t(\Omega)=\frac{T}{8\pi D^{3/2}}\frac{1-i}{(|\Omega|\gamma_2)^{1/2}}. \tag{46}$$

As an interpolation for $\Pi_t(\mathbf{q},\Omega)$ one can use

$$\Pi_t(\mathbf{q},\Omega)=\frac{T}{8\pi D^{3/2}}\left(\frac{1}{[D(2q/\pi)^2+\gamma_2|\Omega|]^{1/2}}-\frac{i\gamma_2\Omega}{(Dq^2+\gamma_2|\Omega|)^{3/2}}\right). \quad (47)$$

Taking into account $m\neq 0$ leads to an additional diverging term in $\mathrm{Im}\,\Pi_t(\mathbf{q},\Omega)=-i\Omega\Pi_{t2}(\mathbf{q},\Omega)$ that can be estimated as

$$\delta\Pi_{t2}(\mathbf{q},\Omega)=\frac{T}{8\pi D^2 q}\frac{1}{\Gamma_2} \quad (48)$$

for $q\ll\Gamma_2/c$. In the opposite limit, for $q\gg\Gamma_2/c$, one finds

$$\delta\Pi_{t2}(\mathbf{q},\Omega)=\frac{T}{4\pi D^2 q}\frac{1}{cq}. \quad (49)$$

One sees that at at small frequencies which we are interested in ($\Omega_L\ll\Gamma_2$) $\delta\Pi_{t2}(\mathbf{q},\Omega)$ can be neglected comparing with $\Pi_{t2}(\mathbf{q},\Omega)$ given by Eq. (47) if $q\ll\Gamma_2/c$, but at $q\gg\Gamma_2/c$ it provides the leading contribution. Note that the rhs of Eqs. (49) and (A7) are the same, up to a numerical factor. Thus the specific ''softness'' of the transversal ''displacements'' influence the longitudinal response function for a relatively small part of the $q$ space and in the main part of this space the effect of the transversal fluctuations is similar to that of one-component order parameter.

We are now in position to discuss the SLR rate due to the transversal and longitudinal fluctuations. Under special conditions and in special points of the NMR spectrum one can probe these SLR rates separately.[6] Let us restrict ourselves to the one-phonon processes.

The transversal SLR rate can be calculated using the conventional formula[6] but taking into account that the damping constant ($\Gamma_2$) in the formula should have the order of magnitude given by Eq. (30) and does not coincide with the damping constant of the soft mode measured from neutron or light-scattering spectra.

Qualitatively it is valid as well for the longitudinal fluctuations excluding the effect of the region of small $q$ that should be discussed separately. Designating the longitudinal damping constant for $q\sim r_c^{-1}$ as $\Gamma_1$ and redefining the zero-order response function just by replacing $\Gamma$ by $\Gamma_1$ to incorporate the nondivergent part of the renormalization one can calculate the correction due to the small-$q$ divergence. To reveal the role of this divergence it is enough to compare $\int \mathrm{Im}\,\chi_{11}^{(0)}(\mathbf{q},\Omega_1)d\mathbf{q}$ and $\int \mathrm{Im}\,\delta\chi_{11}(\mathbf{q},\Omega_1)d\mathbf{q}$, (see Sec. II) the first integral being calculated for all the $q$ region and the second for the region $q<\Gamma_2/c$ with

$$\chi_{11}^{(0)}=\frac{1}{B\eta_{1e}^2+Dq^2+i\gamma_1\Omega_L};$$

$$\delta\chi_{11}=\frac{i\Omega_L 4B\eta_{1e}^2\Pi_{t2}}{(B\eta_{1e}^2+Dq^2+i\gamma_1\Omega_L)^2}, \quad (50)$$

where $\gamma_1=m\Gamma_1$ and $\Pi_{t2}$ is given by Eq. (47). One finds for the ratio of the second and the first integral:

$$\frac{\delta T_{1l}^{-1}}{T_{1l}^{-1(0)}}\approx\frac{1}{2^{3/2}\pi^2}\frac{\Gamma_2}{\Gamma_1}\xi\ln\frac{\Gamma_2}{\Omega_L}. \quad (51)$$

One sees that the correction connected with the transversal softness depends logarithmically on the Larmor frequency but it is smaller than the frequency-independent part in the region of applicability of the perturbation theory ($\xi\ll 1$).

## V. ORDER-DISORDER SYSTEMS

Within the one-ion model there is no region of applicability of the perturbation theory for order-disorder systems[10,11] but the crystals studied experimentally are usually rather far from this model and it might be of some interest to discuss, within the perturbation theory, if the above considered effects could be of some importance for order-disorder systems as well. Such a discussion will be inevitably rather speculative because, as we have seen, the anhamonicity governed corrections to the low-frequency response function can be determined by the high-frequency order-parameter dynamics and the latter is hardly known and not universal for the order-disorder systems. For the sake of illustration we shall assume that to obtain the order-disorder case one can just put $m\to 0$ in the above formulas.

It is worthwhile to discuss, of course, the case of the nonsymmetrical phase only. The new third-order anharmonicities appearing at the transition to this phase that were of main interest above can be expected to be of less importance for order-disorder systems: the latter are strongly anharmonic by their nature and some change of the anharmonicity can hardly produce a drastic effect. Specifically one can set $m\to 0$ in Eq. (A1) to see that for one-component the anharmonicity arising at the transition provides a correction proportional to $\xi$ to the original viscosity constant ($\gamma$), i.e., it is a part of the corrections to the mean-field theory due to the critical fluctuations.[38] For an IC phase Eq. (47) becomes valid for all the wave vectors (smaller than the atomic one, $d^{-1}$) with $\gamma_2=\gamma_1$.

$$\frac{\delta T_{1l}^{-1}}{T_{1l}^{-1(0)}}\approx\frac{1}{2^{3/2}\pi^2}\xi\ln\frac{\Omega_D}{\Omega_L}, \quad (52)$$

where it is estimated that $Dd^2/\gamma\sim\Omega_D$. The correction due to the longitudinal-transversal anharmonic interaction may be more here than in the displacement case but insignificantly because of the logarithm.

## VI. CONCLUSIONS

We attempted to improve the theory of the SLR rate anomalies at structural phase transitions taking into account the relevant results for the lattice response functions that were not taken into account in the previous versions of the theory. The most remarkable of these results is, probably, a strong wave-vector dispersion of the low-frequency damping constant that influences essentially the results on the SLR rate anomaly. An essential difference in the damping constants for a different component of the order parameter might be considered as an unexpected result as well. A specific feature of the IC phase, the divergence of the transversal

fluctuations at zero wave vectors, reveals itself in the Larmor frequency dependence of the longitudinal (''amplitudon'') SLR rate. However this dependence is rather weak, a logarithmic one, and proves to be more weak than it was found in some experiments (see, e.g., Ref. 24). Therefore, the longitudinal-transverse interaction in an *ideal crystal* considered in this paper cannot explain the experiments, one can speculate that the defects which are known to influence dramatically the properties of IC phases might be responsible for the disagreement. As well, one can suspect that the ''transversal contamination'' of the longitudinal contribution somehow took place.

One of the most studied effect of the defects in IC phases is so-called ''phason gap'' that has been reported repeatedly (for a review see Ref. 39). The method to find the ''phason gap'' uses the assumption that the phason (transversal) and the amplitudon (longitudinal) damping constants are equal. We have shown above that it is not the case. Still, as they are expected to be of the same order of magnitude, one may hope that the order of magnitude of the reported ''phason gaps'' is fairly close to the real one.

## APPENDIX

Integrating in Eq. (26) over $\omega$ one obtains

$$\Pi_2(\mathbf{q},\Omega=0)=\frac{T\gamma}{8\pi^3}\int\frac{\left[A(k)+A(\mathbf{q}-\mathbf{k})+2\frac{\gamma^2}{m}\right]d\mathbf{k}}{A(k)A(\mathbf{q}-\mathbf{k})\left\{[A(\mathbf{q}-\mathbf{k})-A(k)]^2+2\frac{\gamma^2}{m}[A(k)+A(\mathbf{q}-\mathbf{k})]\right\}},\tag{A1}$$

where $A(\mathbf{k})=m\omega_0^2(\mathbf{k})=-2A+Dk^2$. After integration over the spherical angles one obtains

$$\Pi_2(\mathbf{q},\Omega=0)=\frac{T\gamma}{4\pi^2D^3}\left[\int_0^{k_{\rm at}}\frac{\tan^{-1}[kq/k_1(k^2+r_c^{-2}+q^2/4)^{1/2}]}{qk_1(k^2+r_c^{-2}+q^2/4)^{3/2}}\,kdk+\int_0^{k_{\rm at}}\frac{\ln[(k+q/2)^2+r_c^{-2}/(k-q/2)^2+r_c^{-2}]}{q(k^2+r_c^{-2}+q^2/4)^2}\,kdk\right],\tag{A2}$$

where $k_1=\gamma/(mD)^{1/2}=\Gamma/c$, $k_{\rm at}\sim d^{-1}$ and $r_c^{-2}=-2A/D$. The characteristic wave vector $k_1$ is important for what follows. It is worthwhile to mention that $k_1\ll r_c^{-1}$ in all the region of applicability of the perturbation theory.

Let us discuss first the $q$ dependence of the first integral ($I_1$) in Eq. (A2). It is seen that the main contribution to this integral comes from the region $k\leqslant(r_c^{-2}+q^2/4)^{1/2}$. So for $q\ll k_1$ the $\tan^{-1}$ can be replaced by its argument and one obtains

$$I_1\approx 2k_1^{-2}\int_0^{k_{\rm at}}\frac{k^2dk}{(k^2+r_c^{-2}+q^2/4)^2}\approx\frac{\pi}{2}\,k_1^{-2}(r_c^{-2}+q^2/4)^{-1/2},\tag{A3}$$

where it is taken into account that $k_{\rm at}\gg r_c^{-1}$. For $q\gg k_1$ one can replace the $\tan^{-1}$ by $\pi/2$ to find

$$I_1\approx\pi k_1^{-1}q^{-1}\int_0^{k_{\rm at}}\frac{kdk}{(k^2+r_c^{-2}+q^2/4)^{3/2}}\approx\pi k_1^{-1}q^{-1}(r_c^{-2}+q^2/4)^{-1/2}.\tag{A4}$$

One sees that at $q=k_1$ the two formulas match perfectly.

The second integral in Eq. (A2) ($I_2$) can be estimated for $q\ll r_c^{-1}$. The logarithm can be approximated here as $2kq/(k^2+r_c^{-2}+q^2/4)$ to find

$$I_2\approx 2\int_0^{k_{\rm at}}\frac{k^2dk}{(k^2+r_c^{-2}+q^2/4)^{3/2}}\approx\frac{\pi}{8}\,(r_c^{-2}+q^2/4)^{-3/2}.\tag{A5}$$

As $k_1\ll r_c^{-1}$ one can neglect $I_2$ for $q\ll r_c^{-1}$ and, as the region $q\gg r_c^{-1}$ is of no importance to treat the spin-lattice relaxation rate anomaly, we neglect it as well. Therefore, in the $q$ regions of our interest we can use, as a good approximation, the formula

$$\Pi_2(\mathbf{q},\Omega=0)\approx\frac{T}{8\pi D^2\Gamma}\,\frac{r_c}{(1+q^2r_c^2/4)^{1/2}(1+qk_1^{-1}/2)}.\tag{A6}$$

For $q\gg r_c^{-1}$ this formula becomes very simple:

$$\Pi_2(\mathbf{q},\Omega=0)\approx\frac{T}{2\pi D^2cq^2}.\tag{A7}$$

For the order-of-magnitude estimations one can use this formula for $q\sim r_c^{-1}$ as well.

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