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# *EFFECT OF ANHARMONICITY ON THE INTENSITY OF THE MÖSSBAUER LINE*

M. A. KRIVOGLAZ

Institute of Metal Physics, Academy of Sciences, Ukrainian SSR

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The effect of anharmonicity on the intensity of the Mössbauer line is treated for the case when local modes are present. Even a weak anharmonicity produces a qualitative change in the time dependence of the correlation function and results in the disappearance from the expression for the intensity of the product of Bessel functions A defined in formula (2). The corresponding intensity difference is distributed around the Mössbauer line over a band of width $\Gamma_\varkappa$, where $\Gamma_\varkappa$ is the level width of the local mode. The effect of anharmonicity on the Debye-Waller factor is discussed. Including anharmonicity changes, the dependence of the Mössbauer line intensity on the temperature and wave vector and, in particular, gives an anisotropy of the effect in cubic crystals. Similar effects can occur in solid solutions in the harmonic approximation. The effect of anharmonicity on the one-phonon spectrum is considered.

**1.** In ideal crystals the intensity of the Mössbauer line for no-phonon emission or absorption of photons is given by the familiar Debye-Waller factor:

$$e^{-2W} = |\langle \exp(i\mathbf{ku})\rangle|^2 \approx \exp[-\langle(\mathbf{ku})^2\rangle]. \qquad (1)$$

Here **k** is the photon wave vector, **u**, is the thermal displacement of the atom, $\langle : \ldots \rangle$ denotes a statistical average. In nonideal crystals, e.g., in the case of a Mössbauer effect on impurity nuclei, local modes may occur. The intensity of the Mössbauer line when local modes are present was considered in [1,2], where it was found to be given by the factor

$$e^{-2W}A = e^{-2W}\prod_\varkappa I_0\left(2W_\varkappa \operatorname{sh}^{-1}\frac{\hbar\omega_\varkappa}{2k_B T}\right). \qquad (2)^*$$

Here $\varkappa$ numbers the normal modes, $2W_\varkappa$ is the contribution to 2W from normal mode number $\varkappa$, $\omega_\varkappa$ is the frequency of this vibration, $k_B$ is the Boltzmann constant, T is the temperature, and $I_0$ is a Bessel function of zero order and imaginary argument; this factor differs from unity when $N \to \infty$ only in the terms corresponding to local modes. This factor also determines the intensity of elastic neutron scattering when local modes are present.[3]

Formula (2) and the approximate expression in formula (1) are derived in the harmonic approximation of the theory of lattice vibrations, and are rigorous within this approximation. However, as we shall show, including anharmonicity causes an essential change in the expression for the intensity of the Mössbauer line when local modes are present. Even for weak anharmonicity, when $\epsilon^2 \ll 1$ (the order of magnitude of $\epsilon^2$ is the ratio of the frequency shift due to anharmonicity to the frequency itself), the intensity of the Mössbauer line is changed not by an amount $\sim \epsilon^2$ but by a finite factor independent of $\epsilon^2$. The consequence of this change is that when the anharmonicity is taken into account the product of Bessel functions drops out of the factor giving the intensity of the Mössbauer line, the factor decreases by A, and, when local modes are present, is given not by (2) but by (1), just as for the case when there are no local modes. The difference in intensity of the Mössbauer line, calculated with and without anharmonicity, is proportional to $(A - 1)\exp(-2W)$, and is distributed around the Mössbauer line within a band of width $\sim\Gamma_\varkappa$ ($\Gamma_\varkappa$ is the level width for the local mode) which is much greater than the width of the Mössbauer line but much less than the widths of the spectral distributions corresponding to one- or multiphonon processes.

Inclusion of the anharmonicity also changes the temperature dependence of the mean square atomic displacement and gives rise to a more complicated dependence of W on **k** than in the last approximate expression (1). As a result, when anharmonicity is included (and also in the harmonic approximation for the case of solid solutions) the intensity of the Mössbauer line is anisotropic even in cubic crystals. These effects,

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*sh = sinh.

as well as the peculiarities of the spectrum of nucleus-phonon emission that are caused by the anharmonicity, will be discussed.

2. We shall start from the formulas of [4,5], which express the (normalized) probability $J(\omega)$ for emission of photons of frequency $\omega$ in terms of the correlation function

$$J(\omega)=\frac{1}{2\pi}\int\limits_{-\infty}^{\infty}\exp\left[-i(\omega-\omega_0)t-\frac{1}{2}\Gamma|t|\right]g_s(\mathbf{k},t)\,dt. \tag{3}$$

Here $\hbar\omega_0$ is the difference in energy of the nuclear levels, and $g_s(\mathbf{k}, t)$ is the Fourier component of the autocorrelation function introduced by Van Hove [6] and defined by the expression

$$g_s(\mathbf{k},t)=\langle\exp[i\mathbf{ku}(t)]\exp[-i\mathbf{ku}(0)]\rangle, \tag{4}$$

where $\mathbf{u}(t)$ is the operator for the thermal displacements in the Heisenberg representation.

A simpler approximate expression for the function $g_s(\mathbf{k}, t)$ was proposed by Baym.[5] Assuming that displacements caused by forces involving the photon are proportional to those forces, it was shown in [5] that

$$g_s(\mathbf{k},t)=\exp\{-\langle(\mathbf{ku}(0))^2\rangle+\langle(\mathbf{ku}(t))(\mathbf{ku}(0))\rangle\}. \tag{5}$$

As we show in the Appendix, the difference between (4) and (5) is proportional to $\epsilon^2$ for all t. At first in carrying out the calculation we shall not consider terms $\sim\epsilon^2$ but shall include terms $\sim\epsilon^2/\omega_\kappa$. We can then use (5) for $g_s(\mathbf{k}, t)$ and the problem reduces to calculating the correlation function $\langle(\mathbf{k}\cdot\mathbf{u}(t))(\mathbf{k}\cdot\mathbf{u}(0))\rangle$. To carry out the computation we expand the displacements in the normal coordinates of the nonideal crystal (which in general does not give plane waves). The scalar product $\mathbf{k}\cdot\mathbf{u}$ can be written in the form

$$\mathbf{ku}=\sum_{\varkappa}(\alpha_\varkappa a_\varkappa+\overset{*}{\alpha}_\varkappa a_\varkappa^{+}), \tag{6}$$

where $a_\kappa^{+}$ and $a_\kappa$ are the phonon creation and annihilation operators (corresponding to the continuous or the local spectrum), and the $\alpha_\kappa$ are coefficients proportional to k, whose detailed form is unimportant for the sequel.

The computation of $\langle(\mathbf{k}\cdot\mathbf{u}(t))(\mathbf{k}\cdot\mathbf{u}(0))\rangle$ thus reduces to calculating the phonon correlation functions. These functions were determined in [7,8] in the course of investigating the energy distribution of inelastically scattered neutrons. It was found that the correlation functions for $\kappa\neq\kappa'$ and also $\langle a_\kappa(t)a_\kappa(0)\rangle$ and $\langle a_\kappa^{+}(t)a_\kappa^{+}(0)\rangle$ are of order $\epsilon^2$. Therefore

$$\langle(\mathbf{ku}(t))(\mathbf{ku}(0))\rangle$$
$$=\sum_{\varkappa}|\alpha_\varkappa|^2[\langle a_\varkappa^{+}(t)a_\varkappa(0)\rangle+\langle a_\varkappa(t)a_\varkappa^{+}(0)\rangle]. \tag{7}$$

In [7,8] they also determined the spectral representations $\varphi_\kappa'(\omega)$ and $\varphi_\kappa''(\omega)$ of the functions $\langle a_\kappa(t)a_\kappa^{+}(0)\rangle$ and $\langle a_\kappa^{+}(t)a_\kappa(0)\rangle$:

$$\langle a_\varkappa(t)a_\varkappa^{+}(0)\rangle=\int\limits_{-\infty}^{\infty}\varphi_\varkappa'(\omega)e^{i\omega t}\,dt;$$
$$\langle a_\varkappa^{+}(t)a_\varkappa(0)\rangle=\int\limits_{-\infty}^{\infty}\varphi_\varkappa''(\omega)e^{i\omega t}\,dt \tag{8}$$

and it was found that

$$\varphi_\varkappa'(-\omega)=\frac{1}{\pi}\frac{\Gamma_\varkappa(\omega)}{[\omega-\omega_\varkappa-P_\varkappa(\omega)]^2+\Gamma_\varkappa^2(\omega)}[n(\omega)+1],$$

$$\varphi_\varkappa''(\omega)=\frac{1}{\pi}\frac{\Gamma_\varkappa(\omega)}{[\omega-\omega_\varkappa-P_\varkappa(\omega)]^2+\Gamma_\varkappa^2(\omega)}n(\omega), \tag{9}$$

where $n(\omega)=[\exp(\hbar\omega/k_BT)-1]^{-1}$, $\Gamma_\kappa(\omega)$ and $P_\kappa(\omega)$ are the damping and shift of the phonon frequency due to anharmonicity (since the selected normal coordinates of the nonideal crystal are exact in the harmonic approximation, the broadening of these phonons corresponding to local modes or to distortion of waves by crystal defects is related only to the anharmonicity and not to static defects).

The damping $\Gamma_\kappa$ of the local mode was determined in [7] [cf. formula (37) in [7], in which, however, one must add a factor ½; in formula (37) for the frequency shift $P_\kappa$ of the local phonon, terms were omitted corresponding to virtual processes of creation or annihilation of three or two phonons; $P_\kappa$ and $\Gamma_\kappa$ are correctly defined by formulas (15) and (14) in [8] or by formula (A.18)]. In the case where processes of decay of a local phonon into two crystal phonons are possible, at high temperatures $\Gamma_\kappa$ and $P_\kappa$ are proportional to T, while for T = 0 they tend toward a nonzero limit. In order of magnitude, $\Gamma_\kappa$ and $P_\kappa$ may be $\sim(10^{-1}\text{–}10^{-2})\,\omega_\kappa$. In the case of weak anharmonicity, one can neglect the dependence of $\Gamma_\kappa$ and $P_\kappa$ on $\omega$ in the region $\omega\approx\pm(\omega_\kappa+P_\kappa)$ and regard $\Gamma_\kappa$ and $P_\kappa$ as constants. Carrying out the integration for this case in formulas (7), (8), we find from (3)—(8) the following integral representation for $J(\omega)$:

$$J(\omega)=\frac{1}{2\pi}\exp[-\langle(\mathbf{ku})^2\rangle]$$
$$\times\int\limits_{-\infty}^{\infty}\exp\left[-i(\omega-\omega_0)t-\frac{1}{2}\Gamma|t|\right]$$

$$\times \exp\Big\{\sum_{\varkappa} |\alpha_\varkappa|^2 [(\bar{n}_\varkappa + 1) \exp\{-i(\omega_\varkappa + P_\varkappa)t - \Gamma_\varkappa |t|\} + \bar{n}_\varkappa \exp\{i(\omega_\varkappa + P_\varkappa)t - \Gamma_\varkappa |t|\}]\Big\}\, dt. \quad (10)$$

Here $\bar{n}_\kappa = n(\omega_\kappa + P_\kappa)$.

The broadening $\Gamma_\kappa$ of the local phonon is much larger than the natural width of the Mössbauer line. For example, for $Fe^{57}$, $\hbar\Gamma = 4.5 \times 10^{-9}$ eV, whereas $\hbar\Gamma_\kappa$ is of order $10^{-2}$—$10^{-4}$ eV. Thus for frequencies $|\omega - \omega_0| \lesssim \Gamma$, one can neglect the contribution of the integration region $|t| \lesssim \Gamma_\kappa^{-1}$ in the integral (10) compared to that from the region $\Gamma_\kappa^{-1} \ll |t| \lesssim \Gamma^{-1}$. But for $|t| \gg \Gamma_\kappa^{-1}$ the quantity $\exp(-\Gamma_\kappa |t|)$ and the exponent of the last exponential in the integrand of (10) become exponentially small, and this exponential can be replaced by unity. Thus neglecting terms $\sim \Gamma/\Gamma_\kappa$ and $\sim \epsilon^2$, the intensity of the Mössbauer line is given, whether local modes are present or not, by the expression

$$J(\omega) = e^{-2W} \frac{1}{2\pi} \frac{\Gamma}{(\omega - \omega_0)^2 + \Gamma^2/4}. \quad (11)$$

Thus in accordance with our remarks the intensity of the Mössbauer line is actually given by the factor $e^{-2W}$ without the multiplier A which is a product of Bessel functions [it is easy to see that this factor appears immediately if we set $\Gamma_\kappa = 0$ in formula (10)]. To get a change of the line intensity by a finite factor (factor A) it is sufficient to include a very small anharmonicity. This amount is limited below only by the very mild requirement that $\Gamma_\kappa \gg \Gamma$, which is practically always satisfied. Essentially the role of the anharmonicity reduces simply to a change in the behavior of the autocorrelation function $g_s(\mathbf{k}, t)$ at long times. In the absence of local modes this function, even in the harmonic approximation, after a time $\sim \omega_m^{-1}$ (where $\omega_m$ is the maximum vibration frequency) tends toward the constant limiting value $g_s(\mathbf{k}, \infty)$.[6] If there are local modes present, then in the harmonic approximation at large t the function $g_s(\mathbf{k}, t)$ does not approach a constant limiting value but oscillates around it, which causes the appearance of the Bessel functions in formula (2).[2] But when anharmonicity is present, even for the case of crystals with local modes, $g_s(\mathbf{k}, t)$ attains the constant limiting value $g_s(\mathbf{k}, \infty)$. True this occurs after a time $\sim \omega_m^{-1}$ and not $\sim \omega_m^{-1}\epsilon^{-2}$. But just as when there are no local modes, the intensity of the Mössbauer line is determined by the limiting value $g_s(\mathbf{k}, \infty) = e^{-2W}$.

The last exponential factor in (10) does not affect the intensity of the Mössbauer line, but when local modes are present it results in the appearance of a peak of width $\sim\Gamma_\kappa$ around the line. Remembering that for not too weak Mössbauer lines $|\alpha_\kappa|^2 < 1$, the intensity of this peak can be found by expanding (10) in powers of $|\alpha_\kappa|^2$ and stopping at the quadratic terms ($\sim \alpha_\kappa^4$). Then in addition to the peaks with frequencies $\omega \approx \omega_0 \pm m(\omega_\kappa + P_\kappa)$ ($m = 1, 2$), corresponding to processes involving excitation or absorption of local phonons, which were treated in [1], there should also be a peak near the frequency $\omega = \omega_0$, whose intensity is given by the expression

$$J'(\omega) = e^{-2W} \frac{1}{2\pi} \sum_{\varkappa} |\alpha_\varkappa|^4 \bar{n}_\varkappa (\bar{n}_\varkappa + 1) \frac{4\Gamma_\varkappa}{(\omega - \omega_0)^2 + 4\Gamma_\varkappa^2}. \quad (12)$$

(If the local modes are degenerate, $|\alpha_\kappa|^2$ is given by a sum over the degenerate vibrations with frequency $\omega_\kappa$.) The width of this peak is actually $\sim\Gamma_\kappa$, and its integral intensity is $(A - 1)e^{-2W}$ [if we include the higher terms in the expansion in powers of $|\alpha_\kappa|^2$ in (12)].

3. So far we have considered only the effect of anharmonicity in changing the behavior of the correlation function at long times and have dropped terms of order $\epsilon^2$. Including these terms enables us to see the effect of anharmonicity on the Debye-Waller factor and on the intensity of the Mössbauer line which, as we have shown above, is determined by the Debye-Waller factor alone. When anharmonicity is included, there is a change in the value of the mean square thermal displacements and in their temperature behavior. For example, at temperatures large compared to the Debye temperature, in the harmonic approximation $\langle(\mathbf{k}\cdot\mathbf{u})^2\rangle$ is proportional to T, whereas when anharmonicity is taken into account there is also a term proportional to $T^2$. Moreover, since when anharmonicity is included the probability distribution for thermal displacements is not Gaussian, the passage from the second to the third expression in formula (1) is no longer rigorous. W will not be given solely by the mean square thermal displacements and will not be a quadratic function of the wave vector $\mathbf{k}$; it will also contain terms corresponding to averages of higher powers of the displacements and proportional to the fourth or higher powers of $\mathbf{k}$.

The effect of anharmonicity on the Debye-Waller factor in an ideal crystal has been treated in [9,10,16] in connection with the determination of the intensity of regular x-ray reflections (cf. also [11], where a one-dimensional model is discussed). According to [10], when the anharmonicity is small and when one can stop at the quadratic terms in the cubic anharmonicity constant and the linear terms in the fourth order constant, the expression for W can be written as

$$W = W^0 + W' + W''. \qquad (13)$$

Here $W^0 + W' = \frac{1}{2}\langle(\mathbf{k}\cdot\mathbf{u})\rangle^2$; $W^0$ is calculated from the usual formula for the Debye-Waller factor in the harmonic vibration theory (for an arbitrary dispersion law). Because of thermal broadening the vibration frequencies depend on temperature, and in this sense the anharmonicity is partially included in $W^0$. $W'$ is determined by the corrections to the mean square displacements caused by the third and fourth order terms in the expansion of the Hamiltonian in powers of the deviations of the atoms from their equilibrium positions (at a given temperature). Like $W^0$, $W'$ is a quadratic function of **k**. Unlike $W^0$ and $W'$, $W''$ is a biquadratic function of the wave vector **k**. Thus even in a cubic crystal W should depend not only on the magnitude but also on the direction of the vector **k**, and one should observe an anisotropy in the intensity of the Mössbauer line in cubic crystals.

At high temperatures, larger than the Debye temperature Θ, according to [10], $W'$ is proportional to $T^2$ and can be written in the form

$$W'/W^0 = \alpha\,(\widetilde{\omega^{-2}}/\overline{\omega^{-2}})\,C'/C_0.$$

Here $\widetilde{\omega}^{-2} \sim \overline{\omega}^{-2}$ are the mean values of the inverse square vibration frequency, taken from some of the sums in [10]; $C'$ results from the anharmonicity of the high temperature correction to the crystal specific heat $C_0 = C_V$ calculated in the harmonic approximation; $\alpha$ is a dimensionless factor of order unity ( $\alpha = 2$ if the main contribution comes from the fourth order anharmonicity, and $\alpha = 3$ if it comes from the third order anharmonic term; in the general case $\alpha$ may be less than 2 or greater than 3). Estimates based on this formula show that the ratio $W'/W^0$ may be several times ten. At low temperatures $W'/W^0$ may be $\sim 10^{-2}$. For $T > \Theta$ the term $W''$ is proportional to $T^3$ and appears more clearly at high temperatures and for large energies of the Mössbauer photons.

The temperature dependence of W has been studied experimentally over a wide temperature range for Sn[11]. The observed deviations from a linear dependence of W on T are caused by anharmonicity. The dependence found could be described quantiatively by setting $\alpha\widetilde{\omega}^{-2}(\overline{\omega}^{-2})^{-1} = 4.5$ in the expression for $W'/W^0$, which is in good agreement with the semiquantitative theoretical estimate.

4. A nonquadratic dependence of W on **k** should also occur, even in the absence of anharmonicity, in solid solutions. In solid solutions the mean square thermal displacements depend on which atoms surround a given atom, and are not the same even for atoms of a definite sort. The probability distribution for the thermal displacements of an atom whose type is not preassigned is then not Gaussian (through the probability distribution for a given atom is of course Gaussian in the harmonic approximation). Thus in this case also the last expression in (1) is not exact.

The Debye-Waller factor for nonideal crystals was determined in [12] by looking at the intensity of regular x-ray reflections. These results can be applied to the problem of the intensity of the Mössbauer line, if we consider that in the absence of coherent effects the static defects do not affect the intensity and we set the static shifts equal to zero in the formulas of[12]. Then, for example, for the case of dilute binary solutions A-B with one atom per unit cell, the value of $W_\alpha$ for atoms $\alpha$ ( $\alpha$ = A, B) is given by the following formula:

$$W_\alpha = \tfrac{1}{2}\langle\overline{(\mathbf{k}\mathbf{u}_\alpha)^2}\rangle + c\sum_s[1 - \exp\{-\tfrac{1}{2}\Delta(\mathbf{k}\mathbf{v}_{s\alpha})^2\} - \tfrac{1}{2}\Delta(\mathbf{k}\mathbf{v}_{s\alpha})^2] \approx \tfrac{1}{2}\langle\overline{(\mathbf{k}\mathbf{u}_\alpha)^2}\rangle - \tfrac{1}{8}c\sum_s[\Delta(\mathbf{k}\mathbf{v}_{s\alpha})^2]^2. \qquad (13')$$

Here c is the concentration of impurity atoms, $\langle(\mathbf{k}\cdot\mathbf{u}_\alpha)^2\rangle$ is determined by the mean square thermal displacements of the atoms $\alpha$, $\Delta(\mathbf{k}\cdot\mathbf{v}_{s\alpha})^2$ determines the average (over the vibrations but not over the atom configurations) of the scalar product $\langle(\mathbf{k}\cdot\mathbf{u}_\alpha)^2\rangle$ for the thermal displacements of the atom of type $\alpha$ ( A or B ) at a given site (the origin) when an impurity atom A is introduced at position s ( a site or an interstitial position). The second, approximate, formula in (13′) is obtained by expanding in powers of $\Delta(\mathbf{k}\cdot\mathbf{v}_{s\alpha})^2$.

The last term in (13) is proportional to $k^4$, and obviously leads to an anisotropy of the intensity of the Mössbauer line in cubic crystals, even when there is no anharmonicity. At high temperatures $\Delta(\mathbf{k}\cdot\mathbf{v}_{s\alpha})^2 \sim T$, i.e., this term is proportional to $T^2$. Obviously the anisotropy should show up more clearly for harder radiation (large k), concentrated solutions (large c) and when there is a marked difference between the force constants and the masses of the different sorts of atoms.

Then at high temperatures the mean square thermal displacements (and $\Delta(\mathbf{k}\cdot\mathbf{v}_{s\alpha})^2$; cf.[12]) do not depend on the masses of the atoms, and only the difference between the force constants is important. At low temperatures the quantities $\Delta(\mathbf{k}\cdot\mathbf{v}_{s\alpha})^2$ are determined by both factors.

5. Anharmonicity may not only affect the intensity of the Mössbauer line, but also the broad distribution of intensity of nucleus-phonon emission or absorption of photons. In particular, including anharmonicity should result in a smearing out of the singularities of this distribution which are caused by the presence of critical points in the vibration spectrum (maxima, minima, and saddle points), which, according to [13], should occur for each vibration branch. This effect is analogous to the smearing out of singularities in the energy distribution of incoherently scattered neutrons, which was treated in [7,8].

The singularities in the spectrum of one-phonon emission or absorption of photons which appear most clearly are those corresponding to processes in which the nuclear transition is accompanied by emission or absorption of one phonon. The emission probability for these processes, taking anharmonicity into account in an ideal crystal, is given, using (3), (5), (7) and (8), by the expression

$$J_1(\omega) = e^{-2W}\sum_{\mathbf{k}j}|\alpha_{\mathbf{k}j}|^2[\varphi'_{\mathbf{k}j}(\omega-\omega_0)+\varphi''_{\mathbf{k}j}(\omega-\omega_0)]. \quad (14)$$

Here, instead of using $\kappa$, we characterize the phonon in the ideal crystal by giving the wave vector $\mathbf{k}$ and the vibration branch j. In (14) we have dropped terms of order $\epsilon^2$ (and $\sim\Gamma/\Gamma_\kappa$) in the expression for the emission probability, but these terms can be kept in the expressions for the correlation functions (9).

Neglecting anharmonicity, the functions $\varphi'_{\mathbf{k}j}$ and $\varphi''_{\mathbf{k}j}$ are δ-functions:

$$\varphi'_{\mathbf{k}j}(\omega) = \delta(\omega+\omega_{\mathbf{k}j})\,[n(\omega_{\mathbf{k}j})+1],$$

$$\varphi''_{\mathbf{k}j}(\omega) = \delta(\omega-\omega_{\mathbf{k}j})\,n(\omega_{\mathbf{k}j}).$$

Then as is easily seen from (14) the appearance of a critical point in any vibration branch j′ at the frequency $\omega_1$ ($\nabla_{\mathbf{k}}\omega_{\mathbf{k}j'} = 0$ for $\omega_{\mathbf{k}j'} = \omega_1$) leads to the appearance of a singularity in the function $J_1(\omega)$ of the type $|\omega-\omega_0-\omega_1|^{1/2}$, just as in the case of incoherent scattering of neutrons.[14] A term proportional to $|\omega-\omega_0-\omega_1|^{1/2}$ appears for frequencies $\omega$ lying only on that side of the point $\omega_0+\omega_1$ for which new poles appear on the surface $S_{j'}(\omega)$ with $\omega_{j'}(\mathbf{k}) = \omega-\omega_0$. For example, for a minimum critical point, the term describing the singularity, for the case where the emission of a photon is accompanied by the absorption of a phonon, has the form

$$\Delta J_1(\omega) = B\begin{cases}(\omega-\omega_0-\omega_1)^{1/2}, & \omega-\omega_0>\omega_1\\ 0 & \omega-\omega_0<\omega_1\end{cases},$$

$$B = e^{-2W}\frac{V}{8\pi^3}\int\limits_{S_{j'}(\omega)}\frac{|\alpha_{\mathbf{k}'j'}|^2\,n(\omega_1)}{|\nabla_{\mathbf{k}'}\omega_{\mathbf{k}'j'}|\,(\omega-\omega_1)^{1/2}}\,dS_{j'}. \quad (15)$$

Here B may be assumed independent of $\omega$.

The singularity in the dependence of $J_1$ on $\omega$ is smeared out if the anharmonicity of the vibrations is taken into account. In fact, when anharmonicity is included the phonon correlation function is given by (9). Except for cases where other branches have no vibrations with frequency $\omega_1$, we can neglect the dependence of $\Gamma_{\mathbf{k}j}$ on $\omega$ over the narrow frequency range $\omega-\omega_0-\omega_{\mathbf{k}j}\sim\Gamma_{\mathbf{k}j}$. If on the surface $S_{j'}$ in the neighborhood of the minimum point of the j′ branch we can neglect the dependence of $\Gamma_{\mathbf{k}'j'} = \Gamma(\omega)$ and $\alpha_{\mathbf{k}'j'}$ on $\mathbf{k}'$, then carrying out the integration in (14) with the correlation function (9) we get

$$\Delta J_1(\omega) = \pi B\int\limits_{\omega_1}^{\infty}\frac{\Gamma(\omega)(\omega'-\omega_1)^{1/2}}{(\omega-\omega')^2+\Gamma^2(\omega)}\,d\omega'$$

$$= \frac{B\Gamma(\omega)}{2[(\omega-\omega_1)^2+\Gamma^2(\omega)]^{1/4}\sin(\theta/2)};$$

$$\operatorname{tg}\theta = \frac{\Gamma(\omega)}{\omega-\omega_1};\quad 0<\theta<\pi. \quad (16)^*$$

Thus the singularity in $\Delta J_1(\omega)$ has been smeared over a frequency interval $\sim\Gamma(\omega)$ according to the same law as in the case of incoherent scattering of neutrons (cf. formula (34) in [7]) which also treated the case where one can neglect the dependence of $\Gamma_{\mathbf{k}'j'}$ on $\mathbf{k}'$).

If the dependence $\Gamma_{\mathbf{k}'j'}(\omega)$ on the position of the point $\mathbf{k}'$ on the surface $S_{j'}(\omega)$ is important, a similar treatment gives the following formula for $\Delta J_1(\omega)$:

$$\Delta J_1(\omega) = \frac{1}{2}\int d\Omega\,\frac{B_{\mathbf{k}'j'}\Gamma_{\mathbf{k}'j'}(\omega)}{[(\omega-\omega_1)^2+\Gamma^2_{\mathbf{k}'j'}(\omega)]^{1/4}\sin(\theta_{\mathbf{k}'j'}/2)};$$

$$\operatorname{tg}\theta_{\mathbf{k}'j'} = \frac{\Gamma_{\mathbf{k}'j'}(\omega)}{\omega-\omega_1}. \quad (17)$$

Here the integration extends over the solid angle with its vertex at the minimum point, while

$$B_{\mathbf{k}'j'} = e^{-2W}\frac{V}{8\pi^3}|\alpha_{\mathbf{k}'j'}|^2\frac{dS_{j'}}{d\Omega}\frac{1}{|\nabla_{\mathbf{k}'}\omega_{\mathbf{k}'j'}|\,(\omega-\omega_1)^{1/2}} \quad (18)$$

may be assumed to be independent of $\omega$. It is obvious that (18) gives qualitatively the same dependence of $\Delta J_1(\omega)$ on $\omega$ as does (16).

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*tg = tan.

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## APPENDIX

We shall calculate the correlation function $\langle a_\kappa^+(t)a_\kappa(t)a_\kappa^+(0)a_\kappa(0)\rangle$ and show that the approximate expression (5) proposed by Baym [5] for the function $g_s(\mathbf{k}, t)$ is valid for all t up to terms $\sim \epsilon^2$.

Let us consider the difference between the exact and approximate expressions (4) and (5) for the correlation function:

$$K(t) = \langle \exp(i\mathbf{ku}(t)) \exp(-i\mathbf{ku}(0))\rangle - \exp[-\langle(\mathbf{ku}(0))^2\rangle + \langle(\mathbf{ku}(t))(\mathbf{ku}(0))\rangle]. \quad (A.1)$$

For simplicity we restrict ourselves to the case where each atom is a center of symmetry and the radiation is not too hard, so that in the expansion of (A.1) in powers of **k** we can stop with terms $\sim k^4$ (an assumption which is satisfied in all cases where the Mössbauer effect has been established experimentally). Then

$$\begin{aligned} K(t) = {} & \tfrac{1}{12}\langle(\mathbf{ku})^4\rangle - \tfrac{1}{6}\langle(\mathbf{ku}(t))^3(\mathbf{ku}(0))\rangle \\ & - \tfrac{1}{6}\langle\mathbf{ku}(t))(\mathbf{ku}(0))^3\rangle + \tfrac{1}{4}\langle(\mathbf{ku}(t))^2(\mathbf{ku}(0))^2\rangle \\ & - \tfrac{1}{2}[\langle(\mathbf{ku})^2\rangle]^2 + \langle(\mathbf{ku})^2\rangle\langle(\mathbf{ku}(t))(\mathbf{ku}(0))\rangle \\ & - \tfrac{1}{2}[\langle(\mathbf{ku}(t))(\mathbf{ku}(0))\rangle]^2. \end{aligned} \quad (A.2)$$

We carry out the computation of K(t) neglecting terms $\sim \epsilon^2$. As an example we consider the terms $K_0(t)$ in K(t) which do not contain oscillating factors of the type $\exp(\pm i\omega_\kappa t)$, $\exp[(\pm\omega_\kappa \pm \omega_{\kappa'})it]$, etc. (which determine the intensity of the Mössbauer line), and not the smooth spectral distribution in its wings. Considering the expansion (6) and the commutation rules $[a_\kappa, a_{\kappa'}^+] = \delta_{\kappa\kappa'}$, and neglecting terms $\sim \epsilon^2$, we can write $K_0(t)$ in the form

$$K_0(t) = \sum_\kappa |\alpha_\kappa|^4 [\langle n_\kappa(t)\, n_\kappa\rangle - \bar{n}_\kappa^2 - \langle a_\kappa(t)\, a_\kappa^+(0)\rangle\langle a_\kappa^+(t)\, a_\kappa(0)\rangle], \quad (A.3)$$

where $n_\kappa(t) = a_\kappa^+(t)a_\kappa(t)$, $n_\kappa = n_\kappa(0)$, while $\bar{n}_\kappa$ is the average occupation number. In obtaining (A.3) it was assumed that the local modes are not degenerate (though it is not difficult to carry through the treatment without this assumption). In the harmonic approximation $K_0(t) = 0$. But the fact that when we include anharmonicity $K_0(t) \sim \epsilon^2$ at all times requires separate proof since the time dependence of the differnce of the first two terms in (A.3) and the last term can be different, in general. It is therefore necessary to calculate the correlation functions appearing in (A.3) for a vibration Hamiltonian which includes the anharmonicity:

$$H = \sum_\kappa \omega_\kappa a_\kappa^+ a_\kappa + \tfrac{1}{2}\sum_{\kappa\kappa'\kappa''}(V_{\kappa\kappa'\kappa''}\, p a_\kappa a_{\kappa'}^+ a_{\kappa''}^+ + \tfrac{1}{3}V'_{\kappa\kappa'\kappa''} a_\kappa a_{\kappa'} a_{\kappa''} + \text{herm. conj.}). \quad (A.4)$$

Here ħ = 1, the anharmonicity constants V and V′ are proportional to the small parameter $\epsilon$ and $pa_\kappa a_{\kappa'}^+ a_{\kappa''}^+ = \frac{1}{3}(a_\kappa a_{\kappa'}^+ a_{\kappa''}^+ + a_{\kappa'}^+ a_\kappa a_{\kappa''}^+ + a_{\kappa'}^+ a_{\kappa''}^+ a_\kappa)$.

When anharmonicity is taken into account, the last term in (A.3) is given by formulas (8) and (9). To determine the difference of the first two terms one must find the correlation function $\langle n_\kappa(t), n_\kappa - \bar{n}_\kappa\rangle$. The calculation of this correlation function can be done by the method of retarded and advanced temperature Green's functions.[15] We introduce the retarded Green's function $g_\kappa(t)$ for the operators $n_\kappa$:

$$g_\kappa(t) = \langle\langle n_\kappa(t); n_\kappa - \bar{n}_\kappa\rangle\rangle = -i\theta(t)\langle\{n_\kappa(t), n_\kappa - \bar{n}_\kappa\}\rangle, \quad (A.5)$$

where the curly brackets denote the anticommutator, and $\theta(t) = 1$ for $t > 0$ and $\theta(t) = 0$ for $t < 0$. For the advanced Green's function $\theta(t)$ is replaced by $-\theta(-t)$. In this definition of the Green's function we take the anticommutator instead of the commutator in order that the equation of motion will be inhomogeneous.

From (A.4) and (A.5) it follows that the equation of motion for the function $g_\kappa(t)$ has the form

$$\begin{aligned} i\frac{dg_\kappa}{dt} = {} & 2\delta(t)(\overline{n_\kappa^2} - \bar{n}_\kappa^2) \\ & + \frac{1}{2}\sum_{\kappa'\kappa''}\{-V_{\kappa\kappa'\kappa''}\langle\langle p a_\kappa a_{\kappa'}^+ a_{\kappa''}^+;\ n_\kappa - \bar{n}_\kappa\rangle\rangle + \\ & + V^*_{\kappa\kappa'\kappa''}\langle\langle p a_\kappa^+ a_{\kappa'} a_{\kappa''};\ n_\kappa - \bar{n}_\kappa\rangle\rangle \\ & + 2V_{\kappa'\kappa\kappa''}\langle\langle p a_{\kappa'} a_\kappa^+ a_{\kappa''}^+;\ n_\kappa - \bar{n}_\kappa\rangle\rangle - \\ & - 2V^*_{\kappa'\kappa\kappa''}\langle\langle p a_{\kappa'}^+ a_\kappa a_{\kappa''};\ n_\kappa - \bar{n}_\kappa\rangle\rangle - V'_{\kappa\kappa'\kappa''}\langle\langle a_\kappa a_{\kappa'} a_{\kappa''}; \\ & n_\kappa - \bar{n}_\kappa\rangle\rangle + V'^*_{\kappa\kappa'\kappa''}\langle\langle a_\kappa^+ a_{\kappa'}^+ a_{\kappa''}^+;\ n_\kappa - \bar{n}_\kappa\rangle\rangle\}. \end{aligned} \quad (A.6)$$

Following the method for truncating the chain of equations for the Green's function [15], we construct the equation of motion for the functions on the right of (A.6), and truncate them in an approximation corresponding to the second order perturbation theory for the inverse Green's function. For example, the equations for the first two Green's functions on the right of (A.6) have the form:

$$
\begin{aligned}
i\frac{d}{dt}\langle\langle pa_\varkappa a^+_{\varkappa'}a^+_{\varkappa''};\quad n_\varkappa-\bar n_\varkappa\rangle\rangle &= \delta(t)\langle\{pa_\varkappa a^+_{\varkappa'}a^+_{\varkappa''};\ n_\varkappa-\bar n_\varkappa\}\rangle\\
&+(\omega_\varkappa-\omega_{\varkappa'}-\omega_{\varkappa''})\langle\langle pa_\varkappa a^+_{\varkappa'}a^+_{\varkappa''};\quad n_\varkappa-\bar n_\varkappa\rangle\rangle\\
&-V^*_{\varkappa\varkappa'\varkappa''}(1+\bar n_{\varkappa'}+\bar n_{\varkappa''})g_\varkappa-\\
&-\tfrac{1}{2}\sum_{\varkappa_1}(\delta_{\varkappa\varkappa'}V^*_{\varkappa_1\varkappa_1\varkappa''}+\delta_{\varkappa\varkappa''}V^*_{\varkappa_1\varkappa_1\varkappa'})(2\bar n_{\varkappa_1}+1)g_\varkappa,
\end{aligned}
\tag{A.7}
$$

$$
\begin{aligned}
i\frac{d}{dt}\langle\langle pa^+_\varkappa a_{\varkappa'}a_{\varkappa''};\ n_\varkappa-\bar n_\varkappa\rangle\rangle &= \delta(t)\langle\{pa^+_\varkappa a_{\varkappa'}a_{\varkappa''},\ n_\varkappa-\bar n_\varkappa\}\rangle\\
&-(\omega_\varkappa-\omega_{\varkappa'}-\omega_{\varkappa''})\langle\langle pa^+_\varkappa a_{\varkappa'}a_{\varkappa''};\ n_\varkappa-\bar n_\varkappa\rangle\rangle\\
&+V_{\varkappa\varkappa'\varkappa''}(1+\bar n_{\varkappa'}+\bar n_{\varkappa''})g_\varkappa+\\
&+\tfrac{1}{2}\sum_{\varkappa_1}(\delta_{\varkappa\varkappa'}V_{\varkappa_1\varkappa_1\varkappa''}+\delta_{\varkappa\varkappa''}V_{\varkappa_1\varkappa_1\varkappa'})(1+2\bar n_{\varkappa_1})g_\varkappa.
\end{aligned}
\tag{A.8}
$$

We note that the other terms on the right of (A.7) and (A.8), for example terms containing the Green's functions $\langle\langle a^+_{\kappa'}a_{\kappa'}a^+_{\kappa''}a_{\kappa''};\ n_\kappa-\bar n_\kappa\rangle\rangle$, are proportional to $\epsilon^2$ and can be dropped only when the Green's functions are suitably chosen (with anticommutators); when the quantity to the right of the semicolon is $n_\kappa-\bar n_\kappa$, whose average is zero (and not $n_\kappa$, for example).

The averages for a single time in the first terms of (A.7) and (A.8) can be calculated to terms $\sim\epsilon$ using the wellknown expansion of the exponential operator:

$$
e^{-\lambda(H_0+H_1)}=e^{-\lambda H_0}-\lambda\int_0^1 du\,e^{-\lambda(1-u)H_0}H_1e^{-\lambda uH_0}+\ldots,
\tag{A.9}
$$

applied to the operator $\exp(-\lambda H)$ $(\lambda=(k_BT)^{-1})$. The averages appearing in (A.7) and (A.8) are equal to

$$
\begin{aligned}
\langle\{a_\varkappa a^+_{\varkappa'}a^+_{\varkappa''};\ n_\varkappa-\bar n_\varkappa\}\rangle &= -\langle\{a^+_\varkappa a_{\varkappa'}a_{\varkappa''};\ n_\varkappa-\bar n_\varkappa\}^*\rangle\\
&=2V^*_{\varkappa\varkappa'\varkappa''}(\omega_\varkappa-\omega_{\varkappa'}-\omega_{\varkappa''})^{-1}\\
&\times[1-\exp\{-\lambda(\omega_\varkappa-\omega_{\varkappa'}-\omega_{\varkappa''})\}]\\
&\times(1+\bar n_\varkappa)(1+2\bar n_\varkappa)\bar n_{\varkappa'}\bar n_{\varkappa''}.
\end{aligned}
\tag{A.10}
$$

Here $\kappa\neq\kappa'\neq\kappa''$ and we use the fact that $\overline{n^2_\kappa}=\bar n_\kappa(1+2\bar n)$.

Next proceeding as usual from the Green's functions to their Fourier transforms, we find that, for example, the Fourier transform of $\langle\langle a_\kappa a^+_{\kappa'}a^+_{\kappa''};\ n_\kappa-\bar n_\kappa\rangle\rangle$ is a linear function of the Fourier transform $g_\kappa(\omega)$. After substituting these linear functions on the right of the Fourier transform equation (A.6), we see from (A.10) and similar expressions for other averages that the free terms under the summation sign cancel one another and the sum will be proportional to $g_\kappa(\omega)$. From the linear equation found for $g_\kappa(\omega)$ it follows that

$$
g_\varkappa(\omega)=\frac{1}{\pi}\frac{\overline{n^2_\varkappa}-\bar n^2_\varkappa}{\omega-R_\varkappa(\omega)+R_\varkappa(-\omega)},
\tag{A.11}
$$

$$
\begin{aligned}
R_\varkappa(\omega)=\frac{1}{2}\sum_{\varkappa'\varkappa''}\Bigg[&\frac{|V_{\varkappa\varkappa'\varkappa''}|^2(1+\bar n_{\varkappa'}+\bar n_{\varkappa''})}{\omega-\omega_\varkappa+\omega_{\varkappa'}+\omega_{\varkappa''}}\\
&-\frac{2|V_{\varkappa'\varkappa\varkappa''}|^2(\bar n_{\varkappa'}-\bar n_{\varkappa''})}{\omega-\omega_\varkappa+\omega_{\varkappa'}-\omega_{\varkappa''}}-\frac{|V'_{\varkappa\varkappa'\varkappa''}|^2(1+\bar n_{\varkappa'}+\bar n_{\varkappa''})}{\omega-\omega_\varkappa-\omega_{\varkappa'}-\omega_{\varkappa''}}\Bigg].
\end{aligned}
\tag{A.12}
$$

The Fourier component $\nu_\kappa(\omega)$ of the correlation function $\langle n_\kappa(t),\ n_\kappa-\bar n_\kappa\rangle$ is related to the Fourier component of the Green's function $g_\kappa(\omega)$ by the relation (cf.[15])

$$
\nu_\varkappa(\omega)=i(e^{\lambda\omega}+1)^{-1}[g_\varkappa(\omega+i\xi)-g_\varkappa(\omega-i\xi)]
\tag{A.13}
$$

$(\xi\to+0)$. Remembering that $(x+i\xi)^{-1}=Px^{-1}-i\pi\delta(x)$ (where P denotes the principal value), we find from (A.11)—(A.13) the following expression for $\nu_\kappa(\omega)$:

$$
\nu_\varkappa(\omega)=\frac{1}{\pi}\frac{\gamma_\varkappa(\omega)+\gamma_\varkappa(-\omega)}{[\omega-\pi_\varkappa(\omega)+\pi_\varkappa(-\omega)]^2+[\gamma_\varkappa(\omega)+\gamma_\varkappa(-\omega)]^2}\times\bar n_\varkappa(\bar n_\varkappa+1).
\tag{A.14}
$$

Here $\pi_\kappa(\omega)$ and $\gamma_\kappa(\omega)$ are gotten from (A.12) by replacing the factor $(\omega-a)^{-1}$ in each of the terms by $P(\omega-a)^{-1}$ and $\pi\delta(\omega-a)$, respectively. The functions $\pi_\kappa(\omega)$ and $\gamma_\kappa(\omega)$ are proportional to $\epsilon^2$. Neglecting their variation over the region of small $|\omega|\lesssim\gamma_\kappa$, we can set $\pi_\kappa(\omega)=\pi_\kappa(-\omega)$ and replace $\gamma_\kappa(\omega)$ and $\gamma_\kappa(-\omega)$ by the constant quantity

$$
\gamma_\varkappa(0)=\frac{\pi}{2}\sum_{\varkappa'\varkappa''}|V_{\varkappa\varkappa'\varkappa''}|^2(1+\bar n_{\varkappa'}+\bar n_{\varkappa''})\,\delta(\omega_\varkappa-\omega_{\varkappa'}-\omega_{\varkappa''}).
\tag{A.15}
$$

Transforming from the Fourier component $\nu_\kappa(\omega)$ to the correlation function, we get the following expression for the difference of the first two terms in the square bracket in (A.3):

$$
\langle n_\varkappa(t),\ n_\varkappa-\bar n_\varkappa\rangle=\bar n_\varkappa(\bar n_\varkappa+1)\exp\{-2\gamma_\varkappa(0)|t|\}.
\tag{A.16}
$$

It follows from (8) and (9) that for small $\epsilon$ the last term is equal to

$$
\bar n_\varkappa(\bar n_\varkappa+1)\exp\{-2\Gamma_\varkappa(\omega_\varkappa)|t|\}.
\tag{A.17}
$$

The damping $\Gamma_\kappa(\omega)$ and shift $P_\kappa(\omega)$ of the phonons for the anharmonic Hamiltonian in a nonideal crystal (A.4) are determined by the relation (cf.[7,8]):

$$P_\varkappa(\omega) - i\Gamma_\varkappa(\omega) = R_\varkappa(\omega + i\xi),$$

where

$$R_\varkappa(\omega) = \frac{1}{2}\sum_{\varkappa'\varkappa''}\left\{\frac{|V_{\varkappa\varkappa'\varkappa''}|^2(1+\bar{n}_{\varkappa'}+\bar{n}_{\varkappa''})}{\omega-\omega_{\varkappa'}-\omega_{\varkappa''}} - \frac{|V'_{\varkappa\varkappa'\varkappa''}|^2(1+\bar{n}_{\varkappa'}+\bar{n}_{\varkappa''})}{\omega+\omega_{\varkappa'}+\omega_{\varkappa''}} - \frac{2|V_{\varkappa'\varkappa\varkappa''}|^2(\bar{n}_{\varkappa'}-\bar{n}_{\varkappa''})}{\omega-\omega_{\varkappa'}+\omega_{\varkappa''}} + V_{\varkappa\varkappa\varkappa''}V^*_{\varkappa'\varkappa'\varkappa''}(1+2\bar{n}_{\varkappa'}) \times \left(\frac{1}{\omega-\omega_\varkappa-\omega_{\varkappa''}} - \frac{1}{\omega-\omega_\varkappa+\omega_{\varkappa''}}\right)\right\}. \quad (A.18)$$

From (A.15) and (A.18) $\gamma_K(0) = \Gamma_K(\omega_K)$. Therefore neglecting terms $\sim \epsilon^2$ (but not assuming that $\epsilon^2 t\omega_K$ is small) the expression in square brackets in (A.3) goes to zero. It then follows that $K_0(t)$ is of order $\epsilon^2$.

By similar arguments one can also show that the other oscillating part of K(t) is of order $\epsilon^2$.

Note added in proof (Jan. 14, 1964). Recently it has been shown (M. A. Krivoglaz, FTT, in press) that inclusion of anharmonicity not only gives the effect considered here of a reduction in the intensity of the Mössbauer line by a finite factor (when local modes are present), but can also cause a reduction by several orders of magnitude of the effect predicted by Snyder and Wick [H. S. Snyder and G. C. Wick, Phys. Phys. Rev. 120, 128 (1960)], which is a broadening of the line line through interaction with the local vibrations.

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