Low-frequency phonon dynamics and spin-lattice relaxation time in the incommensurate phase at low temperatures

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The low-frequency phonon-response functions, both for transverse (phason) and longitudinal (amplitudon) fluctuations, are calculated for a displacive incommensurate system at low temperatures. The results obtained are used to calculate the spin-lattice relaxation rate governed either by transverse or longitudinal fluctuations at temperatures much lower than the Debye temperature but much higher than the energy corresponding to the Larmor frequency. It is found that direct processes determine the spin-lattice relaxation rate in both cases. The transverse spin-lattice relaxation rate is proportional to temperature T and does not depend on the Larmor frequency. The longitudinal contribution is proportional to T^3 and does not depend on the Larmor frequency as well. [S0163-1829(97)02137-1]

I. INTRODUCTION

It is quite natural to expect that specific features of the lattice dynamics of incommensurate(IC) phases should manifest themselves specifically in their low-temperature behavior. Until recently, the only way to study this behavior was to work on quasi-one-dimensional compounds with Peierls transition.[1,2] Now the low-T properties of the nonconductive molecular crystal bis(4-chlorophenyl)sulfone (BCPS) have attracted some attention.[3–13] This crystal has an IC phase that exists in the T interval from 150 K down to the lowest temperatures investigated.[5,8] ^{35}Cl NQR measurements have revealed[5,6,8] a strong decrease in the spin-lattice relaxation rate T_1^{-1} below 110 K. Recently, measurements have been extended[12] down to several K and a monotonous decrease of T_1^{-1} as the temperature decreases has been found. The interpretation of the experiments raises, however, some questions. The aim of this paper is to present a systematic discussion of the low-T and low-frequency lattice dynamics of IC phases which is relevant to the interpretation of the T_1 data.

It is known[14] that NQR and NMR signals in IC phases exhibit a broad spectrum and the spin-lattice relaxation rate varies over the spectrum. This is due to different contributions of the longitudinal and transverse fluctuations (to the harmonic approximation—of the amplitudon and phason ones) to the spin-lattice relaxation rate at different parts of the line. (We shall not consider this question but rather discuss the situations when the spin-lattice relaxation rate is governed completely either by the transverse or by the longitudinal fluctuations.) In fact, what is calculated are definite correlation functions. In the general case these correlation functions enter the formulas for the transition probabilities defining the evolution of the level occupation numbers, so the generalization of the results of the present paper is straightforward but is beyond it. For the main part of the spectrum the transverse fluctuations contribution is more important than that of longitudinal ones. That is why we begin with the discussion of the first contribution. For the sake of brevity we shall use the notations of phason and amplitudon instead of transverse and longitudinal fluctuations even though we shall deal with anharmonic effects.

The paper is organized as follows. In Sec II we discuss the relative role of the direct and the Raman processes in the phason-mediated spin-lattice relaxation in the IC phase and come to the conclusion that the phason-governed T_1^{-1} is proportional to T in the low-temperature region whose boundary is determined by the T-dependence of the phason damping constant. In Sec. III we investigate this temperature dependence in the low-T limit. In Sec IV we discuss the T-dependence of the amplitudon contribution to the spin-lattice relaxation time. In Sec V we summarize the results obtained and discuss their relevance to the experimental data.

II. THE PHASON-MEDIATED SPIN-LATTICE RELAXATION RATE

We shall be interested in the spin-lattice relaxation rate as measured in NMR and NQR experiments at temperatures from about 1 K. As 1 K corresponds to frequencies about 10^{11} s^{-1} and the NMR frequencies are normally of the order of 10^7 s^{-1} (the NQR ones can be much lower) we shall assume in this paper that $\hbar\Omega_L \ll T$ where Ω_L is the Larmor frequency. At the temperatures of our interest the effects of the phonon bottleneck can be neglected: according to Ref. 12 the spin-lattice relaxation time for ^{35}Cl in BCPS at $T \approx 1$ K is about 1 s^{-1} while the phason lifetime is much less at the same temperatures [see Eq. (22) below]. Thus one can use for the spin-lattice relaxation rate the same formulas as in Ref. 14.

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.[15] 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 (EFG) 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. Here 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. In this case[15]

$$T_1^{-1}=B\langle|F(\Omega_L)|^2\rangle, \tag{1}$$

where B is a constant, Ω_L is the Larmor frequency, $F(\omega)=\int F(t)\exp(-i\omega t)dt$, and $\langle\,\rangle$ designates the statistical average. Introducing the order parameter for normal-IC phase transition, $\eta=\eta_1+i\eta_2$, and assuming that the equilibrium value of η_1 is nonzero in the IC phase but that of η_2 is zero one associates the fluctuations of η_1 (the longitudinal ones) with amplitudons and those of η_2 (the transverse ones) with phasons. Then the phason contribution is singled out from the right-hand side (rhs) of Eq. (1) as[14]

$$T_{1\text{dph}}^{-1}=a_1^2\sum_k\langle|\eta_2(\mathbf{k},\Omega_L)|^2\rangle, \tag{2}$$

where

$$\eta_2(\mathbf{k})=\frac{1}{V}\int\eta_2(\mathbf{r})\exp(-i\mathbf{kr})d\mathbf{r},$$

V is the volume and the coefficient a_1 appears in the expansion of the coefficient F:

$$F=F_0+a_1\eta_2+a_2\eta_2^2+\cdots. \tag{3}$$

For further calculations[14] one takes into account that the phason is overdamped and considers it as a relaxator, i.e., in the classical limit one has

$$\langle|\eta_2(\mathbf{k},\Omega_L)|^2\rangle=\frac{\gamma_2T}{\pi V(D^2k^4+\gamma_2^2\Omega_L^2)}, \tag{4}$$

where γ_2 is the phason "viscosity constant," $\gamma_2/2m=\Gamma_2$ is referred to as the phason damping constant, m is the phason mass density, or, rather, the mass density corresponding to the soft mode normal coordinate, and $(D/m)^{1/2}=c$ is referred to as the phason velocity.

As we are interested in the low-T region and the phason viscosity is expected to go to zero when $T\to0$ it is more reasonable to use a less simplified formula than Eq. (4) and represent the phason as an oscillator with damping.

$$\langle|\eta_2(\Omega_L,\mathbf{k})|^2\rangle=\frac{\gamma_2T}{\pi V[(-m\Omega_L^2+Dk^2)^2+\gamma_2^2\Omega_L^2]}. \tag{5}$$

The classical expression is applicable as long as $\hbar\Omega_L\ll T$. Substituting Eq. (5) into Eq. (2) one obtains

$$T_{1\text{dph}}^{-1}=a_1^2\frac{T}{2^{3/2}\pi^2D^{3/2}}\frac{1}{\Omega_L}[((m\Omega_L^2)^2+(\gamma_2\Omega_L)^2)^{1/2}+m\Omega_L^2]^{1/2} \tag{6}$$

the inertial term becomes important, naturally, when

$$\Gamma_2\equiv\gamma_2/2m<\Omega_L. \tag{6a}$$

At low enough temperatures when this condition is satisfied

$$T_{1\text{dph}}^{-1}=a_1^2\frac{Tm^{1/2}}{2\pi^2D^{3/2}}, \tag{7}$$

i.e., the spin-lattice relaxation rate does not depend on Ω_L. Let us emphasize that the contribution of the phason direct processes to the spin-lattice relaxation rate [formula (7)] is quite different from the contribution of the ordinary acoustic phonons direct processes.[15] In the latter case the formula for the spin-lattice relaxation rate contains, comparing with Eq. (7), a very small factor, $(\Omega_L/\omega_D)^2$, where ω_D is the Debye frequency, by the order of magnitude.[15] The reason is that the spin-Hamiltonian depends directly on the transverse (phason) "displacement" [see Eq. (3)] unlike to the case of acoustic phonons where the coefficients of the spin Hamiltonian depend on the strain tensor components, i.e., on the derivatives of the displacements. Therefore, the "background" due to ordinary phonons can be completely neglected comparing with the phason governed contribution.

Now we estimate the spin-lattice relaxation time due to the Raman processes. For the low-T region where the phason damping can be neglected it is the most straightforward to use the standard formula (see, e.g., Ref. 15) rewritten in our terms, i.e., taking into account, once more, that the parameters of the spin Hamiltonian depend on the "phason displacement" η_2. In the form where the classical limit is easily obtainable one has

$$T_{1\text{Rph}}^{-1}=a_2^2\int_0^\infty d\omega\,\frac{(\hbar\omega)^2\gamma_2^2}{2\pi^2}\sinh^{-2}\frac{\hbar\omega}{2T}\times\left[\sum_k\frac{1}{(Dk^2-m\omega^2)^2+\gamma_2^2\omega^2}\right]^2, \tag{8}$$

where the limit $\gamma_2\to0$ will be taken in the final result. One obtains

$$T_{1\text{Rph}}^{-1}=a_2^2\frac{mT^3}{\pi^4\hbar D^3}\int_0^{\hbar\omega_{\max}/T}\frac{y^2dy}{\sinh^2y} \tag{9}$$

where $\omega_{\max}^2=Dk_{\max}^2/m$, and $k_{\max}$ is the cutoff of the problem. For $T\ll T_D$ the upper limit of the integral can be replaced by ∞ and one obtains

$$T_{1\text{Rph}}^{-1}=a_2^2\frac{mT^3}{16\pi^4\hbar D^3}. \tag{10}$$

Recall that the contribution of the Raman processes with participation of ordinary acoustic phonons is proportional to T at $T \ll T_D$ (see, e.g., Ref. 15), i.e., the phason-mediated Raman processes are far more effective. However, as we shall show now, they are still far less effective than the direct processes due to phason.

Let us take into account that if $\eta_2 = \eta_{2\,\max} \approx d$, where d is the interatomic distance, all the terms in Eq. (3) are of the same order of magnitude, i.e., one can set $a_1 \approx a_2 d$. Now we can compare the contribution due to the Raman processes [Eq. (10)] and that due the direct ones in the low-T region [Eq. (7)]. One has

$$\frac{T_{1\mathrm{Rph}}^{-1}}{T_{1\mathrm{dph}}} = \frac{a_2^2}{a_1^2} \frac{T^2 m^{1/2}}{8\pi^2 \hbar D^{3/2}} \approx \frac{1}{4\pi} \frac{T}{T_{\mathrm{Dph}}} \frac{T}{T_{\mathrm{at}}}, \tag{11}$$

where $T_{\mathrm{Dph}} = \hbar\omega_{\mathrm{Dph}} = \hbar (D/m)^{1/2} 2\pi/d$ is of the order of magnitude of the Debye temperature, and T_{at} is the ''atomic temperature'' ($10^4 - 10^5$ K). It has been taken into account that if one assumes $(D/m)^{1/2}$, i.e., the phason velocity, c, to be of order of magnitude of the sound velocity and the phason mass density of the order of magnitude of ordinary mass density one finds that $D \approx T_{\mathrm{at}}$ by the order of magnitude. One sees from Eq. (12) that the Raman processes can be neglected and to find the T-dependence of the phason contribution to T_1^{-1} it is quite enough to restrict ourselves by the direct processes.

At high temperatures, instead of Eq. (7) one obtains, of course, the well known formula[14]

$$T_{1\mathrm{dph}}^{-1} = \frac{a_1^2 T \gamma_2^{1/2}}{2^{3/2} \pi^2 D^{3/2} \Omega_L^{1/2}}. \tag{12}$$

It is convenient to introduce an (albeit ill-defined) ''boundary'' temperature T_b which separates the two temperature regions and is determined by the condition

$$\Gamma_2(T_b) \approx \Omega_L. \tag{13}$$

To reveal the real meaning of this condition, the T-dependence of the ''viscosity coefficient'' γ_2, is the topic of the next section.

III. TEMPERATURE DEPENDENCE OF THE PHASON DAMPING CONSTANT

General features of the phonon loss calculation at low T are well discussed elsewhere.[16] Due to the limitation chosen ($\hbar\Omega_L \ll T$) we are interested in the $\omega \to 0$ limit of the viscosity coefficient, i.e., only the part of loss that is proportional to ω is of interest for us. This part is due to the so-called association processes: the phonon in question and a thermal one convert into two other phonons. As at low T the thermal phonons are the acoustic ones and due to the energy conservation only acoustic phonons are created, it is enough to take into account the interaction of the vibration in question with the acoustic phonons only. The phason branch in the IC crystals is an additional acoustic branch, therefore, one can expect that the phason-phason and the phason-acoustic interaction is of interest for calculation of the ''phason viscosity.'' However, the phason branch is acoustic as to its dispersion law but is optic as to its coupling with other branches. This difference results in much more importance of the phason-phason interaction than the phason-acoustic one. In fact, the amplitudon branch is also acoustic-like due to the amplitudon-phason interaction.[17,18] Thus it is not so surprising that it is impossible to neglect the amplitudons, as we shall see from what follows.

We shall use the continuous medium approximation which is especially relevant for the low-T region. To write down the potential energy it is convenient to start with the ''effective Hamiltonian'' used in the fluctuation theory of phase transitions (see, e.g., Ref. 19). In our case the density of the effective Hamiltonian, or, rather, the density of the effective potential energy, as a function of the order parameter introduced in Sec. II, is

$$u = \frac{A}{2}(\eta_1^2 + \eta_2^2) + \frac{B}{4}(\eta_1^2 + \eta_2^2)^2 + \frac{D}{2}[(\nabla\eta_1)^2 + (\nabla\eta_2)^2]. \tag{14}$$

As in Sec. II we assume that the equilibrium value of $\eta_1(\eta_{1e})$ is nonzero while $\eta_{2e} = 0$, thus associating $\eta_1 = \eta_1 - \eta_{1e}$ with amplitudons and η_2 with phasons. When the fluctuations are neglected $\eta_{1e}^2 = -A/B$ and one has

$$u = u_e + \frac{(2B\eta_{1e}^2)\eta_1'^2}{2} + \frac{D}{2}[(\nabla\eta_1')^2 + (\nabla\eta_2)^2] + B\eta_{1e}\eta_1'\eta_2^2 + B\eta_{1e}\eta_1'^3 + \frac{B}{2}\eta_1'^2\eta_2^2 + \frac{B}{4}\eta_1'^4 + \frac{B}{4}\eta_2^4. \tag{15}$$

This form is more convenient than that of Eq. (14): one can consider η_{1e} as the exact (and unknown) value to be found in every approximation by minimizing the free energy of the system calculated to the same approximation. The density of the kinetic energy we shall assume to be as usual

$$\frac{1}{2m}(p_1^2 + p_2^2), \tag{16}$$

where $p_1(\mathbf{r}), p_2(\mathbf{r})$ are momenta corresponding to $\eta_1(\mathbf{r})$, $\eta_2(\mathbf{r})$, and m is the ''optical'' mass introduced already in Sec. II.

To demonstrate the problems that arise when calculating the phason damping constant let us begin with the classical limit and an approximation that might seem to be reasonable at least far from the normal-IC phase transition. Having in mind that the amplitudon is more ''hard'' than the phason which is quite analogous to the soft mode at a second order phase transition point we shall omit all the terms containing η_1' in Eq. (15). Then one has as the equation of motion for η_2:

$$m\ddot{\eta}_2 - D\nabla^2\eta_2 + B\eta_2^3 = 0. \tag{17}$$

Apart from a renormalization of the stiffness constant, which is not important being compensated by the change of η_{1e}, the anharmonicity described by the last term in the rhs of Eq. (15) provides a damping which is infinite. Indeed, Eq. (17) is that for an ordinary soft mode at the phase transition temperature (the mentioned compensation being taken into account) and the soft mode damping at the phase transition temperature is known to be infinite (see, e.g., Ref. 20).

In fact, it is not the case. The matter is that there are other relevant anharmonicities. Indeed, the fourth term in the rhs of Eq. (15) describes changes of the amplitudon coordinate due to the phason fluctuations as well as changes in the phason fluctuations due to changes in the amplitudon coordinates. In other words it describes the indirect (via amplitudon) interaction of the phason fluctuations. That means that to obtain the lower order result for the phason damping at low T one has to use the following potential energy density:

$$u=u_e+\frac{2B\eta_{1e}^2}{2}\,\eta_1'^2+\frac{D}{2}\,[(\nabla\eta_1')^2+(\nabla\eta_2)^2]+B\eta_{1e}\eta_1'\eta_2^2+\frac{B}{4}\,\eta_2^4. \tag{18}$$

The calculation can be made using the Matsubara technique of the temperature Green functions (see, e.g., Ref. 21). More details about the calculations are to be found in Appendix A. We would like to mention that all the perturbation theory terms are divergent but their sum is finite. Such a situation is typical for IC phases (see, e.g., Refs. 17 and 22).

For the low-frequency phason damping constant ($\hbar\Omega \ll T$) one obtains

$$\Gamma_2(q\approx 0,\Omega\approx 0)=\frac{\pi\hbar^3}{m^4T}\int\int\frac{d\mathbf{k}_1 d\mathbf{k}_2}{(2\pi)^6}\,\frac{B^2\left[\dfrac{D(\mathbf{k}_1+\mathbf{k}_2)^2-D(k_1+k_2)^2}{2B\eta_{1e}^2+D(\mathbf{k}_1+\mathbf{k}_2)^2-D(k_1+k_2)^2}\right]^2}{8\omega_2(k_1)\omega_2(k_2)\omega_2(\mathbf{k}_1+\mathbf{k}_2)}\,\frac{n_2(k_1)n_2(k_2)}{1-\exp\left(-\dfrac{\hbar[\omega_2(k_1)+\omega_2(k_2)]}{T}\right)}$$
$$\times\delta[\omega_2(k_1)+\omega_2(k_2)-\omega_2(\mathbf{k}_1+\mathbf{k}_2)], \tag{19}$$

where $\omega_2^2(\mathbf{k})=m^{-1}Dk^2$ and $n_2(k)=[\exp(\hbar\omega_2(\mathbf{k})T^{-1}-1]^{-1}$.

A rough estimation of Γ_2 can be made taking into account that the main contribution to the integral comes from the region $\hbar\omega_2(\mathbf{k})\sim T$, or $\omega_2\sim\omega_T$, $k\sim k_T$. As a result one finds

$$\Gamma_2(\Omega\approx 0,q\approx 0)\sim 10^{-2}\,\frac{\hbar^3k_T^{10}B^2D^2}{m^4T\omega_T^4|A|^2}\sim 10^{-2}\omega_T\,\frac{T^2}{|A|^2}\,k_T^2\,\frac{B^2}{D^2}, \tag{20}$$

where $|A|=B\eta_{1e}^2$. To estimate $|A|,B,D$ for displacive system one can follow Vaks[23] to obtain

$$|A(T\approx 0)|\sim T_cd^{-5},\quad B\sim T_{\rm at}d^{-7},\quad D\sim T_{\rm at}d^{-3}, \tag{21}$$

where T_c is the temperature of phase transition from the normal to the IC phase, $T_{\rm at}$ is a typical ''atomic temperature'' ($T_{\rm at}\sim 10^4$–10^5 K), d is the lattice constant, and the order parameter is considered as an atomic displacement. Assuming that the ''phason velocity'' $c=(D/m)^{1/2}$ is of the same order of magnitude as the sound one we find that $k_T=d^{-1}(T/T_D)$, where T_D is the Debye temperature. For ω_T one has, naturally, $\omega_T\sim\Omega_D(T/T_D)$, where Ω_D is the Debye frequency. As a result one finds

$$\Gamma_2\sim(10^{-1}-10^{-2})\Omega_D\left(\frac{T}{T_D}\right)^5, \tag{22}$$

where it has been assumed that $T_c\sim T_D$. To compare this result with that for an ''ordinary'' optical phonon let us recall, e.g., that the low-frequency damping constant for a polar optical phonon (such a constant determines the low-frequency dielectric losses) is proportional to T^7 at $T\ll T_D$ if the crystal is nonpolar,[24] i.e., at low enough temperatures the phason damping constant is much more than those of ''ordinary'' optical phonons.

Using Eqs. (13) and (22) one finds an estimation for the ''boundary temperature'' T_b:

$$T_b\sim T_D\left(\frac{\Omega_L}{10^{-2}\Omega_D}\right)^{1/5}. \tag{23}$$

According to Eq. (23) T_b can be one order of magnitude lower than T_D. One sees from Eq. (23) that T_b depends very weakly on the Larmor frequency albeit it does not seem impossible to detect this dependence. A careful experimental study of temperature dependence of T_1^{-1} at low temperatures and for different Larmor frequencies seems to be of interest.

IV. THE AMPLITUDON MEDIATED SPIN-LATTICE RELAXATION TIME

On the same condition as in Sec. II ($\hbar\Omega_L\ll T$) one has for the amplitudon contribution to the spin-lattice relaxation rate[14]

$$T_{1\rm dam}^{-1}=Ab_1^2\sum_{\mathbf{k}}\,\langle|\eta_1(\mathbf{k},\Omega_L)|^2\rangle, \tag{24}$$

$$\langle|\eta_1(\mathbf{k},\Omega_L)|^2\rangle=\frac{T}{\pi\Omega_L}\,{\rm Im}\,\chi_1(\mathbf{k},\Omega_L), \tag{25}$$

where $\chi_1(\mathbf{k},\Omega_L)$ is the longitudinal (amplitudon) response function. To the zero approximation one can write

$$\chi_1(\mathbf{k},\Omega_L)=\chi_{10}(\mathbf{k},\Omega_L)=\frac{1}{V(-m\Omega_L^2+2B\eta_{1e}^2+Dk^2-i\gamma_1\Omega_L)}=\frac{1}{Vm[\omega_1^2(\mathbf{k})-\Omega_L^2-2i\Gamma_1\Omega_L]}. \tag{26}$$

Here $\omega_1^2(\mathbf{k})=(2B\eta_{1e}^2+Dk^2)/m$, $\Gamma_1=\gamma_1/2m$; γ_1 is due to all the anharmonic interactions excluding those with the participation of the amplitudon and the phasons only.

Now we shall study just the effect of interaction between amplitudon and phasons. Specifically, we shall calculate the longitudinal response function $\chi_1(\Omega_L,\mathbf{k})$ taking into account the phase fluctuations.

The response function in question is the same as the retarded Green function as defined in Appendix A. The calculations of the amplitudon Green function for low temperature are to be found in Appendix B. One can write

$$\chi_1(\mathbf{q},\Omega)=\frac{1}{Vm[\omega_1^2(\mathbf{q})-\Omega^2-2i\Gamma_1\Omega]}+\frac{2T(B\eta_{1e})^2}{Vm^4[\omega_1^2(\mathbf{q})-\Omega^2-2i\Gamma_1\Omega]^2}\Phi(\mathbf{q},\Omega),\tag{27}$$

where $\Phi(\mathbf{q},\Omega)$ is found in Appendix B.

Using Eqs. (24) and (25) one can calculate now the amplitudon governed spin-lattice relaxation rate provided that the function $\chi_1(\Omega_L,\mathbf{k})$ is known. To simplify the discussion we shall first assume that $\Gamma_1\approx 0$ and then discuss the limits of this assumption. Within the limits of its validity the spin-lattice relaxation rate is determined by the imaginary part of $\Phi(\mathbf{q},\Omega)$. It is shown in Appendix B that for $\Omega\ll\omega_T$ the value of Im $\Phi(\mathbf{q},\Omega)$ is small for $q>k_T$ so that one can integrate up to $k=k_T$ in Eq. (24). Using Eqs. (24), (25), (27), and (B8) one finds the estimation

$$T_{1\text{dam}}^{-1}\sim b_1^2\,\frac{mT^3}{16\pi^4\hbar D^3\,\eta_{1e}^2(T\approx 0)}.\tag{28}$$

We shall see below that it is, in fact, the main part of the amplitudon-governed spin-lattice relaxation rate at low temperatures. Taking into account[23] that for displacive systems

$$\eta_{1e}^2(T0)\sim d^2\,\frac{T_i}{T_{\text{at}}}.\tag{29}$$

one obtains the estimation

$$T_{1\text{dam}}^{-1}\sim b_2^2\,\frac{mT^3}{16\pi^4\hbar D^3}\,\frac{T_{\text{at}}}{T_i},\tag{30}$$

where $b_2\approx b_1/d$. We can now compare this formula with Eqs. (7) and (10). Assuming reasonably that b_2 is of the same order of magnitude as a_2 one sees that the amplitudon-mediated spin-lattice relaxation rate is much smaller than the phason-mediated one, the ratio is about $(T/T_D)^2$ assuming that $T_i\approx T_D$, but it is more that the phason-mediated spin-lattice relaxation time is due to the Raman processes, the ratio being of the order of T_{at}/T_i.

Let us now discuss the temperature dependence of Γ_1. This problem is more difficult than in the case of phason. The matter is that the amplitudon vibrations are accompanied by the vibrations of temperature, unlike the phason ones. It is quite natural because the amplitudon is a fully symmetrical vibration and in this aspect is similar to the longitudinal sound wave. However, for the frequency region $\omega>\tau_N^{-1}$, where τ_N is the relaxation time for normal processes (see, e.g., Refs. 16 and 25), the only important mechanism of the longitudinal sound attenuation is that of Landau-Rumer[26] when the quantum many-phonon processes are considered (for the longitudinal sound the three-phonon ones prove to be the most important). In this frequency region the concept of temperature cannot be used: the thermalization time is less that the period of the vibrations.

The estimation for τ_N reads[16]

$$\tau_N^{-1}\sim\frac{T^5}{\rho\hbar^4c^5}.\tag{31}$$

This gives for $T\approx 1$ K the value of τ_N^{-1} about 10^2-10^3 s^{-1} and for $T\approx 10$ K the value about 10^7-10^8 s^{-1}. Therefore it seems reasonable to use the Landau-Rumer mechanism to estimate Γ_1 having in mind the calculations of the spin-lattice relaxation rate in ordinary conditions when Ω_L is about 10^7-10^8 s^{-1}.

The most effective three-phonon interaction corresponds to the term which, in Eq. (15), would have the form $r\eta_{1e}\eta_1'\eta_2^2u_{ii}$, where r is a coefficient and u_{ij} is the dilatation [in Eq. (14) the corresponding term would be $(r/2)(\eta_1^2+\eta_2^2)^2u_{ii}$]. A calculation similar to that of Appendix A shows that $\Gamma_1\propto T^7$. The same temperature dependence, but with a smaller coefficient, provides as well the coupling of the amplitudon with the acoustic phonons due to to the coupling term $\eta_{1e}\eta_1'u_{ii}^2$. One sees that the corrections to spin-lattice relaxation rate due to the finite value of Γ_1 are proportional at least to T^8 and can be neglected, at low temperatures, comparing with the contribution given by Eq. (30).

V. CONCLUSIONS

We have shown that, in the low-T region, the T-dependence of the spin lattice relaxation rates in IC phases is quite different from that in ordinary crystal phases. First, the phason contribution is proportional to T and does not depend on the Larmor frequency Ω_L while the linear in T term due to the direct processes with acoustic phonons is proportional to Ω_L^2. Second, the amplitudon, which is an optical mode to the harmonic approximation, was found to provide a contribution to the spin-lattice relaxation rate which is smaller than the phason one but is much larger than any contribution of the acoustic phonons (at $T>\hbar\Omega_L$).

The treatment of the amplitudon contribution was referred to high enough Ω_L. This is because we have not made calculations of the "bare" amplitudon damping coefficient $\Gamma_1(\mathbf{q},\Omega_L)$ for small Ω_L but large q $(q>k_T)$ which is necessary to calculate the low-frequency spin-lattice relaxation time. Such a $(\mathbf{q},\Omega_L)$ region corresponds to a nonhydrodynamic regime and should be treated separately. Thus it is possible, in principle, that the conclusions of the present paper concerning the amplitudon contribution to the spin-lattice relaxation rate are not valid for small Ω_L (e.g., for that characteristic for some NQR experiments). Still it is not very probable taking into account that for higher frequencies the role of Γ_1 has been found negligible.

APPENDIX A: THE PHASON DAMPING CONSTANT

Considering $\eta_1(\mathbf{r},t)$, $\eta_2(\mathbf{r},t)$ as quantum operators one has for the amplitudon or phason retarded Green functions

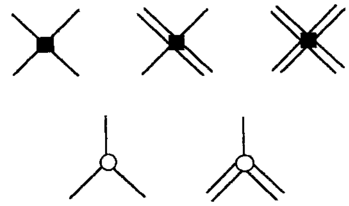

FIG. 1. Vertices of anharmonic part of $H_{\rm eff}$: ■, fourth-order vertex B; ○, third-order vertex $B\eta_{1e}$; —, Green function $G_1(\omega_n,\mathbf{k})$; ═, Green function $G_2(\omega_n,\mathbf{k})$.

$$G_j^R(\mathbf{r},t)=\frac{i}{\hbar}\,\theta(t)\langle\,\eta_j(\mathbf{r},t)\,\eta_j^+(0,0)-\eta_j^+(0,0)\,\eta_j(\mathbf{r},t)\rangle, \tag{A1}$$

where $j=1,2$; $\theta(t)=1$ for $t>0$ and $\theta(t)=0$ for $t<0$; the $\langle\ \rangle$ is thermodynamic average. The Fourier transform of these functions can be written as

$$G_j^R(\mathbf{q},\Omega)=\frac{1}{Vm[\,\omega_j^2(\mathbf{q})-\Omega^2+\Sigma_j(\mathbf{q},\Omega)\,]}=\frac{1}{Vm[\,\widetilde{\omega}_j^2(\mathbf{q})-\Omega^2-2i\Omega\Gamma_j(\mathbf{q},\Omega)\,]}, \tag{A2a}$$

where $\omega_j(\mathbf{q})$ is the dispersion law in the harmonic approximation, $\omega_1^2(\mathbf{q})=m^{-1}(2B\eta_{1e}^2+Dq^2)$, $\omega_2^2(\mathbf{q})=m^{-1}Dq^2=c^2q^2$; $\widetilde{\omega}_j(\mathbf{q})$ is the renormalized dispersion law; $\Sigma_j(\mathbf{q},\Omega)$ is the self-energy and $\Gamma_j(\mathbf{q},\Omega)$ is the damping constant, i.e., $\Gamma_j(\mathbf{q},\Omega)=-(2\Omega)^{-1}\,\mathrm{Im}\,\Sigma_j(\mathbf{q},\Omega)$. The Green functions (A2a) are obtainable from corresponding thermal Green functions[21,25] $G_j(\mathbf{q},\Omega_n)$, where $\Omega_n=2\pi Tn$, $n=0,\pm1,\ldots$,

$$G_j^R(\mathbf{q},\Omega)=\frac{1}{T}\,G_j[\mathbf{q},-i(\Omega+i\delta)\hbar], \tag{A3}$$

where $\delta\to+0$, i.e., in the upper half of the complex Ω plane, where the retarded Green function $G_j^R(\mathbf{q},\Omega)$ is analytical, $T^{-1}G_j(\mathbf{q},\Omega_n)$ and $G_j^R(\mathbf{q},\Omega)$ coincide at $\Omega=i\Omega_n/\hbar$. Similarly to Eq. (A2a) one has

$$G_j(\mathbf{q},\Omega_n)=\frac{T}{Vm}\,\frac{1}{\omega_j^2(\mathbf{q})+\dfrac{\Omega_n^2}{\hbar^2}+\Sigma_j(\mathbf{q},\Omega_n)}. \tag{A2b}$$

The diagram technique for the thermal Green functions is standard.[22,25] It is seen from Eq. (15) that the anharmonic part of the Hamiltonian contains four different vortices (Fig. 1).

To calculate the phason damping constant we find first the phason self-energy. The contribution of the lowest-order diagrams is of no interest for us. The diagram containing one fourth-order vortex contributes to the real part of the self-energy only. The diagram with two third-order vertices contributes both to the real part and to the imaginary part but the last contribution is exponentially small at low T. Important lowest-order diagrams for the self-energy are represented in Fig. 2; their sum is a diagram with renormalized phason-phason vertex (Fig. 3). One finds

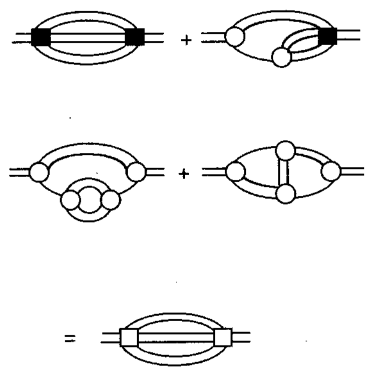

FIG. 2. Compensation of divergent diagrams of $G_2(\omega_n,\mathbf{k})$. The second order of the perturbation theory.

$$\Sigma_2(\mathbf{q},\Omega_n)=-\frac{T}{Vm}\sum_{\substack{\mathbf{k}_1,\mathbf{k}_2\\ m,1}}G_{2,0}(\mathbf{k}_1,\omega_m)G_{2,0}(\mathbf{k}_2,\omega_l)\times G_{2,0}(\mathbf{q}-\mathbf{k}_1-\mathbf{k}_2,\Omega_n-\Omega_m-\omega_l)\left[\left(\frac{V}{T}\right)^2\times 6\widetilde{B}^2(\mathbf{k}_1+\mathbf{k}_2,\mathbf{k}_1,\omega_m+\omega_l,\omega_m,\mathbf{q},\Omega_n)\right] \tag{A4}$$

where

$$\begin{aligned}\widetilde{B}^2(\mathbf{k}_1+\mathbf{k}_2,\mathbf{k}_1,\omega_m+\omega_l,\omega_m,\mathbf{q},\Omega_n)&=B^2-\frac{V}{T}4B^3\eta_{1e}^2G_{1,0}(\mathbf{k}_1+\mathbf{k}_2,\omega_m+\omega_l)\\&+\left(\frac{V}{T}\right)^2\frac{4}{3}B^4\eta_{1e}^4G_{1,0}^2(\mathbf{k}_1+\mathbf{k}_2,\omega_m+\omega_l)\\&+\left(\frac{V}{T}\right)^2\frac{8}{3}B^4\eta_{1e}^4G_{1,0}(\mathbf{k}_1+\mathbf{k}_2,\omega_m+\omega_l)\\&\times G_{1,0}(\mathbf{q}-\mathbf{k}_1,\Omega_n-\omega_m)\end{aligned} \tag{A5}$$

and

$$G_{j,0}(\mathbf{k},\omega_n)=\frac{T}{Vm}\,\frac{1}{\omega_j^2(\mathbf{k})+\dfrac{\omega_n^2}{\hbar^2}}. \tag{A6}$$

The summation over m,l can be made with the help of the formula[25]

$$\sum_{m=-\infty}^{m=\infty}f(i\omega_m)=\frac{\hbar}{2\pi Ti}\int_C f(z)n(z)dz, \tag{A7}$$

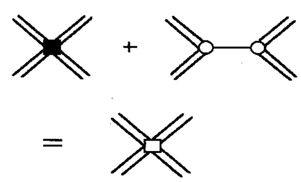

FIG. 3. Renormalization of the phason-phason vertex.

where $n(z)=[\exp(\hbar z/T)-1]^{-1}$ and the contour C is taken around the singularities of the function $f(z)$. The integral is evaluated by the usual residue theorem. As the poles of the integrand are those of the Green functions $G_j(\mathbf{k},i\omega_n=z)$ and function $n(z)$ at the poles is the mean occupation of the mode the contribution of the poles of $\widetilde{B}$ can be neglected. Indeed, it is seen from Eq. (A5) that they are due to the poles of the harmonic amplitudon Green function $G_{1,0}(\mathbf{k},z)$, i.e., at $z=\omega_1(\mathbf{k})$ and for the temperatures under consideration $\hbar\omega_1(\mathbf{k})\ll T$ and $n[\omega_1(\mathbf{k})]$ is close to zero. Still the formula for $\Sigma_2(\mathbf{q},\omega_n)$ is fairly clumsy and we shall not write it down.

The next step is to transform this formula to a continuous variable such that $f(\omega)=f(i\omega_n)$ when $\omega=i\omega_n$. This is achieved by writing $i\omega_n/\hbar\rightarrow\omega+i\delta$ where $\delta\rightarrow+0$. Then one takes into account that

$$\lim_{\delta-0}\frac{1}{x-i\delta}-\frac{1}{(x)_p}+i\pi\delta(x), \tag{A8}$$

where the subscript p indicates the principal part, to obtain for the imaginary part of $\Sigma_2(\mathbf{q},\Omega)$

$$-\operatorname{Im}\Sigma_2(\mathbf{q},\Omega)=\frac{\pi\hbar^2}{m^4}\int\int\frac{d\mathbf{k}_1 d\mathbf{k}_2}{(2\pi)^6}\frac{9[n_2(k_1)n_2(k_2)-n_2(|\mathbf{q}-\mathbf{k}_1-\mathbf{k}_2|)(1+n_2(k_1)+n_2(k_2)]}{4\omega_2(\mathbf{k}_1)\omega_2(\mathbf{k}_2)\omega_2(\mathbf{q}-\mathbf{k}_1-\mathbf{k}_2)}\widetilde{B}^2[\mathbf{k}_1+\mathbf{k}_2,\mathbf{k}_1,\omega_2(\mathbf{k}_1)$$
$$+\omega_2(\mathbf{k}_2),\omega(\mathbf{k}_1),\mathbf{q},\Omega]\delta[\omega_2(\mathbf{k}_1)+\omega_2(\mathbf{k}_2)-\omega_2(\mathbf{q}-\mathbf{k}_1-\mathbf{k}_2)-\Omega], \tag{A9}$$

where $n_2(k)=n[\omega_2(\mathbf{k})]$. Here only the phonon association processes[16] have been taken into account because we are interested in the low-frequency damping coefficient and the contribution of the neglected decay processes to this coefficient is zero at $\omega\rightarrow 0$. Setting $\mathbf{q}=0$ and taking into account that because of the δ function

$$n_2(|-\mathbf{k}_1-\mathbf{k}_2|)=\exp\left(\frac{\hbar(\Omega-\omega_2(\mathbf{k}_1)-\omega_2(\mathbf{k}_2)}{T}-1\right)^{-1}, \tag{A10}$$

one can show that

$$n_2(k_1)n_2(k_2)-n_2(|-\mathbf{k}_1-\mathbf{k}_2|)[1+n_2(k_1)+n_2(k_2)]=n_2(k_1)n_2(k_2)\frac{\exp\left(\frac{\hbar\Omega}{T}-1\right)}{1-\exp\left(-\frac{\hbar[\omega_2(\mathbf{k}_1)+\omega_2(\mathbf{k}_2)]}{T}\right)}. \tag{A11}$$

Then, for $\hbar\Omega\ll T$, we obtain Eq. (19) from Eq. (A9).

APPENDIX B: THE AMPLITUDON RESPONSE FUNCTION

The amplitudon response function is the same as its retarded Green function as defined by Eqs. (A1), (A2a), and (A2b). The method to calculate this Green function is, in principle, the same as for the case of phason (Appendix A). However, it is more convenient now to develop the perturbation expansion not for $\Sigma_1(\mathbf{q},\omega_n)$ but for the Green function itself: it is a specific feature of IC systems that a compensation of some divergent higher order diagrams takes places as it has been commented for the Heisenberg magnet[17] and this compensation is revealed more easily in the perturbation expansion of the Green (response) function.

The leading correction to the zeroth order Green function (A6) is

$$G_1(\mathbf{q},\Omega_n)=G_{1,0}(\mathbf{q},\Omega_n)+(2B\eta_{1e})^2G_{1,0}^2(\mathbf{q},\Omega_n)\left(\frac{V}{T}\right)^2\sum_{\mathbf{k},\omega_m}G_{2,0}(\mathbf{k},\omega_m)G_{2,0}(\mathbf{q}-\mathbf{k},\Omega_n-\omega_m). \tag{B1}$$

Performing the summation over ω_m (see Appendix A) and transforming the formula to the continious variable $\Omega[\Omega_n\rightarrow -i(\Omega+i\delta)\hbar]$ we find

$$G_1^R(\mathbf{q},\Omega)=\frac{1}{Vm[\omega_1^2(\mathbf{q})-\Omega^2-2i\Omega\Gamma_1]}-\frac{2T(B\eta_{1e})^2}{Vm^4[\omega_1^2(\mathbf{q})-\Omega^2-2i\Omega\Gamma_1]^2}\Phi(\mathbf{q},\Omega), \tag{B2}$$

where Γ_1 is the ''bare'' amplitudon damping constant (see Sec. IV) and

$$\mathrm{Re}\ \Phi(\mathbf{q},\Omega)=\frac{\hbar}{T(2\pi)^3}\int\frac{d\mathbf{k}}{4\omega_2(\mathbf{k})\omega_2(\mathbf{q}-\mathbf{k})}\left[[n(\omega_2(\mathbf{q}-\mathbf{k}))-n(\omega_2(\mathbf{k}))]\left(\frac{1}{\omega_2(\mathbf{k})-\omega_2(\mathbf{q}-\mathbf{k})-\Omega}+\frac{1}{\omega_2(\mathbf{k})-\omega_2(\mathbf{q}-\mathbf{k})+\Omega}\right)\right.$$
$$\left.+[n(\omega_2(\mathbf{k}))+n(\omega_2(\mathbf{q}-\mathbf{k}))+1]\left(\frac{1}{\omega_2(\mathbf{k})-\omega_2(\mathbf{q}-\mathbf{k})-\Omega}-\frac{1}{\omega_2(\mathbf{k})-\omega_2(\mathbf{q}-\mathbf{k})+\Omega}\right)\right], \tag{B3a}$$

$$\mathrm{Im}\ \Phi(\mathbf{q})=\frac{\pi\hbar}{T(2\pi)^3}\int\frac{d\mathbf{k}[n(\omega_2(\mathbf{q}-\mathbf{k}))-n(\omega_2(\mathbf{k}))]}{4\omega_2(\mathbf{k})\omega_2(\mathbf{q}-\mathbf{k})}[\delta(\omega_2(\mathbf{k})-\omega_2(\mathbf{q}-\mathbf{k})-\Omega)-\delta(\omega_2(\mathbf{k})-\omega_2(\mathbf{q}-\mathbf{k})+\Omega)]. \tag{B3b}$$

Here only the phonon association processes are taken into account (cf. Appendix A) as we are interested in the low-frequency response function.

One sees that at $T=0$

$$T\ \mathrm{Re}\ \Phi(\mathbf{q},\Omega)=\frac{\pi\hbar}{(2\pi)^3}\int\frac{d\mathbf{k}}{4\omega_2(\mathbf{k})\omega_2(\mathbf{q}-\mathbf{k})}\left[\frac{1}{\omega_2(\mathbf{k})+\omega_2(\mathbf{q}-\mathbf{k})-\Omega}+\frac{1}{\omega_2(\mathbf{k})+\omega_2(\mathbf{q}-\mathbf{k})+\Omega}\right], \tag{B4}$$

i.e., the real part of the response function diverges logarithmically at $\mathbf{q}\to 0$, $\Omega\to 0$. In other words the quantum fluctuations of phase, as well as the classical ones, lead to a divergence of the static amplitudon response function but the quantum divergences are more weak than the classical ones.

Of more interest, within this paper, is the imaginary part of the response function. Let us calculate Im $\Phi(\mathbf{q},\Omega)$. It is easy to see that

$$n(\omega_2(\mathbf{q}-\mathbf{k}))-n(\omega_2(\mathbf{k}))=\frac{1}{2}\frac{\sinh\left[\frac{\hbar(\omega_2(\mathbf{k})-\omega_2(\mathbf{q}-\mathbf{k}))}{2T}\right]}{\sinh\left[\frac{\hbar\omega_2(\mathbf{k})}{2T}\right]\sinh\left[\frac{\hbar\omega_2(\mathbf{q}-\mathbf{k})}{2T}\right]}. \tag{B5}$$

Taking into account then that because of the δ functions in Eq. (B3b) one has $|\omega_2(\mathbf{k})-\omega_2(\mathbf{q}-\mathbf{k})|=\Omega$, and reminding ourselves that it is the low frequency region ($\Omega\ll\omega_T=T/\hbar$) which is under the study we arrive at the formula

$$\mathrm{Im}\ \Phi(\mathbf{q},\Omega\to 0)\approx\frac{\pi\Omega}{(2\pi)^3}\frac{\hbar^2}{T^2}\int\frac{d\mathbf{k}\delta[\omega_2(\mathbf{k})-\omega_2(\mathbf{q}-\mathbf{k})]}{8\omega_2^2(\mathbf{k})\sinh^2\left[\frac{\hbar\omega_2(\mathbf{k})}{2T}\right]}. \tag{B6}$$

One can see that the minimum vector $\mathbf{k}$ to contribute to the integral is of the length $q/2$ and the effective cutoff of the integral is about $T/\hbar c=k_T$. One sees as well that the integral is very small for $q>k_T$ and, therefore, the integral can be estimated for small $\mathbf{q}$. Introducing spherical polar coordinates with the z axis along the vector $\mathbf{q}$ one has (for $k>q/2$):

$$\omega_2(\mathbf{k})-\omega_2(\mathbf{k}-\mathbf{q})\approx-\frac{\partial\omega_2}{\partial\mathbf{k}}\mathbf{q}=-cq\cos\theta. \tag{B7}$$

We find that Eq. (B6) can be rewritten as

$$\mathrm{Im}\ \Phi(\mathbf{q},\Omega)\approx\frac{\Omega}{8\pi^2qc^5}\int_{q/2<k<k_T}\frac{\delta(\cos\theta)d\sin\theta k^2dk}{k^4}\approx\frac{\Omega}{4\pi^2q^2c^5}. \tag{B8}$$

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