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Quantum-Mechanical Model of Mössbauer Line Narrowing*

SAMUEL M. HARRIS
Department of Physics, Purdue University, Lafayette, Indiana
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A model for Mössbauer radiation is developed which includes both radiation and lattice-relaxation processes. The model describes an initially excited nucleus in an excited lattice. Both radiation and relaxation phenomena are treated in lowest order, so that a quantum-mechanical solution is possible. It is found that under certain conditions, Mössbauer lines result which are narrower than the natural line, although line broadening is the more usual effect of lattice relaxation. The maximum narrowing in this approximation never exceeds 36% of the natural width.

RECENTLY, Dash and Nussbaum[1] proposed that it might be possible to observe a narrowing of a Mössbauer line due to lattice relaxation phenomena. They consider a radiating Mössbauer nucleus which is vibrating in a long-lived localized mode. It is assumed that the vibrational amplitude is initially greater than the equilibrium value. As time progresses, this amplitude damps down to the equilibrium value with some characteristic relaxation time. Since the recoil-free fraction f depends on the amplitude of vibration, the emission of a recoilless photon becomes relatively more probable as time increases (as the vibration amplitude becomes smaller). They argue that the effect of this relaxation mode is to impose an additional time dependence on the usual amplitude for recoilless photon emission. They predict line shape narrowing in *all cases* and under certain circumstances, the resulting line may attain a limit of zero width (although, unfortunately, with zero intensity).

Although their treatment is simple, it is open to some serious criticism. Since there exist two competing mechanisms, nuclear photon emission and lattice relaxation, it is not clear that one can simply consider the photon to be modulated by the relaxation process as they have done. Also, it is dubious that one can simply use the positive square root of the time-dependent recoil-free fraction when calculating the photon amplitude. In particular, we find that the phase of the recoil-free amplitude (the square of which gives the recoil-free fraction) influences the line shape greatly. We have constructed a simple model which includes both photon emission and lattice-relaxation processes and has the added advantage that it possesses a quantum-mechanical solution.

We consider a nucleus in a two-state lattice. If we allow the nucleus to possess one excited state, then there are a total of four possible states for the system, lattice and nucleus both excited, both in their ground states, etc. For the sake of simplicity, we will consider space to be one dimensional although our conclusions will in no way depend on this restriction. Units are chosen so that $\hbar=c=1$.

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[1] J. G. Dash and R. H. Nussbaum, Phys. Rev. Letters 16, 567 (1966).

The lattice relaxation is handled by introducing the phonon concept. The lattice de-excites by phonon emission which allows energy conservation. We consider a Hamiltonian,

$$H=H_0+H_\gamma+H_\beta. \tag{1}$$

H_0 contains the pure-nuclear, pure-lattice, and free-photon and phonon-radiation-field Hamiltonians. H_γ is the interaction term which leads to photon emission (nuclear de-excitation); H_β leads to phonon emission (lattice relaxation). Eigenstates of H_0 are easily written since H_0 is completely separable into nuclear, lattice, and radiation parts. An eigenstate is represented by a four-product wave function. The operators H_γ and H_β are responsible for transitions between eigenstates of H_0. H_β has nonzero matrix elements between states which differ only in phonon and lattice composition. H_γ matrix elements are nonzero for states which differ in nuclear and photon quantum numbers and may or may not differ in lattice composition. If the lattice quantum number does not change, we are led to recoilless photon emission. If the lattice quantum number changes, we obtain photon emission accompanied by nuclear recoil.

We consider our system to be initially in an eigenstate of H_0 in which both nucleus and lattice are excited with no photons or phonons present. Only those amplitudes will be considered which are of zeroth or first order in phonon and photon coupling constants; we never consider states with more than one photon and/or phonon present. We rationalize this limitation on the basis of the weakness of the coupling constants involved. This approximation is the usual one for photon emission. Since we expect the interference between lattice relaxation and nuclear de-excitation modes to be most significant when the lattice-relaxation time and nuclear lifetime are comparable, it seems consistent to limit ourselves to one-phonon states also.

We must consider six amplitudes which correspond to eigenstates of H_0:

A, both nucleus and lattice excited, state vector $|N^*L^*\rangle$;

B_p, nucleus de-excited, lattice excited, photon present with momentum p, $|NL^*p\rangle$;

C_k, nucleus excited, lattice de-excited, phonon k, $|N^*Lk\rangle$;
D_{pk}, nucleus and lattice de-excited, photon p, phonon k, $|NLpk\rangle$;
F_p, nucleus and lattice de-excited, photon p, $|NLp\rangle$;
G_{pk}, nucleus de-excited, lattice excited, photon p, phonon k, $|NL^*pk\rangle$.

The energy scale is fixed by taking the state $|NL\rangle$ to have zero energy. Nuclear and lattice-excitation energies are denoted by ω_N and ω_L, respectively. The energies associated with a photon of momentum p and a phonon with momentum k are ω_p and ω_k.

We approach this problem via the time-dependent Schrödinger equation,

$$(H_0+H')\Psi(t)=i(\partial/\partial t)\Psi(t). \tag{2}$$

We attempt a solution in terms of eigenstates of H_0,

$$\Psi(t)=\sum_m a_m(t)\varphi_m(t), \tag{3}$$

where

$$H_0\varphi_m(t)=\omega_m\varphi_m(t). \tag{4}$$

Upon substitution of (3) into (2), we obtain a coupled system of differential equations for the amplitudes,

$$i\dot{a}_n(t)=\sum_m a_m(t)\exp[i(\omega_n-\omega_m)t](\varphi_n, H'\varphi_m)+i\delta_{n0}\delta(t). \tag{5}$$

In (5), the time dependence of the matrix element has been explicitly displayed. The inhomogeneous term expresses the boundary condition that $a_n(0)=\delta_{n0}$; at $t=0$, the system is in the pure state denoted by the index 0. H' is the operator that leads to transitions between eigenstates of H_0.

It is simpler to use the Fourier transform of (5) than the differential equation itself.[2,3] If we introduce the Fourier transformed amplitudes $a_n(\omega)$ by

$$a_n(t)=-(2\pi i)^{-1}\int_{-\infty}^{\infty}d\omega\exp[i(\omega_n-\omega)t]a_n(\omega), \tag{6}$$

then Eq. (5) becomes

$$(\omega-\omega_n+i\epsilon)a_n(\omega)=\sum_m a_m(\omega)(\varphi_n, H'\varphi_m)+\delta_{n0}. \tag{7}$$

The introduction of $+i\epsilon$ in (7) insures that the system will display the correct behavior for $t<0$ and leads to the proper causality conditions.

This formalism is easily applied to the current problem. We obtain six coupled equations using (7),

$$\begin{aligned}(\omega-\omega_N-\omega_L+i\epsilon)A(\omega)&=1+\sum_p B_p(\omega)\langle N^*L^*|H_\gamma|NL^*p\rangle\\&+\sum_k C_k(\omega)\langle N^*L^*|H_\beta|N^*Lk\rangle\\&+\sum_p F_p(\omega)\langle N^*L^*|H_\gamma|NLp\rangle,\end{aligned} \tag{8a}$$

$$\begin{aligned}(\omega-\omega_L-\omega_p+i\epsilon)B_p(\omega)&=A(\omega)\langle NL^*p|H_\gamma|N^*L^*\rangle\\&+\sum_k D_{pk}(\omega)\langle NL^*p|H_\beta|NLpk\rangle,\end{aligned} \tag{8b}$$

$$\begin{aligned}(\omega-\omega_N-\omega_k+i\epsilon)C_k(\omega)&=A(\omega)\langle N^*Lk|H_\beta|N^*L^*\rangle\\&+\sum_p D_{pk}(\omega)\langle N^*Lk|H_\gamma|NLpk\rangle\\&+\sum_p G_{pk}(\omega)\langle N^*Lk|H_\gamma|NL^*pk\rangle,\end{aligned} \tag{8c}$$

$$\begin{aligned}(\omega-\omega_p-\omega_k+i\epsilon)D_{pk}(\omega)&=B_p(\omega)\langle NLpk|H_\beta|NL^*p\rangle\\&+C_k(\omega)\langle NLpk|H_\gamma|N^*Lk\rangle,\end{aligned} \tag{8d}$$

$$\begin{aligned}(\omega-\omega_p+i\epsilon)F_p(\omega)&=A(\omega)\langle NLp|H_\gamma|N^*L^*\rangle\\&+\sum_k G_{pk}(\omega)\langle NLp|H_\beta|NL^*pk\rangle,\end{aligned} \tag{8e}$$

$$\begin{aligned}(\omega-\omega_L-\omega_p-\omega_k+i\epsilon)G_{pk}(\omega)&=C_k(\omega)\langle NL^*pk|H_\gamma|N^*Lk\rangle\\&+F_p(\omega)\langle NL^*pk|H_\beta|NLp\rangle.\end{aligned} \tag{8f}$$

These equations can be written more concisely if we introduce the following definitions:

$$\langle Np|H_\gamma|N^*\rangle=\gamma, \tag{9a}$$

$$\langle Lk|H_\beta|L^*\rangle=\beta, \tag{9b}$$

$$\langle NL^*p|H_\gamma|N^*L^*\rangle=f_{11}\gamma, \tag{9c}$$

$$\langle NLp|H_\gamma|N^*L^*\rangle=f_{12}\gamma, \tag{9d}$$

$$\langle NLp|H_\gamma|N^*L\rangle=f_{22}\gamma. \tag{9e}$$

Here γ and β are the photon and phonon radiative matrix elements, respectively. The square of the magnitude of f_{11} and f_{22} yields the recoilless fractions for photon emission from lattice states L^* and L. The square of the magnitude of f_{12} yields the fraction of off-resonance photons (emission accompanied by nuclear recoil).

It is shown in Appendix A that f_{11} and f_{22} are real if the lattice wave functions are eigenstates of parity. Furthermore, we have

$$f_{21}=\pi f_{12}, \tag{10}$$

where π is the product of the parities associated with L and L^*. If L and L^* have the same (different) parity, then $\pi=+1(-1)$. f_{12} can always be made real by a proper choice of relative phases of lattice wave functions. Assuming that the matrix elements do not depend strongly on the momenta p and k, we can take

[2] W. Heitler, *The Quantum Theory of Radiation* (Oxford University Press, New York, 1954).
[3] S. M. Harris, Phys. Rev. **124**, 1178 (1961).

them outside of the summations, so that (8) becomes

$$(\omega-\omega_N-\omega_L+i\epsilon)A=1+\gamma^*f_{11}\sum_p B_p+\beta^*\sum_k C_k+\gamma^*f_{12}\sum_p F_p, \quad (11a)$$

$$(\omega-\omega_L-\omega_p+i\epsilon)B_p=A\gamma f_{11}+\beta^*\sum_k D_{pk}, \quad (11b)$$

$$(\omega-\omega_N-\omega_k+i\epsilon)C_k=A\beta+\gamma^*f_{22}\sum_p D_{pk}+\gamma^*f_{12}\pi\sum_p G_{pk}, \quad (11c)$$

$$(\omega-\omega_p-\omega_k+i\epsilon)D_{pk}=\beta B_p+\gamma f_{22}C_k, \quad (11d)$$

$$(\omega-\omega_p+i\epsilon)F_p=\gamma f_{12}A+\pi\beta\sum_k G_{pk}, \quad (11e)$$

and

$$(\omega-\omega_L-\omega_p-\omega_k+i\epsilon)G_{pk}=\gamma f_{12}\pi C_k+\beta^*\pi F_p. \quad (11f)$$

Everywhere G_{pk} occurs, it is preceded by π so that we can redefine this amplitude $\pi G_{pk}\to G_{pk}$. Making this substitution, π explicitly disappears from our set of coupled equations.

These equations can be handled in a manner similar to the treatment used in our earlier discussion of time-dependent Mössbauer transmission.[3] Details of the solution of (11a)–(11f) are given in Appendix B. The results are

$$A(\omega)=[\omega-\omega_N-\omega_L+\tfrac{1}{2}i(\Gamma_\gamma+\Gamma_\beta)]^{-1}, \quad (12a)$$

$$B_p(\omega)=\gamma f_{11}[\omega-\omega_L-\omega_p+\tfrac{1}{2}i\Gamma_\beta]^{-1}\times[\omega-\omega_N-\omega_L+\tfrac{1}{2}i(\Gamma_\gamma+\Gamma_\beta)]^{-1}, \quad (12b)$$

$$C_k(\omega)=\beta[\omega-\omega_N-\omega_k+\tfrac{1}{2}i\Gamma_\gamma]^{-1}\times[\omega-\omega_N-\omega_L+\tfrac{1}{2}i(\Gamma_\gamma+\Gamma_\beta)]^{-1}, \quad (12c)$$

$$D_{pk}(\omega)=(\beta B_p(\omega)+\gamma f_{22}C_k(\omega))[\omega-\omega_p-\omega_k+i\epsilon]^{-1}, \quad (12d)$$

$$F_p(\omega)=\gamma f_{12}[\omega-\omega_p+\tfrac{1}{2}i\Gamma_\beta]^{-1}\times[\omega-\omega_N-\omega_L+\tfrac{1}{2}i(\Gamma_\gamma+\Gamma_\beta)]^{-1}, \quad (12e)$$

and

$$G_{pk}(\omega)=(\gamma f_{12}C_k(\omega)+\beta^*F_p(\omega))[\omega-\omega_L-\omega_p-\omega_k+i\epsilon]^{-1}. \quad (12f)$$

In the above, Γ_γ and Γ_β are the natural radiative and lattice linewidths. The nuclear lifetime is $\tau_\gamma=1/\Gamma_\gamma$. The lattice-relaxation time is $\tau_\beta=1/\Gamma_\beta$. We are interested in the Fourier transforms of these amplitudes at large time ($t\to\infty$). The time-dependent amplitudes may be calculated from (6) using (12.a)–(12.f). Thus, for example, $|A(t\to\infty)|^2$ is the probability of finding the state $|N^*L^*\rangle$ populated at large times. All Fourier transforms can be evaluated easily by calculating residues, since only simple poles enter into our integrals.

Looking at (6), we see that at $t\to+\infty$ only those terms will survive for which $(\omega_n-\omega)$ does not contain a positive imaginary part. Thus a pole for which ω has a negative imaginary part will not contribute to $a_n(t\to\infty)$. If we return to (12), we see that all poles in A, B_p, C_k, and F_p lie in the lower half-plane, so that

$$A(t\to\infty)=B_p(t\to\infty)=C_k(t\to\infty)=F_p(t\to\infty)=0. \quad (13)$$

The amplitudes D_{pk} and G_{pk} each contain one pole in the lower half-plane which moves up to the real axis as $\epsilon\to 0$. These amplitudes will therefore remain finite as $t\to\infty$.

To obtain the time dependence of D, we must evaluate

$$D_{pk}(t)=-(2\pi i)^{-1}\int_{-\infty}^{\infty}d\omega\times\frac{\exp[it(\omega_p+\omega_k-\omega)]}{(\omega-\omega_p-\omega_k+i\epsilon)(\omega-\omega_N-\omega_L+\frac{1}{2}i\Gamma_\gamma+\frac{1}{2}i\Gamma_\beta)}\times\beta\gamma\left[\frac{f_{11}}{(\omega-\omega_L-\omega_p+\frac{1}{2}i\Gamma_\beta)}+\frac{f_{22}}{(\omega-\omega_N-\omega_k+\frac{1}{2}i\Gamma_\gamma)}\right]. \quad (14)$$

All poles are in the lower half-plane. For $t>0$, we must close our contour of integration in the lower half-plane. [For $t<0$, we must close our contour in the upper half-plane which contains no poles, so that $D(t<0)=0$, a requirement of causality.] At large time, only the pole at $\omega=\omega_p+\omega_k-i\epsilon$ which is very close to the real axis can contribute. The result, after performing the integration specified in (14), is

$$D_{pk}(t\to\infty)=\beta\gamma(\omega_p+\omega_k-\omega_N-\omega_L+\tfrac{1}{2}i\Gamma_T)^{-1}\times[f_{11}(\omega_k-\omega_L+\tfrac{1}{2}i\Gamma_\beta)^{-1}+f_{22}(\omega_p-\omega_N+\tfrac{1}{2}i\Gamma_\gamma)^{-1}] \quad (15)$$

where $\Gamma_T=\Gamma_\gamma+\Gamma_\beta$ is the total width for decay from the initial state. The probability of finding the nucleus and lattice both de-excited at large time with one photon p and one phonon k present is given by the square of the amplitude in (15). Since we only observe the photon, not the phonon, we must sum over all unobserved phonon states, k. The same techniques are used in performing the k sums here as in Appendix B. The result is

$$\sum_k|D_{pk}(\infty)|^2=\frac{\Gamma_\gamma}{2L(\Gamma_\gamma+\Gamma_\beta)}\times\left\{\frac{f_{22}(\Gamma_\gamma f_{11}+\Gamma_\beta f_{22})}{(\omega_p-\omega_N)^2+\frac{1}{4}\Gamma_\gamma^2}+\frac{(f_{11}-f_{22})f_{11}(\Gamma_\gamma+2\Gamma_\beta)}{(\omega_p-\omega_N)^2+\frac{1}{4}(\Gamma_\gamma+2\Gamma_\beta)^2}\right\}. \quad (16)$$

The spectrum given in (16) is the sum of two Lorentzians centered about the natural frequency ω_N. The first term gives a line of width Γ_γ, the natural linewidth. The second line is broadened to $\Gamma_\gamma+2\Gamma_\beta$ by the effect of lattice relaxation. This result can be written more concisely if we introduce the parameters $a=\Gamma_\beta/\Gamma_\gamma=\tau_\gamma/\tau_\beta$, $x=(\omega_p-\omega_N)/\Gamma_\gamma$, and $\alpha=f_{11}/f_{22}$,

$$\mathcal{L}(x)=\sum_k |D_{pk}(\infty)|^2=\frac{f_{22}^2}{2\Gamma_\gamma L}(1+a)^{-1}\times\left\{\frac{(a+\alpha)}{x^2+\frac{1}{4}}+\frac{\alpha(\alpha-1)(1+2a)}{x^2+\frac{1}{4}(1+2a)^2}\right\}. \quad (17)$$

In a similar manner, we can evaluate the photon distribution due to G_{pk},

$$\sum_k |G_{pk}(t\rightarrow\infty)|^2=\left(\frac{\Gamma_\gamma\Gamma_\beta}{\Gamma_\gamma+\Gamma_\beta}\right)\frac{f_{12}^2}{2L}\times\{[(\omega_p-\omega_N+\omega_L)^2+\tfrac{1}{4}\Gamma_\gamma^2]^{-1}+[(\Gamma_\gamma+2\Gamma_\beta)/\Gamma_\beta]\times[(\omega_p-\omega_N-\omega_L)^2+\tfrac{1}{4}(\Gamma_\gamma+2\Gamma_\beta)^2]^{-1}\}. \quad (18)$$

This is the probability that the nucleus is de-excited, lattice excited, photon p, and an unobserved phonon present at large time. Since we have limited ourselves to one phonon processes, there is no mechanism available for the lattice to relax back to its ground state. The amplitude G yields two lines centered about $\omega_p=\omega_N\pm\omega_L$. These are the nonresonant lines corresponding to one-phonon transitions. In addition, there is a cross term present in (18) which we have dropped. This latter contribution will be small if the two lines included in (18) do not appreciably overlap.

Returning now to the expression for resonant emission, (17), we see that the special case of $\alpha=1$ leads to an especially simple result,

$$\mathcal{L}(x)=(f_{22}^2/2\Gamma_\gamma L)(x^2+\tfrac{1}{4})^{-1} \qquad (\text{for } \alpha=1). \quad (19)$$

This is just the natural line shape which would be obtained in the absence of lattice relaxation ($\Gamma_\beta=a=0$). Similarly, we obtain the natural line shape if $a=\infty$. This case corresponds to immediate lattice relaxation, so that resonant photon emission always occurs while the lattice is in the ground state. For other combinations of a and α, we can obtain lines which are either narrower or wider than the natural line.[4]

For a true two-level lattice, we must have $|f_{11}|=|f_{22}|$ (Appendix A), so that $\alpha=\pm1$. In the previous paragraph, we saw that $\alpha=+1$ leads to the natural line shape. For the other possibility, $\alpha=-1$, the line is always broadened. The line shape is plotted as a function of x for various values of a ($\alpha=-1$) in Fig. 1.

[4] By linewidth, we mean the maximum width of the line between points with amplitude equal to half of the central amplitude. Because our line shape is in general not a simple Lorentzian, alternative definitions may be used. We could have used the maximum width between points with amplitude equal to half of the maximum amplitude (since in some cases maximum amplitude does not occur at the central frequency) or the width which includes half of the area under the spectral curve. For a pure Lorentzian, all of these definitions yield the same width. The choice of definition may alter our numerical values slightly, but the qualitative results do not depend on definition.

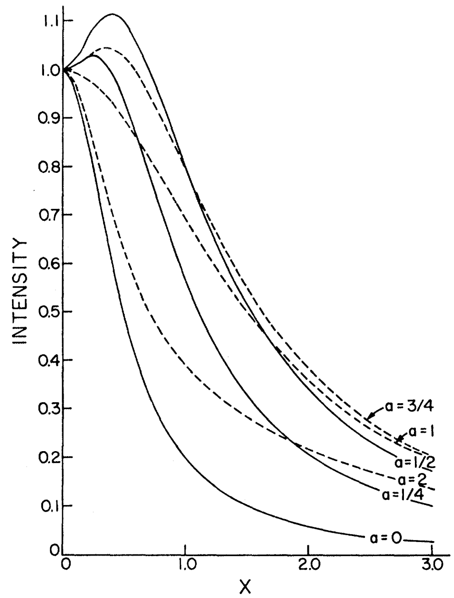

FIG. 1. Resonant line shape as a function of $x=(\omega-\omega_N)/\Gamma_\gamma$ for various values of a ($\alpha=-1$). All curves are normalized to have the same value at $x=0$.

The curve for $a=0$ corresponds to the natural line shape of width Γ_γ. For $0<a<1$, the line has a double hump with a minimum at $x=0$. The case $a=1$ corresponds to a pure Lorentzian of width $3\Gamma_\gamma$. For $a>1$, we obtain a non-Lorentzian line peaked at $x=0$ (only one maximum). The linewidth varies between Γ_γ and $3.3\Gamma_\gamma$. In this case, the line can only be broadened by lattice relaxation.

If we were to relax our conditions on the matrix elements so that $|f_{11}|\neq|f_{22}|$ would be allowed, then our formalism could predict narrowing under certain conditions. We would expect $|f_{11}|\leq|f_{22}|$, since the recoilless fraction should be larger for states of lower excitation. We would then be interested in cases for which $|\alpha|\leq1$. The linewidth as a function of a is plotted for various values of α in Fig. 2. It is apparent that narrowing is only possible for $\alpha>0$. The minimum linewidth possible is $0.869\Gamma_\gamma$, which occurs for $\alpha=0.238$ and $a=0.108$. Thus even though narrowing is possible, it is a small effect, never larger than about 13%. The condition $|f_{11}|\neq|f_{22}|$ might arise in the multilevel lattice where only two levels are important for resonant emission, so that our formalism still applies.

A calculation of the multilevel lattice has been performed in the same approximation as above, namely, considering only amplitudes up to first order in photon and phonon emission. The calculation and results are similar to the two-level case. The resonant line shape is given by the sum of two Lorentzians,

$$\mathcal{L}(x)=\frac{A}{x^2+\frac{1}{4}}+\frac{B}{x^2+\frac{1}{4}(1+2a)^2}, \quad (20)$$

in analogy to (17). The coefficients A and B are given as complicated sums of products of photon and phonon matrix elements between all permissible levels and are strongly model-dependent. In this case, a is the ratio of total phonon width to photon width.

If A, B, and a in (20) are treated as independent parameters subject to the constraints that $\mathcal{L}(x)$ is positive definite and $a>0$, then one finds narrowing only when $A>0$ and $B<0$. The minimum obtainable linewidth is $0.644\Gamma_\gamma$, a reduction of 36%. Note that this narrowing is greater than the optimal 13% effect for the two-level system. In the latter case, A and B were determined by a and α, so that A, B, and a could not be varied independently. Also, the condition $|\alpha|\leq 1$ restricts the possible value of A and B for given a. Consequently, it is possible that slightly sharper lines will result for the multilevel system, but narrowing should not exceed 36%. The effect of higher-order phonon processes (multiple phonon emission) is difficult to estimate. Models which can deal with this question are now being considered.

APPENDIX A

The amplitude for photon emission (momentum p) accompanied by a lattice transition from state $i\rightarrow j$ is proportional to[5]

$$f_{ij}=\langle j\,|\,e^{-ipx}\,|\,i\rangle. \qquad \text{(A1)}$$

If we assume that the lattice states are eigenstates of parity, then

$$P\,|\,i\rangle=\pi_i\,|\,i\rangle, \qquad \pi_i=\pm 1. \qquad \text{(A2)}$$

This situation would exist in our simple model where we consider the lattice to be one nucleus bound in a potential well. Using (A1) and (A2), we obtain

$$\begin{aligned} f_{ij}^* &= \langle j\,|\,e^{-ipx}\,|\,i\rangle^* \\ &= \langle i\,|\,e^{ipx}\,|\,j\rangle \\ &= \langle i\,|\,P^{-1}e^{-ipx}P\,|\,j\rangle \\ &= \pi_i\pi_j\langle i\,|\,e^{-ipx}\,|\,j\rangle \\ &= \pi_i\pi_j f_{ji}. \end{aligned} \qquad \text{(A3)}$$

If we consider diagonal components, then $i=j$, and

$$f_{ii}^*=f_{ii}; \qquad \text{(A4)}$$

the diagonal amplitudes are real.

Using the completeness relation, $\sum_m |\,m\rangle\,\langle m\,|=1$, we can show that

$$\begin{aligned} \sum_m f_{im}^* f_{jm} &= \sum_m \langle m\,|\,e^{-ipx}\,|\,i\rangle^*\,\langle m\,|\,e^{-ipx}\,|\,j\rangle \\ &= \sum_m \langle i\,|\,e^{ipx}\,|\,m\rangle\,\langle m\,|\,e^{-ipx}\,|\,j\rangle \\ &= \delta_{ij}. \end{aligned} \qquad \text{(A5)}$$

For the case of a two-level lattice (A5) yields three relations among the f's,

$$\begin{aligned} f_{11}^2+f_{12}^2 &= 1, \\ f_{22}^2+f_{12}^2 &= 1, \end{aligned} \qquad \text{(A6)}$$

and

$$f_{12}(\pi_1\pi_2 f_{11}+f_{22})=0.$$

In the above, we have used the result that f_{11} and f_{22} are real and that phases can be chosen such that f_{12} is also real. If $f_{12}=0$ (only recoilless emission is possible), then (A6) has a solution f_{11}, $f_{22}=\pm 1$. If $f_{12}\neq 0$, then $f_{22}=-\pi_1\pi_2 f_{11}$. Thus in both cases we have

$$|\,f_{11}\,|=|\,f_{22}\,|. \qquad \text{(A7)}$$

APPENDIX B

We will solve the set of Eqs. (11a)–(11f). We use (11d) to eliminate the amplitude D_{pk} from the remaining equations. From (11d), we get

$$D_{pk}=(\omega-\omega_p-\omega_k+i\epsilon)^{-1}[\beta B_p+\gamma f_{22}C_k]. \qquad \text{(B1)}$$

Substituting (B1) into (11b), we obtain

$$(\omega-\omega_L-\omega_p+i\epsilon)B_p = A\gamma f_{11}+\beta^* \sum_k (\omega-\omega_p-\omega_k+i\epsilon)^{-1}[\beta B_p+\gamma f_{22}C_k]. \qquad \text{(B2)}$$

B_p is independent of k so that the first sum is simply

$$\sum_k (\omega-\omega_p-\omega_k+i\epsilon)^{-1}.$$

This sum can be handled by converting the sum to an integral (plane waves normalized in a length L)

$$\begin{aligned} \sum_k (\omega-\omega_p-\omega_k+i\epsilon)^{-1} &= \frac{L}{2\pi}\int_{-\infty}^{\infty} dk(\omega-\omega_p-\omega_k+i\epsilon)^{-1} \\ &= \frac{L}{\pi}\int_0^{\infty} dk(\omega-\omega_p-\omega_k+i\epsilon)^{-1} \\ &= \frac{L}{\pi}\int_0^{\infty} d\omega_k(\omega-\omega_p-\omega_k+i\epsilon)^{-1} \\ &\approx \frac{L}{\pi}\int_{-\infty}^{\infty} d\omega_k(\omega-\omega_p-\omega_k+i\epsilon)^{-1} \\ &= \frac{L}{\pi}\int_{-\infty}^{\infty} d\omega_k[\mathcal{P}(\omega-\omega_p-\omega_k)^{-1}-i\pi\delta(\omega-\omega_p-\omega_k)]. \end{aligned} \qquad \text{(B3)}$$

The principal value integral is usually small and contributes a line shift which will be neglected. Thus we get

$$\sum_k (\omega-\omega_p-\omega_k+i\epsilon)^{-1}\approx -iL. \qquad \text{(B4)}$$

Using (B4), we can write (B2) as

$$(\omega-\omega_L-\omega_p+\tfrac{1}{2}i\Gamma_\beta)B_p = A\gamma f_{11}+\beta^*\gamma f_{22}\sum_k (\omega-\omega_p-\omega_k+i\epsilon)^{-1}C_k, \qquad \text{(B5)}$$

[5] W. E. Lamb, Jr., Phys. Rev. 55, 190 (1939).

where $\Gamma_\beta = 2L|\beta|^2$ is the rate for lattice relaxation from the initial state.

In a similar manner, we can use (11f) to eliminate G_{pk} from the equations with the result

$$(\omega-\omega_N-\omega_k+\tfrac{1}{2}i\Gamma_\gamma)C_k = A\beta + f_{22}\gamma^*\beta \sum_p (\omega-\omega_p-\omega_k+i\epsilon)^{-1}B_p + f_{12}\gamma^*\beta^* \sum_p (\omega-\omega_L-\omega_p-\omega_k+i\epsilon)^{-1}F_p \quad \text{(B6)}$$

and

$$(\omega-\omega_p+\tfrac{1}{2}i\Gamma_\beta)F_p = \gamma f_{12}A + \beta\gamma f_{12}^* \sum_k (\omega-\omega_L-\omega_p-\omega_k+i\epsilon)^{-1}C_k. \quad \text{(B7)}$$

In obtaining (B6) we have used the result $f_{22}^2+f_{12}^2=1$, so that only the total radiation width Γ_γ appears.

In calculating $A(\omega)$, we require $\sum_p F_p$, $\sum_k C_k$, and $\sum_p B_p$. Using (B7), we get

$$\sum_p F_p = \sum_p (\omega-\omega_p+\tfrac{1}{2}i\Gamma_\beta)^{-1} \times [\gamma f_{12}A + \beta\gamma f_{12}^* \sum_k (\omega-\omega_L-\omega_p-\omega_k+i\epsilon)^{-1}C_k]. \quad \text{(B8)}$$

The only p dependence in the bracketed term in (B8) occurs in the energy denominator $(\omega-\omega_L-\omega_p-\omega_k+i\epsilon)^{-1}$. The first term can be summed in analogy with (B4),

$$\sum_p (\omega-\omega_L-\omega_p-\omega_k+i\epsilon)^{-1} \approx -iL. \quad \text{(B9)}$$

The second sum over p can be performed by reducing to partial fractions,

$$\sum_p (\omega-\omega_p+\tfrac{1}{2}i\Gamma_\beta)^{-1}(\omega-\omega_L-\omega_p-\omega_k+i\epsilon)^{-1} = (\omega_L+\omega_k+\tfrac{1}{2}i\Gamma_\beta)^{-1} \sum_p [-(\omega-\omega_p+\tfrac{1}{2}i\Gamma_\beta)^{-1} + (\omega-\omega_L-\omega_p-\omega_k+i\epsilon)^{-1}] \approx 0, \quad \text{(B10)}$$

since both sums are approximately equal.

We can evaluate (B8) using (B9) and (B10),

$$\sum_p F_p = -iL\gamma f_{12}A. \quad \text{(B11)}$$

Similarly, we obtain

$$\sum_k C_k = -iL\beta A \quad \text{(B12)}$$

and

$$\sum_p B_p = -iL\gamma f_{11}A. \quad \text{(B13)}$$

Substituting (B11)–(B13) into (11a), we obtain a solution for the amplitude A,

$$A(\omega) = (\omega-\omega_N-\omega_L+\tfrac{1}{2}i\Gamma_\gamma+\tfrac{1}{2}i\Gamma_\beta)^{-1}. \quad \text{(B14)}$$

The decay rate from the initial state is simply the sum of the rates for radiative and relaxation processes.

To calculate C_k from (B6), we need sums like

$$\sum_p (\omega-\omega_p-\omega_k+i\epsilon)^{-1}B_p$$

and

$$\sum_p (\omega-\omega_L-\omega_p-\omega_k+i\epsilon)^{-1}F_p.$$

The first of these can be rewritten using (B5),

$$\sum_p (\omega-\omega_p-\omega_k+i\epsilon)^{-1}(\omega-\omega_L-\omega_p+\tfrac{1}{2}i\Gamma_\beta)^{-1} \times [A\gamma f_{11} + \beta^*\gamma f_{22} \sum_k (\omega-\omega_p-\omega_k+i\epsilon)^{-1}C_k]. \quad \text{(B15)}$$

The first term vanishes by (B10). The second sum over p is a sum over a product of three energy denominators which can again be handled by partial fractions and also is equal to zero. Thus the term (B15) does not contribute to C_k. A similar treatment yields a similar result for $\sum_p (\omega-\omega_L-\omega_p-\omega_k+i\epsilon)^{-1}F_p$.

Setting the last two terms in (B6) equal to zero, we obtain

$$C_k(\omega) = (\omega-\omega_N-\omega_k+\tfrac{1}{2}i\Gamma_\gamma)^{-1}A\beta = (\omega-\omega_N-\omega_k+\tfrac{1}{2}i\Gamma_\gamma)^{-1} \times (\omega-\omega_N-\omega_L+\tfrac{1}{2}i\Gamma_\gamma+\tfrac{1}{2}i\Gamma_\beta)^{-1}\beta. \quad \text{(B16)}$$

Treating the remaining amplitudes in the same way we obtain the solutions (12a)–(12f).

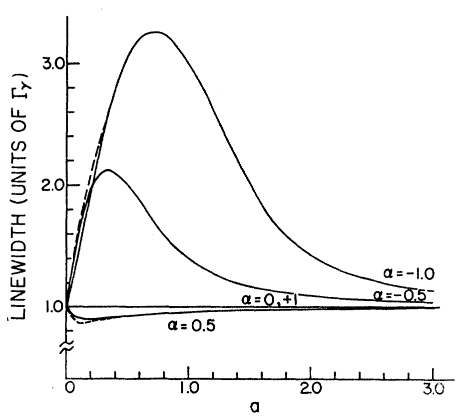

FIG. 2. Linewidth (in units of Γ_γ) as a function of a for various values of α, $1 \geq \alpha \geq -1$. The dashed curves indicate the envelope for all accessible values of linewidth.