where we have used $\langle S,M_S|S_+S_-|S,M_S\rangle=\frac{1}{2}(S+M_S)\times(S-M_S+1)$. Also

$$\begin{aligned}\langle S_+F_{-4}F_5F_{-5}F_4S_-\rangle &=\tfrac{1}{2}\langle S_+S_-\rangle\langle F_{-4}(H_2+H_1)F_4\rangle\\ &=\tfrac{1}{2}\langle S_+S_-\rangle\langle(H_2+H_1+2)F_{-4}F_4\rangle\\ &=\tfrac{1}{4}(S+M_S)(S-M_S+1)(\epsilon_0+K+2)\\ &\qquad\times(\tfrac{1}{2}\mu-\tfrac{1}{2}K)(\tfrac{1}{2}\mu+\tfrac{1}{2}K+1). \qquad (A3)\end{aligned}$$

In the last line we have used the fact that $-(\sqrt{\frac{1}{2}})F_{\pm 4}$ acts like $J_{\pm}$ in a space with "angular momentum" $J=\frac{1}{2}\mu$, and $M_J=\frac{1}{2}K$.[10]

Similarly,

$$\langle S_-F_5F_{-5}S_+\rangle=\tfrac{1}{4}(S-M_S)(S+M_S+1)(\epsilon_0+K), \qquad (A4)$$

$$\begin{aligned}\langle S_-F_4F_1F_{-1}F_{-4}S_+\rangle &=\tfrac{1}{4}(S-M_S)(S+M_S-1)(\epsilon_0\epsilon K+2)\\ &\qquad\times(\tfrac{1}{2}\mu+\tfrac{1}{2}K)(\tfrac{1}{2}\mu-\tfrac{1}{2}K+1). \qquad (A5)\end{aligned}$$

There are two off-diagonal terms:

$$\begin{aligned}\langle S_+F_1F_{-5}F_4S_-\rangle=\langle S_+S_-\rangle\langle F_{-4}F_{+4}\rangle &=\tfrac{1}{2}(S+M_S)(S-M_S+1)\\ &\qquad\times(\tfrac{1}{2}\mu-\tfrac{1}{2}K)(\tfrac{1}{2}\mu+\tfrac{1}{2}K+1), \qquad (A6)\end{aligned}$$

$$\begin{aligned}\langle S_-F_5F_{-1}F_{-4}S_+\rangle &=\tfrac{1}{2}(S-M_S)(S+M_S+1)\\ &\qquad\times(\tfrac{1}{2}\mu+\tfrac{1}{2}K)(\tfrac{1}{2}\mu-\tfrac{1}{2}K+1). \qquad (A7)\end{aligned}$$

The inhomogeneous terms, which come from the left side of (6.6), must be calculated separately for each case, although the commutors may be used to reduce the operators:

$$\begin{aligned}\langle S_+F_1\textstyle\sum_i F_{-1}(i)S_-(i)\rangle &=\tfrac{1}{2}\langle S_+\textstyle\sum_i[H_2(i)-H_1(i)]S_-(i)\rangle,\\ \langle S_-F_5\textstyle\sum_i F_{-5}(i)S_+(i)\rangle &=\tfrac{1}{2}\langle S_-\textstyle\sum_i[H_2(i)+H_1(i)]S_+(i)\rangle,\\ \langle S_+F_{-4}F_5\textstyle\sum_i F_{-1}(i)S_-(i)\rangle &=\langle S_+F_{-4}\textstyle\sum_i F_4(i)S_-(i)\rangle,\\ \langle S_-F_4F_1\textstyle\sum_i F_{-5}(i)S_+(i)\rangle &=\langle S_-F_4\textstyle\sum_i F_{-4}(i)S_+(i)\rangle.\end{aligned} \qquad (A8)$$

The right sides of these equations are calculated by using the determinant forms (Table II) and (6.7).

PHYSICAL REVIEW VOLUME 124, NUMBER 4 NOVEMBER 15, 1961

Quantum Mechanical Calculation of Mössbauer Transmission*†

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(Received June 26, 1961)

A quantum mechanical calculation of the time-dependent Mössbauer transmission has been performed neglecting solid-state effects. The source considered consists of nuclei which decay via a two-photon cascade, the second of which is emitted without recoil and is subject to resonant absorption by a foil whose resonance may be shifted due to a small relative velocity between source and absorber. The transmission is obtained when the transmitted recoiless photon is measured in coincidence with the first photon of the cascade. The result is in agreement with that obtained by considering the absorber as a classical dielectric slab capable of absorption and dispersion. The initial condition has been investigated in detail by considering the full cascade. In this manner, one sees that the usual simple assumption that the nucleus is in the first excited state immediately after the emission of the first photon, gives the correct boundary condition.

INTRODUCTION

THE most common Mössbauer experiment is performed by measuring the transmission of recoiless radiation through a thin resonant absorber which may be in motion relative to the source. In this manner, the hyperfine structure of the isotope employed may be investigated.[1] An interesting variation of this simple experiment has been performed by several groups.[2–5] They make use of the most popular Mössbauer isotope, Fe^{57}. The source contains Co^{57} which decays by electron capture to Fe^{57m} which decays in turn by a 122-kev photon followed by a

* Work partially supported by joint contract with the Office of Naval Research and the U. S. Atomic Energy Commission.
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14.4-kev photon (competing with internal conversion) to the ground state. The 14.4-kev photon is the one which is emitted without recoil about 60% of the time even at room temperature.[6] These investigators measure the transmission of the 14.4-kev photon through a resonant absorber foil. The recoiless photons, however, are counted in delayed coincidence with the 122-kev photons. The spectrum of transmitted radiation gives information on the temporal aspects of the emission and absorption processes involved, with well-defined initial conditions. In this paper, an attempt is made to calculate the line shape (transmission vs absorber velocity) of the transmitted radiation as a function of the delay time, employing the principles of quantum mechanics.

The experimental results are in general agreement with the theory developed here. For extremely short times, the transmitted line is greatly broadened. As the delay time grows longer, the line develops a damped oscillatory behavior, the central peak of which approaches the natural linewidth. For very long times, the oscillations become quite rapid, and the central peak becomes narrower than the natural line while the area under the other peaks becomes more significant. One extremely interesting point is that for certain delays and relative velocities the transmission is greater than it would be if the absorber were not present. Thus, the resonant absorber seems to produce a time bunching of photons.

Hamermesh[7] has performed a simple classical calculation of this effect. He assumes that the source and absorber are composed of a collection of single level damped harmonic oscillators with natural frequency ω_0 and damping factor Γ. The radiation field emitted by the source can be represented as a damped electric field,

$$a(t)=\exp[i(\omega_0+i\Gamma/2)t]. \tag{1}$$

If this field is Fourier decomposed, each frequency component sees a different index of refraction $n(\omega)$ as it traverses the absorber. The standard expression for the dielectric constant ϵ ($\epsilon=n^2$) of such a collection of damped oscillators is

$$\epsilon(\omega)=1+r(\omega_0'^2-\omega^2+i\omega\Gamma)^{-1}. \tag{2}$$

In the above, ω_0' is the resonant frequency of each absorber nucleus, and r is a constant which depends on the density and properties of the absorber nuclei. In this manner, the Doppler shift due to the possible relative motion of source and absorber is taken into account. The frequency distribution of the transmitted field $a'(\omega)$ differs markedly from $a(\omega)$, the field leaving the source, each component being altered by a frequency-dependent phase change due to the presence of the absorber of thickness d,

$$a'(\omega)=a(\omega)\exp[i\omega d n(\omega)]. \tag{3}$$

The inverse Fourier transform of (3) is $a'(t)$, the square of the magnitude of which is the transmission,

$$T(t)=|a'(t)|^2=e^{-\Gamma t}\left|\sum_{m=0}^{\infty}(4i\Delta\omega/\beta\Gamma)^m(\tfrac{1}{4}\beta\Gamma t)^{m/2}\times J_m[(\beta\Gamma t)^{\frac{1}{2}}]\right|^2. \tag{4}$$

In the above, we have $\Delta\omega=\omega_0-\omega_0'$, β is given by $\beta=-(rd/\Gamma)$, and $J_m[(\beta\Gamma t)^{\frac{1}{2}}]$ is the usual Bessel function of order m. Curves of the transmission have been plotted by Hamermesh for various values of the parameters β and $(\Delta\omega/\Gamma)$.

QUANTUM MECHANICAL APPROACH

The time development of a quantum system is determined by its Hamiltonian. In what is to follow, we separate the Hamiltonian into two parts, H_0, which includes the nuclear effects and the free radiation field, and H, the interaction term which is responsible for transitions between the pure states $|\varphi_p\rangle$ of the free Hamiltonian. The true state $|\psi\rangle$ (belonging to the total Hamiltonian) may be expanded in terms of the $|\varphi_p\rangle$,

$$|\psi(t)\rangle=\sum_p a_p(t)|\varphi_p(t)\rangle. \tag{5}$$

In the usual manner,[8] we arrive at the coupled system of equations which the expansion amplitudes $a_p(t)$ must satisfy,

$$i\dot{a}_p(t)=\sum_m a_m(t)e^{i(\omega_p-\omega_m)t}\langle\varphi_p|H|\varphi_m\rangle+i\delta_{pl}\delta(t). \tag{6}$$

In the above, ω_p is the energy corresponding to $|\varphi_p\rangle$ (i.e., $H_0|\varphi_p\rangle=\omega_p|\varphi_p\rangle$) and the time dependence of the matrix elements has been explicitly displayed. The inhomogeneous term expresses the boundary condition $a_p(0)=\delta_{pl}$; at $t=0$, the system is in the pure state corresponding to $p=l$. More useful than (6) is its Fourier transform.[9] If we write

$$a_p(t)=-(2\pi i)^{-1}\int_{-\infty}^{\infty}d\omega\exp[i(\omega_p-\omega)t]A_p(\omega), \tag{7}$$

then (6) can be written as

$$(\omega-\omega_p+i\epsilon)A_p(\omega)=\sum_m A_m(\omega)\langle\varphi_p|H|\varphi_m\rangle+\delta_{pl}. \tag{8}$$

The introduction of $+i\epsilon$ in (8) ensures that the system will display the correct behavior for $t<0$ and gives us the proper causality conditions.

For a single nucleus having only one excited level (at energy ω_0) and two competing modes of decay, radiative (matrix element $\langle|H|\rangle$) and internal conversion (matrix element $\langle|h|\rangle$), there exist only three

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significant amplitudes. They are as follows: (1) $A(\omega)$, nucleus in excited state, energy ω_0; (2) $B_{\mathbf{k}}(\omega)$, nucleus in ground state, one photon $\mathbf{k}$ present, energy ω_k; (3) $C_{\mathbf{p}}(\omega)$, nucleus in ground state, one conversion electron $\mathbf{p}$ present, energy ω_p. If this nucleus is initially excited, (8) may be written as

$$(\omega-\omega_0+i\epsilon)A(\omega)=1+\sum_{\mathbf{k}} H_{\mathbf{k}}B_{\mathbf{k}}(\omega)+\sum_{\mathbf{p}} h_{\mathbf{p}}C_{\mathbf{p}}(\omega), \quad (9a)$$

$$(\omega-\omega_k+i\epsilon)B_{\mathbf{k}}(\omega)=H_{\mathbf{k}}^*A(\omega), \quad (9b)$$

and

$$(\omega-\omega_p+i\epsilon)C_{\mathbf{p}}(\omega)=h_{\mathbf{p}}^*A(\omega). \quad (9c)$$

For simplicity, $\langle\varphi_{\text{ex}}|H_{\mathbf{k}}|\varphi_{\text{gnd}}\rangle$ has been written as $H_{\mathbf{k}}$ and $\langle\varphi_{\text{ex}}|h_{\mathbf{p}}|\varphi_{\text{gnd}}\rangle$ as $h_{\mathbf{p}}$. When substituting (9b, 9c) into (9a), we obtain

$$(\omega-\omega_0+i\epsilon)A(\omega)=1+\sum_{\mathbf{k}}\frac{A(\omega)|H_{\mathbf{k}}|^2}{(\omega-\omega_k+i\epsilon)}+\sum_{\mathbf{p}}\frac{A(\omega)|h_{\mathbf{p}}|^2}{(\omega-\omega_p+i\epsilon)}. \quad (10)$$

Each of the sums in (10) may be evaluated by first converting them to integrals and then employing the symbolic identity

$$(x+i\epsilon)^{-1}=\mathrm{P}(1/x)-i\pi\delta(x).$$

The δ-function contribution gives an imaginary term which results in a finite linewidth. The principal value term results in an energy shift of the line and will not be considered further since it may be eliminated by correctly choosing our expansion states. In this manner, (10) may be rewritten as

$$\begin{aligned}A(\omega)&=[\omega-\omega_0+i\gamma_c/2+i\gamma_R/2]^{-1}\\&=[\omega-\omega_0+i\Gamma/2]^{-1},\end{aligned} \quad (11)$$

where γ_c, γ_R, and Γ are the partial internal conversion, partial radiative, and total linewidths, respectively. We find, for example, that γ_R is given by $\gamma_R=V\omega^2|H|^2/\pi$ where V is the volume of normalization and $|H|^2$ is the square of the matrix element averaged over all possible $\mathbf{k}$ directions. Let us for the present ignore polarization effects and consider $H_{\mathbf{k}}$ to depend only on $|\mathbf{k}|$. Finally, it should be noted that the Fourier transform of (11) leads to the simple usual exponential decay, $|a(t)|^2=\exp(-\Gamma t)$.

The simplest quantum mechanical model for resonant fluorescence is that in which only two nuclei participate, the source and the absorber. In general, we will never consider the details of the solid-state questions arising in connection with the conditions for recoiless emission or absorption. For our purposes, we will take all processes to be recoiless and will later attempt to overcome this restriction by a phenomenological extension of our results. First we will consider a one-dimensional experiment in which the source nucleus is located at $x=0$ and the absorber at $x=x_0>0$. The initial condition is that the first photon of the source cascade is emitted at $t=0$. Thus, we are able to say that the source nucleus is in the first-excited level with unit probability at that instant. We can write (8) for this system as

$$(\omega-\omega_0+i\gamma_c/2)A(\omega)=1+\sum_k H_kB_k(\omega), \quad (12a)$$

$$(\omega-\omega_k+i\epsilon)B_k(\omega)=A(\omega)H_k^*+C(\omega)H_k^*e^{-ikx_0}, \quad (12b)$$

and

$$(\omega-\omega_0'+i\gamma_c/2)C(\omega)=\sum_k B_k(\omega)H_ke^{ikx_0}. \quad (12c)$$

The three states of interest are as follows: (1) only source excited, energy ω_0, amplitude $A(\omega)$; (2) both nuclei in ground state, one photon, k, present, energy ω_k, amplitude $B_k(\omega)$; (3) only absorber excited, energy ω_0', amplitude $C(\omega)$.

We have written ω_0' for the first energy level of the absorber to allow for the Doppler shift due to its motion relative to the source ($\omega_0'=\omega_0+\omega_0v/c$, where v is the absorber velocity). Actually, the various frequency components of the source radiation experience different Doppler shifts. A more careful calculation reveals that the only effect of this exact treatment (aside from adding mathematical complications) is to alter the final transmission by terms of higher order in v/c which would be immeasurable for such experiments because of the small relative velocities employed ($v\lesssim 1$ cm/sec, typically).

In writing (12), we have already eliminated the amplitudes for internal conversion, resulting in the added factors of ($i\gamma_c/2$), in the previously discussed manner. If the conversion coefficient is large (i.e., $\gamma_c\gg\gamma_R$) or the geometry favorable, the source nucleus will decay in the same manner as it would if the absorber were absent. To good approximation, we may assume that $A(\omega)$ is the same here as given by (11). When substituting (12b) and (11) into (12c), we obtain

$$\begin{aligned}C(\omega)=&-\tfrac{1}{2}i\gamma_Re^{i\omega x_0}(\omega-\omega_0+i\Gamma/2)^{-1}\\&\times(\omega-\omega_0'+i\Gamma/2)^{-1}.\end{aligned} \quad (13)$$

This expression may be used in turn to solve for $B_k(\omega)$ via (12b),

$$B_k(\omega)=H_k^*(\omega-\omega_0+i\Gamma/2)^{-1}(\omega-\omega_k+i\epsilon)^{-1}\times\left[1-i\frac{\gamma_R}{2}\frac{e^{i(\omega-k)x_0}}{(\omega-\omega_0'+i\Gamma/2)}\right]. \quad (14)$$

The time-dependent amplitude can now be obtained from (7). Finally, this allows us to calculate the spatial wave function of the radiation field,

$$\begin{aligned}\psi(x,t)&=\sum_k L^{-\frac{1}{2}}e^{i(kx-\omega_kt)}b_k(t) \quad \text{for} \quad t\geqslant x>x_0\\&=-iL^{\frac{1}{2}}H_{\omega_0}^*e^{i(\omega_0-i\Gamma/2)(x-t)}\\&\quad\times[1-\tfrac{1}{2}i\gamma_R(\omega_0-\omega_0')^{-1}(1-e^{i(\omega_0-\omega_0')(t-x)})],\end{aligned} \quad (15)$$

where L is a normalization length. For $x>x_0$, the square of the magnitude of the wave function is the

transmission,

$$T(\tau)=\tfrac{1}{2}\gamma_R e^{-\Gamma\tau}\{1-\gamma_R(\omega_0-\omega_0')^{-1}\sin[(\omega_0-\omega_0')\tau] +\gamma_R^2(\omega_0-\omega_0')^{-2}\sin^2[\tfrac{1}{2}\tau(\omega_0-\omega_0')]\}, \quad (16)$$

where $\tau=t-x$. The various terms in (16) may be identified as the counting rate due to the emission from the source, the effect of absorption due to the absorber, and the subsequent re-emission from the absorber, respectively. This correspondence becomes more evident if (16) is integrated over τ. This latter case describes an experiment in which no coincidence measurement is made. The result is

$$\langle T\rangle=\left(\frac{\gamma_R}{2\Gamma}\right)\left[1-\left(\frac{\gamma_R}{\Gamma}\right)\frac{\Gamma^2}{(\omega_0-\omega_0')^2+\Gamma^2} +\frac{1}{2}\left(\frac{\gamma_R}{\Gamma}\right)^2\frac{\Gamma^2}{(\omega_0-\omega_0')^2+\Gamma^2}\right]. \quad (17)$$

The probability of a photon emission for the source nucleus is γ_R/Γ. The factor of $\frac{1}{2}$ arises because only half of the emitted photons will be headed in the forward direction in this simple one-dimensional model. This explains the first term as the source emission contribution. The second term is the absorption term since it merely multiplies the incident flux by the absorption cross section. The third term gives the probability of re-emission, the absorption multiplied by the appropriate conversion fraction (another factor of $\frac{1}{2}$ enters here as in the first term).

VERIFICATION OF THE $t=0$ BOUNDARY CONDITION

The boundary condition previously used was the one in which the source was excited with unit probability at $t=0$. All other amplitudes were taken to be zero at that time. This corresponds to the assumption that a measurement of the first cascade photon emitted by the source at $t=0$ establishes the state of the source nucleus. In this section both radiations in the cascade will be included explicitly. The source will be assumed to have two excited states, one at energy W, the second at energy ω_0 ($W>\omega_0$). The Mössbauer level is the lower of these two. At $t=0$, the source is in the state with energy W. At some later time, it emits photon k and goes into the level at energy ω_0. At a still later time, photon q is emitted as the source nucleus goes into its ground state. This is the Mössbauer photon which may or may not be absorbed by the second nucleus. Four amplitudes must be considered for this problem. They are as follows: (1) $A(\omega)$, the source at level W, no photons present; (2) $B_k(\omega)$, the source at level ω_0, one photon present (k); (3) $C_{kq}(\omega)$, the source in its ground state, two photons present (k,q); (4) $D_k(\omega)$, the absorber excited to energy ω_0', one photon present (k).

The appropriate set of equations for these amplitudes is

$$(\omega-W+i\epsilon)A(\omega)=1+\textstyle\sum_k H_kB_k(\omega), \quad (18a)$$

$$(\omega-\omega_0-\omega_k+i\gamma_c/2)B_k(\omega)=H_k^*A(\omega) +\textstyle\sum_q H_qC_{kq}(\omega), \quad (18b)$$

$$(\omega-\omega_k-\omega_q+i\epsilon)C_{kq}(\omega)=H_q^*[B_k(\omega) +D_k(\omega)e^{-iqx_0}], \quad (18c)$$

and

$$(\omega-\omega_k-\omega_0'+i\gamma_c/2)D_k(\omega)=\textstyle\sum_q H_qe^{iqx_0}C_{kq}(\omega). \quad (18d)$$

The chain $A\rightarrow B\rightarrow C$ occurs as it would in the absence of the absorber to the same approximation as discussed previously. Thus, $A(\omega)$ and $B(\omega)$ can be obtained by solving (18a–18c) after setting $D(\omega)$ equal to zero. The result of this calculation is

$$A(\omega)=(\omega-W+i\Lambda/2)^{-1}, \quad (19)$$

and

$$B_k(\omega)=H_k^*(\omega-W+i\Lambda/2)^{-1}(\omega-\omega_0-\omega_k+i\Gamma/2)^{-1}, \quad (20)$$

where Λ is the linewidth of the first-excited state and Γ is the total linewidth of the second-excited state. The radiative linewidth of this second level will again be denoted by γ_R. We can now use (19, 20) together with (18c, 18d) to obtain $C_{kq}(\omega)$. Substituting (18c) into (18d) and using (20), we obtain

$$D_k(\omega)=-\tfrac{1}{2}i\gamma_RH_k^*\frac{e^{i(\omega-\omega_k)x_0}}{(\omega-\omega_0-\omega_k+i\Gamma/2)(\omega-W+i\Lambda/2)(\omega-\omega_k-\omega_0'+i\Gamma/2)}. \quad (21)$$

Substituting (20) and (21) back into (18c) gives the desired result for $C_{kq}(\omega)$,

$$C_{kq}(\omega)=\frac{H_k^*H_q^*}{(\omega-W+i\Lambda/2)(\omega-\omega_0-\omega_k+i\Gamma/2)} \times\left[1-i\frac{\gamma_Re^{i(\omega-\omega_k-q)x_0}}{2(\omega-\omega_0'-\omega_k+i\Gamma/2)}\right]. \quad (22)$$

We can now obtain the Fourier transform of (22),

$$C_{kq}(t)=-(2\pi i)^{-1}\int_{-\infty}^{+\infty}d\omega\,\exp[i(\omega_k+\omega_q-\omega)t] \times C_{kq}(\omega). \quad (23)$$

The wave function corresponding to this amplitude is

$$\psi(x_1,x_2,t)=(L/4\pi^2)\int dkdq\,C_{kq}(\omega)\exp[i(kx_1-\omega_kt)] \times\exp[i(qx_2-\omega_qt)]. \quad (24)$$

Obviously, $\psi(x_1,x_2,t)$ can be considered as a probability amplitude. Therefore, $|\psi(x_1,x_2,t)|^2$ is the probability of simultaneously finding the first photon at x_1 and the second photon at x_2 at a time t. If we place a detector capable of sensing the first radiation at x_1 and another which detects the second at x_2, then $|\psi(x_1,x_2,t)|^2$ becomes the probability of simultaneously counting both photons at the time t. If $x_1>x_2$, then the first photon was emitted from the source at a time $t_1=t-x_1$, and the second was emitted at a time $t_2=t-x_2$. Thus, we have $t_1<t_2$, and a delay has been inserted into the counting system which is $\tau=x_1-x_2$. In this manner, this calculation is seen to parallel the actual physical manipulations made in a laboratory delayed coincidence measurement. Since the experiment described above has no way of determining the length of time which the source nucleus spends before its first decay, we must integrate over the time t. Combining (23) and (24), and using the discussion of this paragraph, we obtain the transmission as a function of the delay time,

$$T(\tau)=L^2(2\pi)^{-6}\int d\omega d\omega' dkdk'dqdq'dt\, C_{kq}(\omega)C_{k'q'}(\omega')^* \times e^{i(\omega_k+\omega_q-\omega)t}e^{-i(\omega_{k'}+\omega_{q'}-\omega')t}e^{i(qx_2-\omega_q t)}e^{-i(q'x_2-\omega_{q'}t)} \times e^{i(k\tau+kx_2-\omega_k t)}e^{-i(k'\tau+k'x_2-\omega_{k'}t)}. \quad (25)$$

The function $C_{kq}(\omega)$ has the proper analyticity so that the integrand is nonzero only for $t>0$. This allows us to perform the t integration from $-\infty$ to $+\infty$. Since the time dependence in the integrand is completely specified, the integral over t is the easiest to do. The result of this integration is $2\pi\delta(\omega'-\omega)$, so that

$$T(\tau)=L^2(2\pi)^{-5}\int d\omega dkdk'dqdq'\, C_{kq}(\omega)C_{k'q'}(\omega)^* \times e^{i(k-k')\tau}e^{i(k+q-k'-q')x_2}. \quad (26)$$

For $x_2>x_0$ and $\tau\geqslant 0$ we find

$$T(\tau)=\tfrac{1}{4}\gamma_R e^{-\Gamma\tau}\{1-\gamma_R(\omega_0-\omega_0')^{-1}\sin[(\omega_0-\omega_0')\tau] +\gamma_R^2(\omega_0-\omega_0')^{-2}\sin^2[\tfrac{1}{2}(\omega_0-\omega_0')\tau]\}. \quad (27)$$

This result is the same as that obtained in (16) except for the added factor of $\frac{1}{2}$ here. This arises since only half of the first emitted radiation is detected by the counter. The rest is emitted in the backward direction.

The detailed treatment of this section verifies the validity of the simpler treatment of boundary conditions which we have used up until now. Once performed rigorously, it is now sufficient to resort to the simpler treatment again which is what we will do in the remainder of this paper.

Everything done up to this point may easily be repeated for a three-dimensional system. The only additional complications which arise are those of geometrical origin and in no way alter the general conclusions already reached. In this case, however, the probability of the source being re-excited by reemission from the absorber is small for reasonable source-absorber separation because of the small solid angle subtended by either nucleus at the site of the other. Therefore, the previous assumption concerning the simple exponential decay of the source is certainly valid here (even if there is no conversion present).

THICK ABSORBERS

Real experiments, however, are performed with absorber foils which are composed of many nuclei. Such a system is described in analogy to (12), by

$$(\omega-\omega_0+i\gamma_c/2)A(\omega)=1+\textstyle\sum_{\mathbf{k}} H_k B_{\mathbf{k}}(\omega), \quad (28a)$$

$$(\omega-\omega_k+i\epsilon)B_{\mathbf{k}}(\omega) =H_k^*[A(\omega)+\textstyle\sum_l e^{-i\mathbf{k}\cdot\mathbf{x}_l}C_l(\omega)], \quad (28b)$$

and

$$(\omega-\omega_0'+i\gamma_c/2)C_l(\omega)=\textstyle\sum_{\mathbf{k}} H_k e^{i\mathbf{k}\cdot\mathbf{x}_l}B_{\mathbf{k}}(\omega). \quad (28c)$$

The amplitudes $A(\omega)$ and $B_{\mathbf{k}}(\omega)$ have the same meaning as those in (12). Now, however, $C_l(\omega)$ is the amplitude for the excitation of the lth absorber nucleus (located at $\mathbf{x}_l$) with no photons present and all other nuclei in their ground states.

Let us consider an absorber slab of cross section L^2 and thickness d. If the source is moved far from the absorber, the photons incident on the absorber may be considered as plane waves traveling in the positive z direction. Under this last approximation, (28b) becomes

$$(\omega-\omega_k+i\epsilon)B_{\mathbf{k}}(\omega)=H_k^*\left\{\omega_0 L\frac{\delta(k_x,0)\delta(k_y,0)\theta(k_z)}{(\omega-\omega_0+i\Gamma/2)} +\textstyle\sum_l e^{-i\mathbf{k}\cdot\mathbf{x}_l}C_l(\omega)\right\}, \quad (29)$$

where we have again assumed that the source decays as it would if no absorber were present. The normalization has been chosen so that the total photon intensity incident on the absorber is $(\pi\gamma_R/\Gamma)$. In the above, $\delta(k,0)$ is the usual Kronecker delta function

$$\delta(k,0)=1 \quad \text{for} \quad k=0$$
$$=0 \quad \text{for} \quad k\neq 0,$$

and $\theta(k)$ is the step function

$$\theta(k)=1 \quad \text{for} \quad k>0$$
$$=0 \quad \text{for} \quad k<0.$$

The time origin has been chosen so that the originally emitted wave front reaches the origin at $t=0$.

Combining (28c) and (29), we obtain

$$(\omega-\omega_0'+i\Gamma/2)C_l(\omega)=-i\frac{\pi\gamma_R e^{i\omega z_l}}{\omega_0 L(\omega-\omega_0+i\Gamma/2)} -\frac{\gamma_R}{2}\sum_{j\neq l} C_j(\omega)\frac{e^{i\omega|\mathbf{x}_j-\mathbf{x}_l|}}{\omega|\mathbf{x}_j-\mathbf{x}_l|}, \quad (30)$$

where the sums over $\mathbf{k}$ have been performed as before. Thus, we have reduced the problem to an integral equation for $C_l(\omega)$. The solution of (30) depends, of course, upon the spatial distribution of the absorber nuclei. The only case treated here will be the one in which the summation sign in (30) can be replaced by an integral (or consider the discrete lattice replaced by a continuous distribution). If the wavelength of the characteristic radiation is much larger than the typical spacing of absorber nuclei, then this approximation is a good one. The 14.4-kev photon in Fe^{57} has $\lambda \approx 10^{-8}$ cm, which is of the same order as the nuclear spacing in the absorber crystal. Thus, we must admit that this approximation does not appear to be too good here. Most Mössbauer experiments, however, use iron absorbers which contain the natural abundance of Fe^{57} nuclei (i.e., 2.17%). In this case, the absorber nuclei will be situated on random lattice sites (only about one out of 50 will be occupied by a resonant absorber). The actual transmission will then be determined by the average concentration of active absorbers, and the continuous absorber will give a good approximation to the physical situation. (A detailed calculation for a one-dimensional random lattice has verified this argument.)

Our formalism up to this point has been sufficiently general to include all absorber configurations. It can also be shown that for well-ordered lattices, one would expect resonant Bragg scattering as in the simple x-ray case. We will, however, restrict ourselves to the case where the continuous absorber approximation may be used. When we introduce an absorber density $\rho(\mathbf{x})$, (30) may be rewritten as

$$(\omega-\omega_0'+i\Gamma/2)C(\mathbf{x},\omega)=-i\frac{\pi\gamma_R e^{i\omega z}}{\omega_0 L(\omega-\omega_0+i\Gamma/2)} -\frac{\gamma_R}{2}\int d\mathbf{x}'\,\rho(\mathbf{x}')C(\mathbf{x}',\omega)\frac{e^{i\omega|\mathbf{x}-\mathbf{x}'|}}{\omega|\mathbf{x}-\mathbf{x}'|}. \quad (31)$$

For convenience, the absorber slab will be taken to be normal to the direction of the incident beam. Therefore, we have $\rho(\mathbf{x})=\rho$ for all x and y when $0<z<d$, and $\rho(\mathbf{x})$ vanishes for all other space points. Using symmetry arguments, it is obvious that $C(\mathbf{x},\omega)$ cannot depend on x or y. The integration over x and y can be performed and leads to

$$(\omega-\omega_0'+i\Gamma/2)C(z,\omega)=-i\frac{\pi\gamma_R e^{i\omega z}}{\omega_0 L(\omega-\omega_0+i\Gamma/2)} -i\frac{\pi\gamma_R\rho}{\omega_0^2}\int_0^d dz'\,C(z',\omega)e^{i\omega|z-z'|}. \quad (32)$$

The absolute value sign in the integrand can be eliminated by separating the integral into two parts,

$$(\omega-\omega_0'+i\Gamma/2)C(z,\omega)=-i\frac{\pi\gamma_R e^{i\omega z}}{\omega_0 L(\omega-\omega_0+i\Gamma/2)} -i\frac{\pi\gamma_R\rho}{\omega_0^2}\left[\int_0^z dz'\,C(z',\omega)e^{i(z-z')\omega}+\int_z^d dz'\,C(z',\omega)e^{i(z'-z)\omega}\right]. \quad (33)$$

An iterative technique can be used to solve (33). If we first assume that the z dependence in $C(z,\omega)$ is given entirely by the inhomogeneous term as $\exp(i\omega z)$, then this substitution for $C(z',\omega)$ in the first integral removes all z' dependence from the integrand. The second integrand, however, will go as $\exp(2i\omega z')$ which fluctuates rapidly as z' varies across the absorber, resulting in a small contribution. This latter integral can, in fact, be identified with waves traveling backward through the absorber. If this second integral is neglected, we can obtain an otherwise exact solution to (33),

$$C(z,\omega)=-i(\pi\gamma_R/\omega_0 L)\times(\omega-\omega_0+i\Gamma/2)^{-1}(\omega-\omega_0'+i\Gamma/2)^{-1}\times\exp\left[i\omega z-i\frac{\pi\rho\gamma_R z}{\omega_0^2(\omega-\omega_0'+i\Gamma/2)}\right]. \quad (34)$$

To obtain a quantitative estimate of the error introduced in neglecting the previously mentioned integral, it may be noted that a more exact solution of (33) has been investigated which retains the backward traveling waves. The lowest-order correction to (34) produces terms which contain added factors of $(\beta_0/2\omega)$ and $(\beta_0^2 z/2\omega)$ where β_0 is given by

$$\beta_0=-i(\pi\rho\gamma_R/\omega_0^2)(\omega-\omega_0'+i\Gamma/2)^{-1}.$$

The maximum value of $|\beta_0/2\omega|$ is proportional to the number of absorber nuclei in a cube with side $\lambda\approx[2/E(\text{kev})]\times10^{-8}$ cm, since

$$|\beta_0/2\omega|_{\max}=\pi(\gamma_R/\Gamma)\rho\lambda^3.$$

We have $\rho^{\frac{1}{3}}<3\times10^7$ cm^{-1}, so that $|\beta_0/2\omega|_{\max}\ll1$ for reasonable transition energies. For Fe^{57} absorbers, we have $|\beta_0/2\omega|_{\max}<10^{-5}$ and $|\beta_0^2 z/2\omega|_{\max}<10^{-1}d$ (cm). Thus for $d<10^{-2}$ cm, the largest correction terms are of the order of 10^{-3}. Thus, if (33) is valid, then (34) will give the transmission to within 0.1% which is sufficient accuracy for our purposes considering that we have already neglected larger effects which will be mentioned later.

We are now in a position to calculate the radiation field from (29) which for the continuous absorber

becomes

$$(\omega-\omega_k+i\epsilon)B_{\mathbf{k}}(\omega)=H_k^*\left\{\omega_0L\frac{\delta(k_x,0)\delta(k_y,0)\theta(k_z)}{(\omega-\omega_0+i\Gamma/2)}+\int d\mathbf{x}\,\rho(\mathbf{x})e^{-i\mathbf{k}\cdot\mathbf{x}}C(z,\omega)\right\}. \quad (35)$$

Using (34) and immediately making use of the lack of x and y dependence in $C(z,\omega)$, we obtain

$$(\omega-\omega_k+i\epsilon)B_{\mathbf{k}}(\omega)=\frac{H_k^*\omega_0L}{(\omega-\omega_0+i\Gamma/2)}\delta(k_x,0)\delta(k_y,0)\times\left\{\theta(k_z)-i\frac{\pi\rho\gamma_R}{\omega_0^2}\int_0^d dz\frac{e^{i(\omega-k_z)z}}{(\omega-\omega_0'+i\Gamma/2)}\times\exp\left(-i\frac{\pi\rho\gamma_Rz}{\omega_0^2(\omega-\omega_0'+i\Gamma/2)}\right)\right\}. \quad (36)$$

Thus we see that the only nonzero photon amplitude in this approximation occurs for $k_x=k_y=0$. The second term does give a finite amplitude for backward scattered photons which, however, is small because of the factor $\exp[i(\omega-k_z)z]$ occurring in the integrand. Other terms of this order have already been neglected in using (34), and in order to be consistent, we will also omit this small contribution. To lowest order, only transmitted photons survive. The argument of $\exp[i(\omega-k_z)z]$ is typically of the order of $\Gamma d=d/c\tau$ where τ is the mean life of the transition. For Fe^{57}, $\tau\approx10^{-7}$ sec so that $(d/c\tau)\approx1/3\times10^{-3}d$ (cm). Hence, for reasonable foil thicknesses, this exponential factor can be replaced by unity. The remaining integral can be performed easily to yield

$$B_k^{\text{fwd}}(\omega)\approx\frac{H_k^*\omega_0L}{(\omega-\omega_k+i\epsilon)(\omega-\omega_0+i\Gamma/2)}\times\exp\left[-i\frac{\pi\rho\gamma_Rd}{\omega_0^2(\omega-\omega_0'+i\Gamma/2)}\right]. \quad (37)$$

The time dependent amplitude $b_k^{\text{fwd}}(t)$ can be obtained by using (7). This latter integral over ω is complicated by the presence of an essential singularity at $\omega=\omega_0'-i\Gamma/2$ in addition to the simple poles at $(\omega_k-i\epsilon)$ and $(\omega_0-i\Gamma/2)$.

We can obtain the spatial wave function of the radiation field from $b_k^{\text{fwd}}(t)$ as

$$\psi(\mathbf{x},t)=\sum_{\mathbf{k}}L^{-\frac{3}{2}}e^{i(\mathbf{k}\cdot\mathbf{x}-\omega_kt)}b_{\mathbf{k}}(t)=\int_0^\infty dk(2\pi)^{-1}L^{-\frac{1}{2}}e^{i(kz-\omega_kt)}b_k^{\text{fwd}}(t), \quad (38)$$

where, in the last line, we have replaced the sum by an integral and made use of the "forward" property of the field amplitude. Combining (37), (7), and (38), we have

$$\psi(z,t)=-\frac{\omega_0L^{\frac{1}{2}}}{4\pi^2i}\times\int_0^\infty dk\int d\omega\,e^{i(kz-\omega_kt)}\frac{H_k^*}{(\omega-\omega_k+i\epsilon)(\omega-\omega_0+i\Gamma/2)}\times\exp\left[-i\frac{\pi\rho\gamma_Rd}{\omega_0^2(\omega-\omega_0'+i\Gamma/2)}\right]. \quad (39)$$

It is useful to perform the k integration first. Assuming that H_k^* is a smooth function of k, we obtain for $z>0$,

$$\psi(z,t)=\frac{\omega_0L^{\frac{1}{2}}}{2\pi}\int d\omega\,e^{i\omega(z-t)}\frac{H_\omega^*}{(\omega-\omega_0+i\Gamma/2)}\times\exp\left[-i\frac{\pi\rho\gamma_Rd}{\omega_0^2(\omega-\omega_0'+i\Gamma/2)}\right]. \quad (40)$$

This integral is of the same form as that considered by Hamermesh. The result of this last integration is, for $t>z>0$,

$$\psi(z,t)=-\omega_0L^{\frac{1}{2}}iH_{\omega_0}^*e^{-i(\omega_0'-i\Gamma/2)(t-z)}\times\sum_{n=0}^\infty[-i(\omega_0-\omega_0')(t-z)]^n\left[\frac{\omega_0^2}{\pi\rho\gamma_Rd(t-z)}\right]^{n/2}\times J_n[(4\pi\rho\gamma_Rd(t-z)/\omega_0^2)^{\frac{1}{2}}]. \quad (41)$$

Consequently, the transmission per unit incident flux may be written as

$$T(\tau)=(\pi\gamma_R/\Gamma)^{-1}L^2|\psi|^2=\Gamma e^{-\Gamma\tau}\left|\sum_{n=0}^\infty[-i\omega_0\Delta\omega(\tau/\pi\rho\gamma_Rd)^{\frac{1}{2}}]^n\times J_n[(4\pi\rho\gamma_R\tau d/\omega_0^2)^{\frac{1}{2}}]\right|^2, \quad (42)$$

where $\tau=t-z$ and $\Delta\omega=\omega_0-\omega_0'$. This result is seen to be identical (except for normalization) with the classically derived (4) if we make the correspondence $\beta=(4\pi\rho d\gamma_R/\omega_0^2\Gamma)$.

One result worth noting is that if (4) or (42) is expanded in ascending powers of β, the first-order term in β agrees with the absorption term of (16). Since β is proportional to absorber thickness, (16) gives the correct absorption for extremely thin absorber foils.

An interesting problem that may be considered concerns the result of a measurement in which photons are detected for all τ. This is the usual simple counting experiment which does not make use of delayed coincidence techniques. The result of integrating (42) over τ can be written as

$$\langle T\rangle=\frac{\Gamma}{2\pi}\int_0^\infty\frac{\exp\left(-\frac{4\pi}{\omega_0^2}\frac{\gamma_R}{\Gamma}\rho d\frac{\Gamma^2/4}{(\omega_k-\omega_0')^2+\Gamma^2/4}\right)}{(\omega_k-\omega_0)^2+\Gamma^2/4}d\omega_k. \quad (43)$$

This is exactly the same result as that predicted by Visscher[10] from simple considerations. The maximum cross section for a single absorption can be seen to be $\sigma_{\max}$, where

$$\sigma_{\max}=(4\pi/\omega_0^2)(\gamma_R/\Gamma). \tag{44}$$

CONCLUSION

We have seen that a quantum mechanical derivation of the Mössbauer transmission (for absorbers which do not have sufficiently good crystalline structure so as to permit Bragg scattering) agrees with the previously mentioned classical result of Hamermesh. In addition, the initial conditions were investigated in detail by considering the first radiation of the cascade which leads to the first-excited state (the Mössbauer level). We were able to explicitly verify that the same result is obtained as when we adopt the simpler approach where we consider the nucleus to be in the first-excited state with unit probability immediately after the emission of the first photon.

The actual problem contains complications not yet considered here. The solid-state aspects have not been taken into account. Let us say that the probability that any nuclear absorption or emission will be recoilless is given by the parameter f (determined from experiment or some other theory). The density of active absorbers is then essentially reduced from ρ to $f\rho$. In addition to this substitution in (42), we must also include an off-resonance flux which is given by $(1-f)$ of the source radiation and cannot be affected by the absorber except for normal scatterings. The effective transmission then becomes

$$T_{\text{eff}}(\tau)=(1-f)\Gamma e^{-\Gamma t}+fT(\tau), \tag{45}$$

in which $T(\tau)$ is given by (42) with the substitution of $f\rho$ for ρ.

Equations (42) and (44) can be combined, which allows us to rewrite the transmission as a function of $\sigma_{\max}$. This is equivalent to setting $\beta=\rho d\sigma_{\max}$ in (4). We have performed our previous calculations considering only "spinless photons." For the case of a physical transition between nuclear levels with spins I_{ex} and I_{g}, we must use

$$\sigma_{\max}=\frac{2\pi}{\omega_0^2}\left(\frac{2I_{\text{ex}}+1}{2I_{\text{g}}+1}\right)\frac{\gamma_R}{\Gamma}$$

in place of (44) when both source and absorber are unpolarized. In the event that we do have polarization, the angular dependence of the cross section must be taken into account.

One of the most difficult problems to compensate for in the previous calculations, is that of hyperfine splitting. In reality, the emission and absorption spectra contain many lines due to the hyperfine interaction between crystalline fields and nuclear moments. In Fe^{57}, the excited and ground states have spins of $\frac{3}{2}$ and $\frac{1}{2}$, respectively, so that a dipole transition produces a six-line spectrum. The simplest comparison with experiment may be obtained by assuming that each emission line only overlaps the corresponding absorption line of the absorber. This requires that one must produce Doppler shifts in the absorber which are small when compared with the hyperfine splitting so that overlap of different components does not occur. This situation is easily obtained if the splittings are large compared with the linewidths involved. An interesting problem which arises is that for very short times, the lines become extremely broad. If the delay time is chosen to be short enough, then the lines are so broad that overlap must occur between different components. No suggestion of the effects caused by this phenomenon will be offered here.

ACKNOWLEDGMENTS

The author would like to express his gratitude to Professor H. Frauenfelder for originally bringing this problem to his attention and to Professor J. D. Jackson, who provided encouragement and advice throughout this work and furnished many suggestions which have been incorporated into this manuscript. He also wishes to thank Dr. J. H. Hetherington for many helpful discussions.

[10] W. M. Visscher, "Evaluation of transmission integral," Los Alamos Scientific Laboratory, 1960 (unpublished).