PHYSICAL REVIEW B VOLUME 10, NUMBER 5 1 SEPTEMBER 1974

Effect of the thermal relaxation of a lattice mode on the Mössbauer radiation*

Ajay Gupta† and K. Rama Reddy
Department of Physics, Aligarh Muslim University, Aligarh-202001, India
(Received 23 May 1973)

The spectral distribution of the Mössbauer radiation emitted from a radiating nucleus when it is in the excited vibrational level of a lattice normal mode has been considered theoretically. In obtaining the results, the relevant correlation functions, which include the thermal relaxation of phonons, are calculated in the harmonic approximation. Results are presented for the line shapes of the zero-, one-, and two-phonon-assisted Mössbauer transitions. The "relaxation splitting" is absent in our results and the narrowing of the zero-phonon Mössbauer line never exceeds $\sim 8\%$ of the natural linewidth.

INTRODUCTION

The line shape of the Mössbauer resonance is an incompletely solved problem; the nature of solid-state interactions causing the perturbations of the Mössbauer radiation are not exactly known. The recent observations of phonon-assisted Mössbauer transitions[1] have created a new interest in the study of the line shapes perturbed either by an rf pumped normal mode or by a localized mode; the continuum mode of the lattice is considered as the normal mode and the localized vibration around the impurity (Mössbauer) nucleus is the localized mode. The influence of these modes[2–4] and their thermal relaxation on the spectral distribution of the Mössbauer radiation has been extensively investigated in recent years. But the theories developed so far have some inconsistencies which are summarized as follows in the next paragraph.

In the theory of Kaufman and Lipkin[2] the high-amplitude localized mode associated with the radiating nucleus would enhance the intensity of the Mössbauer line in comparison with the usual Debye-Waller factor. This enhancement is known in the literature as the Bessel-function enhancement.[5] Lax and Waller[6] have extended this theory to include the phonon-damping effect on the Mössbauer radiation. However, the consequence of the Bessel-function enhancement leads to the absurd conclusion that the highly excited lattice mode should enhance the recoilless factor. Therefore, the phonon-damping effects as calculated by Lax and Waller[6] will be of little value for the comparison of theoretical line shapes with experimental results.

In the theory of Dash and Nussbaum[7] the radiating nucleus is in a highly excited vibrational state of the long-lived localized mode and the thermal-averaged mean-square vibrational amplitude is assumed to decay to its equilibrium value with a characteristic relaxation rate. The physical description of the phenomenon which involved calculations of the photon amplitude by using the positive square root of the time-dependent recoilless fraction has also been the subject of criticism.[8] Further considerations of this theory, as shown in the present work, give "negative intensities" in the Mössbauer spectra which are unphysical in our opinion.

In an attempt to understand the lattice-relaxation phenomenon, Harris[8] has given a quantum-mechanical model which includes both nuclear radiation and the lattice-relaxation processes. This model predicts, besides the changes in line shapes, the existence of an apparent "relaxation splitting" of the Mössbauer line for a certain range of relaxation rates. The serious drawback with this model is the conspicuous absence of the statistical averages for the phonons owing to thermal processes.[9] It is a fact that the Mössbauer spectrum represents an ensemble average over many nuclei and in the calculation of the Mössbauer cross section, a thermal average over all the lattice modes is called for. Therefore, one wonders in a statistical ensemble, while calculating the Mössbauer line shapes, whether the phase coherence between the nuclear radiative transition and the lattice transition amplitudes is maintained. The splitting obtained with this model is the consequence of maintaining the phase coherence between the nucleus and the lattice transition amplitudes.

Abragam[3] and Mishory and Bolef[4] have derived expressions for the relative intensities of side bands produced in the Mössbauer resonance owing to an excited lattice mode. This theory is adequate so far as the rf pumped phonons are concerned but certainly is not so for localized phonons: even the very-low-excited vibrational amplitude of a localized mode can cause considerable perturbation on the Mössbauer radiation. Therefore in the following we have presented model-independent calculations which give an adequate description of the line shapes even in presence of thermally relaxing phonons.

THEORY

In our treatment the excited vibrational state may be a localized mode or a normal mode of the

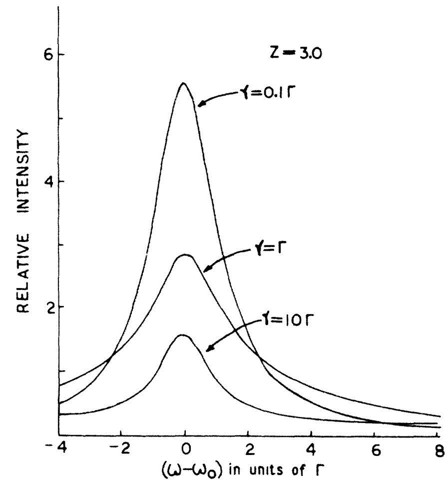

FIG. 1. Zero-phonon Mössbauer line shapes for various values of the phonon relaxation rate in conformity with the Ref. 6. Parameter Z is taken to be 3.0.

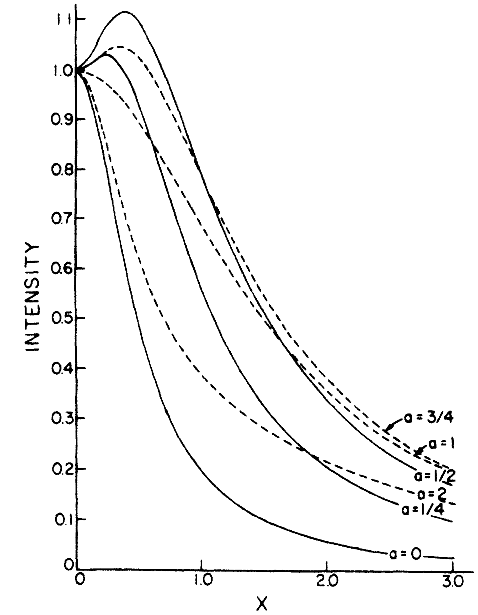

FIG. 3. Zero-phonon Mössbauer line shapes as taken directly from Ref. 8 (parameter $\alpha = -1$). Parameters a and α are related to the phonon relaxation rate γ and Z, respectively. The parameter X is given in units of Γ.

lattice. The excited Mössbauer state of the nucleus is generally created by the radioactive decay of a parent nucleus which may be an impurity in the lattice. In this decay process a large amount of recoil kinetic energy is imparted to the nucleus, leaving the localized mode in the highly excited vibrational state. A particular continuum normal mode can also be excited by applying external perturbation to the sample. The experimental technique to do this could be similar to that of Albanese *et al.*[10] Thus the highly excited vibrational state will have the tendency to relax exponentially

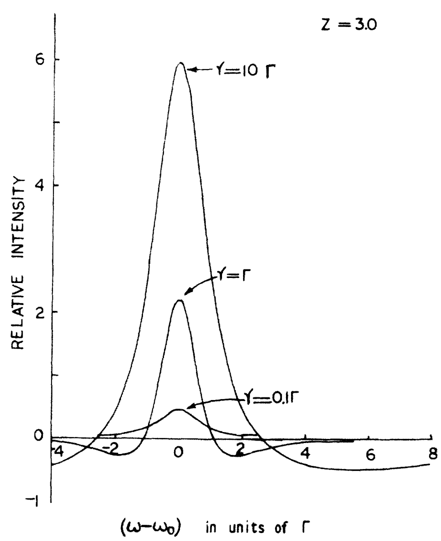

FIG. 2. Zero-phonon Mössbauer line shapes for various values of the phonon relaxation rate as expected on the basis of the theory given in Ref. 7 ($Z = 3.0$).

to its thermal equilibrium value owing to the anharmonic coupling to the lattice. Thus our aim is to investigate the effect of phonons and their thermal relaxation on the frequency distribution of the Mössbauer radiation.

Effect of an undamped lattice mode on the Mössbauer spectrum

In this section we calculate the effect of an excited lattice vibrational mode analogous with the calculation of the self-correlation function for the scattering of slow neutrons by the harmonic mode in a periodic structure.[11] A representative mode designated as q' mode, which is highly excited by external means and for which the average number of phonons is greater than the normally allowed excitation numbers, has a special significance in these calculations. The expression for the recoilless-emission cross section per nucleus for a γ ray can be obtained from the dispersion theory. The result[9] is

$$\sigma(E) = \tfrac{1}{4}\sigma_0 \Gamma \int_{-\infty}^{\infty} e^{-it(E-E_0)-\Gamma|t|/2} \times \langle e^{-i\vec{k}\cdot\vec{U}(t)} e^{i\vec{k}\cdot\vec{U}(0)}\rangle_T \, dt \ , \quad (1)$$

where $\langle\ldots\rangle_T$ denotes the lattice thermal average, $\vec{U}$ is the displacement of the radiating nucleus from its mean position, $\vec{p} = \vec{k}$ with $\hbar = 1$, the linear momentum of the emitted photon, and Γ the width of the decaying nuclear state. Expressing the $\vec{U}$ in terms of normal modes it can be shown that for the harmonic solid[12]

$$\langle e^{-i\vec{k}\cdot\vec{U}(t)}\, e^{i\vec{k}\cdot\vec{U}(0)}\rangle_T = \exp\left(-\frac{k^2}{2MN}\sum_{q\neq q'} \omega_q^{-1}\left[(2n_q+1)(1-\cos\omega_q t)+i\sin\omega_q t\right]\right)$$
$$\times\exp\left(-\frac{k^2}{2MN}\omega_{q'}^{-1}\left[(2n_{q'}+1)(1-\cos\omega_{q'} t)+i\sin\omega_{q'} t\right]\right)\ , \qquad (2)$$

where the contribution of the q'th mode is separated out. Following the argument given by Kittel[12] it can be shown that the first exponential on the right-hand side of Eq. (2) is the usual Debye-Waller factor $f=e^{-2W}$ with the assumption that the contribution of the q'th mode to the f factor is negligible when the mode perturbation is absent. The second exponential of Eq. (2) may be written in terms of Bessel functions[13] and the final result is

$$\langle e^{-i\vec{k}\cdot\vec{U}(t)}\, e^{i\vec{k}\cdot\vec{U}(0)}\rangle_T = e^{-2W} e^{-Z}\left(\sum_{n=-\infty}^{\infty} I_n(Z)\, e^{in\omega_{q'}t}\right)\times\left(\sum_{m=-\infty}^{\infty} J_m(Y)\, e^{im\omega_{q'}t}\right)\ , \qquad (3)$$

where

$$Z=k^2/2MN\left(\frac{2n_{q'}+1}{\omega_{q'}}\right)\ ,\quad Y=k^2/2MN\omega_{q'}\ .$$

$I_n(Z)$ is the modified Bessel function of order n and $J_m(Y)$ is the Bessel function of order m. Combining Eqs. (3) and (1) one obtains

$$\sigma(E)=\sigma_0 e^{-2W} e^{-Z}\sum_{n=0}^{\infty}\sum_{m=0}^{\infty} I_n(Z)\, J_m(Y)\times\frac{\frac{1}{4}\Gamma^2}{(E-E_0\pm n\omega_{q'}\pm m\omega_{q'})^2+\frac{1}{4}\Gamma^2}\ . \qquad (4)$$

From Eq. (4) one finds that the effect of the excited mode is to give rise to an infinite set of phonon-assisted recoilless transitions. These results agree with those of the earlier authors[3,4] provided $Y\to 0$, a situation applicable for the rf pumped phonons; in the usual experimental configurations the rf pumping of the phonons is done only for the continuum modes in which case the number N of the participating atoms is of the order of 10^{20}. However for the localized mode the number N of nuclei participating in the mode is $N\sim 1$; therefore, the parameter Y can be of the order of unity and thus the dependence of a localized mode on the Bessel function $J_m(Y)$ becomes significant.

It may be interesting to compare these calculations with those of Kaufman and Lipkin who found that the presence of a localized mode causes the absorption cross section $\sigma_a(E)$ to be

$$\sigma_a(E)=\sigma_0 e^{-2W}\sum_{n=0}^{\infty} I_n(Z')\times\frac{\frac{1}{4}\Gamma^2}{(E-E_0\pm n\omega_{q'})^2+\frac{1}{4}\Gamma^2}\ , \qquad (5)$$

with

$$Z'=2Z\frac{[n_{q'}(n_{q'}+1)]^{1/2}}{(2n_{q'}+1)}\ .$$

Equation (5) resembles Eq. (4) but with some factors missing. The interpretation of Eq. (5) is as follows. Since the Debye-Waller factor gives the probability of those processes in which the initial and the final states of the lattice are identical, the product of the Bessel function for $n=0$ represents the contribution to the cross section from processes in which the local-mode phonons are excited and deexcited in all possible ways such that the net energy change of the crystal is zero. This in turn will enhance the intensity of the zero-phonon Mössbauer line. However the presence of phonons also increases the probability of those recoilless transitions in which net n phonons are either emitted or absorbed and thus decreases the intensity of the zero-phonon recoilless transition. Equation (4) which incorporates these effects predicts a net decrease in the intensity of the zero-phonon Mössbauer radiation which is contradictory to the results obtained using Eq. (5). And thus the Bessel-function enhancement does not really exist.

The experiments of Perlow[14] on the rf perturbation of the Mössbauer hyperfine spectrum deserves a special mention. The rf perturbation on the spectrum was thought to be due to rapid 180° wall motion. However it was suggested by us[15] and by others[1] that these results could be understood in terms of the magnetostriction model proposed by Pfeiffer *et al.* Our explanation seeks to separate the effects of domain-wall motion and magnetostriction without recourse to additional experiments.[1] Since the number of induced phonons due to magnetostriction coupling is proportional to the rf energy density the rms Z parameter is proportional to the square of the perturbing rf field. Deviation from this relationship must reflect the effect of domain-wall motion, if present.

Effect of the thermal relaxation of phonons on the Mössbauer radiation

In this section we calculate the Mössbauer line shapes while the normal mode is relaxing from its excited vibrational state to the lattice-thermal-equillibrium state. Several authors have attempted this problem to predict the effect of the relaxing phonons on the Mössbauer line shapes. But, as we will see in the following, none of these theories are

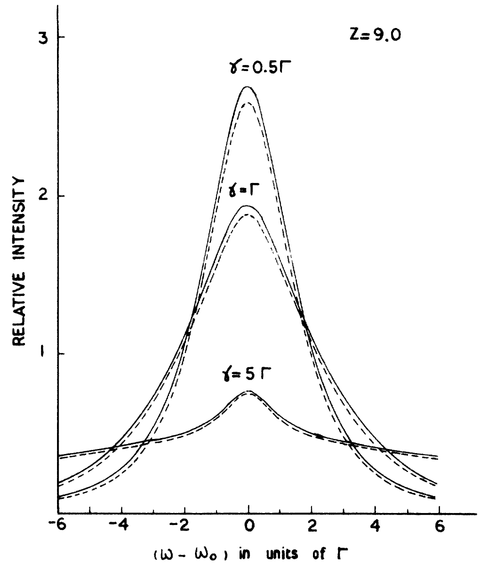

FIG. 4. Zero-phonon Mössbauer line shapes for various values of the phonon relaxation rate γ obtained on the basis of the present work; continuous curves correspond to $Y=0.0$ and dashed curves for $Y=0.5$.

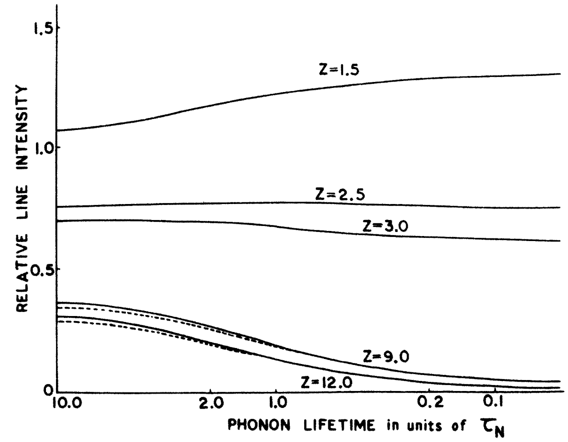

FIG. 6. Intensity of the zero-phonon Mössbauer line is plotted as a function of γ for various values of Z; continuous curves correspond to $Y=0.0$ and dashed curves for $Y=0.5$.

satisfactory. Therefore, before we go on to our calculations, the results of various authors are presented on common footing which allows a meaningful comparision.

(i) The zero-phonon Mössbauer line shape in the presence of relaxing phonons with a decay rate of γ as calculated by Lax and Waller[6] is given as

$$A_0(E)=e^{-2W}\sum_{n=0}^{\infty}\frac{\frac{1}{2}(\Gamma+2n\gamma)}{(E-E_0)^2+\frac{1}{4}(\Gamma+2n\gamma)^2}\,\frac{(\frac{1}{2}Z')^{2n}}{(n!)^2}\;. \tag{6}$$

From this equation it follows that the zero-phonon line is a superposition of a number of Lorentzian lines each having a width $\Gamma+2n\gamma$ and centered at the γ-ray transition energy E_0. The line shapes as calculated from the above equation with $Z'=3.0$ for different phonon-relaxation rates are shown in Fig. 1. From these curves it may be noted that the intensity of the recoilless line increases with

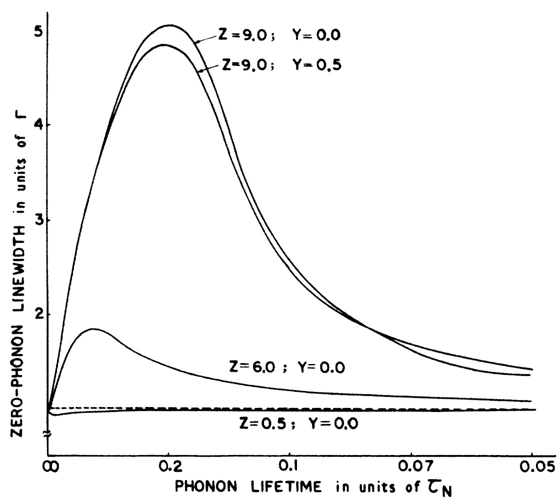

FIG. 5. Zero-phonon Mössbauer linewidth as a function of phonon relaxation rate (linewidth is defined here as the full width at half-maximum).

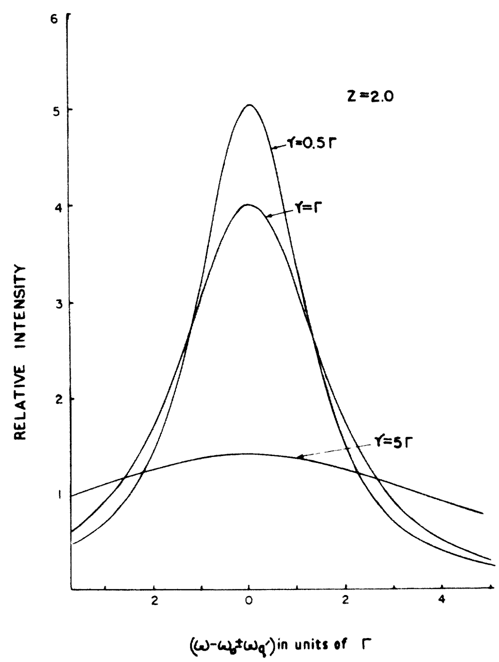

FIG. 7. Line shapes of the one-phonon assisted Mössbauer transition for various values of the lattice-mode relaxation rate (parameters $Z=2.0$ and $Y=0.0$).

decreasing relaxation rate. Further, when $\gamma = 0$ Eq. (6) reduces to Eq. (5) which predicts that by the increase of Z' (which is equivalent to an increase in the number of the phonons in the mode) the intensity of the zero-phonon Mössbauer line should increase. This fact is in contradiction with the experimental observations which may be inferred from the temperature dependence of the intensity of the Mössbauer line.

(ii) A semiclassical treatment[7] of this problem for the line shape of the Mössbauer transition is given by $|E(\omega)|^2$ where

$$E(\omega) = [\langle f \rangle]^{1/2} \int_0^\infty \exp(-Z e^{-\gamma|t|}) \times \exp[-it(E - E_0) - \tfrac{1}{2}\Gamma|t|]\, dt \;. \qquad (7)$$

The line shapes calculated with the help of the above equation are shown in the Fig. 2. The width of the Mössbauer line for all nonzero values of Z and γ, becomes less than the natural linewidth and the Mössbauer intensity can also become negative! For a given value of γ, the linewidth decreases with increasing value of Z and for very large values of Z it may even reduce to zero. The narrowing of the line is also accompanied by a substantial loss in over-all intensity of the Mössbauer radiation.

(iii) A quantum-mechanical model for the Mössbauer line shapes in the presence of lattice relaxation has been attempted by Harris.[8] Figure 3 gives the zero-phonon line shapes obtained on this model as given by Harris. One can see that for certain values of phonon-relaxation rate γ (which is related to a) and Z (which is related to α) the single Mössbauer line is split into two lines. This splitting may be called "relaxation splitting"; it has no relation to the hyperfine splitting.

It is now clear that the results obtained by different authors are in conflict with each other. Therefore, we would like to give our own theory for the phonon-relaxation broadening of the Mössbauer radiation.[16] To determine the Mössbauer line shapes in the presence of lattice relaxation we calculate the correlation function $\langle e^{-i\vec{k}\cdot\vec{U}(t)} e^{i\vec{k}\cdot\vec{U}(0)}\rangle_T$ by taking the average value of the creation and annihilation operators for q'th mode which is decaying with time according to the prescription given as[6]

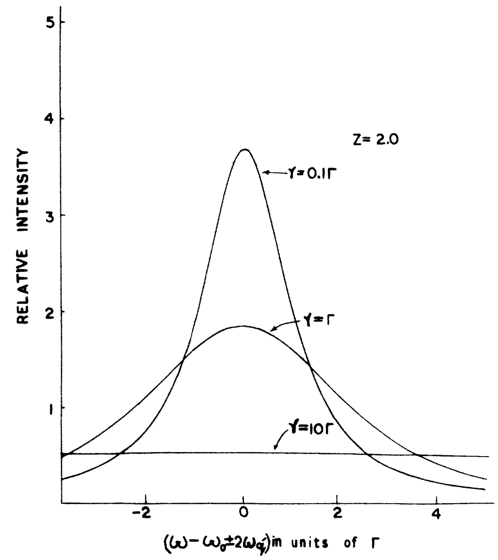

FIG. 8. Line shapes of two-phonon assisted Mössbauer transitions for various values of the phonon relaxation rate (parameters $Z = 2.0$ and $Y = 0.0$).

$$\langle a^\dagger_{q'}(t)\rangle \simeq e^{i\omega_{q'}t} e^{-\gamma|t|/2} \langle a^\dagger_{q'}(0)\rangle \;,$$
$$\langle a_{q'}(t)\rangle \simeq e^{-i\omega_{q'}t} e^{-\gamma|t|/2} \langle a_{q'}(0)\rangle \;. \qquad (8)$$

As usual we calculate the correlation function by expanding $\vec{U}(t)$ and $\vec{U}(0)$ in terms of normal modes. Since the expectation value of terms like $a^\dagger_{q'} a^\dagger_{q'}$ or $a_{q'} a_{q'}$ is zero we may write

$$\langle e^{-i\vec{k}\cdot\vec{U}(t)} e^{i\vec{k}\cdot\vec{U}(0)}\rangle_T = e^{-2W} \exp\Big(\frac{k^2}{4MN}\frac{1}{\omega_{q'}} \langle a_{q'}(t) a^\dagger_{q'}(t) - a_{q'}(t) a^\dagger_{q'}(0) + a^\dagger_{q'}(t) a_{q'}(t) - a^\dagger_{q'}(t) a_{q'}(0) - a_{q'}(0) a^\dagger_{q'}(t) + a_{q'}(0) a^\dagger_{q'}(0) - a^\dagger_{q'}(0) a_{q'}(t) + a^\dagger_{q'}(0) a_{q'}(0) - [a_{q'}(0) + a^\dagger_{q'}(0),\, a_{q'}(t) + a^\dagger_{q'}(t)]\rangle_T\Big) \;. \qquad (9)$$

Now if the phonon-relaxation time is sufficiently large to satisfy the following conditions[17]

$$\hbar\gamma \ll KT, \quad \gamma \ll \omega_{q'}$$

then we can write

$$\langle a^\dagger_{q'}(t) a_{q'}(0)\rangle_T \simeq e^{-\gamma|t|/2} e^{i\omega_{q'}t} \langle a^\dagger_{q'}(0) a_{q'}(0)\rangle_T \;,$$
$$\langle a^\dagger_{q'}(t) a_{q'}(t)\rangle_T \simeq e^{-\gamma|t|} \langle a^\dagger_{q'}(0) a_{q'}(0)\rangle_T \;, \qquad (10)$$

and the correlation function becomes

$$\langle e^{-i\vec{k}\cdot\vec{U}(t)} e^{i\vec{k}\cdot\vec{U}(0)}\rangle_T = e^{-2W} \exp[-\tfrac{1}{2}Z(1 + e^{-\gamma|t|}) + Z e^{-\gamma|t|/2}\cos\omega_{q'}t] \exp[Y e^{-\gamma|t|/2}\sin\omega_{q'}t] \;. \qquad (11)$$

Therefore, combining Eqs. (11) and (1) the expression for the emission cross section becomes

$$\sigma(E)=\tfrac{1}{4}\sigma_0\Gamma e^{-2W}e^{-Z/2}\sum_{n=0}^{\infty}\sum_{m=0}^{\infty}\int_{-\infty}^{\infty}\exp[-it(E-E_0\pm n\omega_{q'}\pm m\omega_{q'})-\tfrac{1}{2}\Gamma|t|]\times\exp(\tfrac{1}{2}Ze^{-\gamma|t|})I_n(Ze^{-\gamma|t|/2})J_m(Ye^{-\gamma|t|/2})\,dt\ . \qquad (12)$$

For $|n\pm m|=0,1,2,\ldots$ Eq. (12) gives the intensity distribution of the 0-, 1-, 2-, etc., phonon-assisted Mössbauer transitions.

Equation (12) is a general expression applicable for the relaxation of both the rf pumped continuum-mode phonons for which $Y\sim 10^{-20}\simeq 0$ and the localized mode with $Y\sim 1$. For several values of $|n\pm m|$, Z, and γ, Eq. (12) has been evaluated numerically using an IBM-1130 computer. The results are shown in Figs. 4–9. In all the cases the natural linewidth Γ was taken to be $7.14\times 10^6\ \text{sec}^{-1}$.

Figure 4 gives the line shape of the zero-phonon Mössbauer line for different values of the phonon-relaxation rate γ. It may be noted that as a result of phonon relaxation the line shape no longer remains a Lorentzian and the intensity of line never becomes negative. The line splitting also does not show up in the present calculations. This conjecture is verified for the line-shape calculation using several values of the Z and Y parameters.

Figure 5 shows the variation of the zero-phonon Mössbauer linewidth with the phonon-relaxation rate. One can see that in most of the cases presented within the framwork of our theory the line gets broadened while the result of others show that the line always gets narrowed under the influence of the phonon relaxation. For $Z\geqslant 2.5$ the width of the line first increases with increasing relaxation rate but after a certain maximum value it starts decreasing with increasing γ and slowly approaches the natural linewidth. The maximum value of linewidth increases with increasing Z but it has no upper limit as obtained by Harris. Around $Z=2.5$ the linewidth becomes equal to the natural linewidth and does not vary with relaxation rate. For $Z<2.5$, the line becomes narrower than the natural linewidth but the maximum narrowing never exceeds $\sim 8\%$ of the natural linewidth. It is interesting to note that the general feature of these curves is similar to those obtained by Harris[8] calculated on the basis of an entirely different formalism.

Figure 6 gives the zero-phonon resonance line intensity as a function of the phonon-relaxation rate γ. For $Z\simeq 2.5$ the intensity of the line remains independent of γ. For other values of Z the intensity either increases or decreases with increasing γ, depending upon whether Z is less than or greater than 2.5. But for a given value of relaxation rate the intensity always decreases with increasing Z which is contrary to the results reported by others. Figures 7 and 8 give the line shape for one-phonon and two-phonon lines. In this case the value of Z is taken to be 2.0. In Fig. 9 the width of the one-phonon Mössbauer line as a function of the phonon relaxation rate is given. From these results and other extensive calculations we conclude that the width of the one-phonon and other higher-phonon lines increases monotonically with the increase of the phonon-relaxation rate. It may be noted further that the relaxation broadening for the phonon-assisted Mössbauer transition is much more pronounced as compared to the zero-phonon Mössbauer transitions.

CONCLUSION

The main results of this work may be summarized as:

(i) A general expression is derived for the intensity distribution of the Mössbauer radiation perturbed by a normal mode. The formalism is applicable for both the rf pumped phonons and the localized mode phonons. The thermal relaxation of phonons affects the Mössbauer line shape with

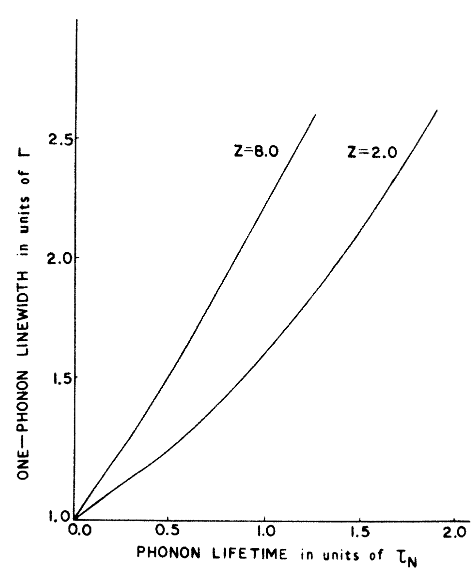

FIG. 9. Linewidth of the one-phonon transition as a function of γ.

the effect being different for the continuum mode and for the localized mode. This result is contrary to the usual expectation.

(ii) Zero-, one-, and two-phonon-assisted Mössbauer line shapes in the presence of the lattice-mode relaxation become non-Lorentzian and the effective linewidths in general are larger than the natural linewidth.

(iii) The splitting reported by Harris of the Mössbauer line owing to the excited lattice-mode relaxation is not seen in our calculations. The splitting[8] may be attributed to the limited applicability of the model proposed by Harris which is discussed in an earlier part of this paper.

---

*Based on the dissertation submitted by Ajay Gupta in partial fulfilment of the Masters degree in Philosophy.

†Junior Research Fellow of the Council of Scientific and Industrial Research, India.

[1]N. D. Heiman, L. Pfeiffer, and J. C. Walker, Phys. Ref. Lett. 21, 93 (1968); G. Asti, G. Albanese, and C. Bucci, Nuovo Cimento B 57, 531 (1968); L. Pfeiffer, N. D. Heiman, and J. C. Walker, Phys. Rev. B 6, 74 (1972); G. Asti, G. Albanese, and C. Bucci, Phys. Rev. 184, 260 (1969).

[2]B. Kaufman and H. J. Lipkin, Ann. Phys. (N.Y.) 18, 294 (1962).

[3]A. Abragam, *L' Effect Mössbauer* (Gordon and Breach, New York, 1964), pp. 22–24.

[4]J. Mishory and D. I. Bolef, *Mössbauer Effect Methodology* (Plenum, New York, 1968), Vol. 4.

[5]See, for example, A. A. Maradudin, Rev. Mod. Phys. 36, 417 (1964).

[6]M. Lax and I. Waller, Phys. Rev. 138, A523 (1965).

[7]J. G. Dash and R. H. Nussbaum, Phys. Rev. Lett. 16, 567 (1966) and *Proceedings of the Fourteenth Colloque Ampere on Nuclear Resonance and Relaxation* (North-Holland, Amsterdam, 1967), pp. 881–902; Rudi H. Nussbaum, *Mössbauer-Effect Methodology* (Plenum, New York, 1966), Vol. 2.

[8]Samuel M. Harris, Phys. Rev. 163, 280 (1967).

[9]K. S. Singwi and A. Sjölander, Phys. Rev. 120, 1093 (1960).

[10]G. Albanese, G. Asti, and S. Rinaldi, Nuovo Cimento B 6, 153 (1971).

[11]G. H. Vineyard, Phys. Rev. 110, 999 (1958).

[12]C. Kittel, *Quantum Theory of Solids* (Wiley, New York, 1964), pp. 394–395.

[13]G. N. Watson, *Theory of Bessel Function* (Cambridge, Mass., 1954), p. 369.

[14]G. J. Perlow, Phys. Rev. 172, 319 (1968).

[15]Ajay Gupta and K. Rama Reddy, Proceedings of the 59th Session of the Indian Science Congress, Part III, 1972 (unpublished), p. 69; Ajay Gupta, Master of Philosophy thesis (Aligarh Muslim University, Aligarh, India, 1973) (unpublished).

[16]Ajay Gupta and K. Rama Reddy, Nucl. Phys. and Solid State Phys. (India) 14C, 277 (1972); an algebric computational error was detected in this symposium report which is rectified in the present work.

[17]M. Lax, Phys. Rev. 129, 3342 (1963).