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1997 J. Phys.: Condens. Matter 9 8749

(http://iopscience.iop.org/0953-8984/9/41/019)

J. Phys.: Condens. Matter **9** (1997) 8749–8765. Printed in the UK PII: S0953-8984(97)77585-4

Quantum mechanical model for nuclear-resonant scattering of gamma radiation

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Received 4 September 1996, in final form 23 June 1997

Abstract. A one-dimensional quantum mechanical model for nuclear-resonant scattering of gamma radiation from matter is developed assuming the source radiation is gamma decay. A closed-form, finite-sum solution for the radiated intensity is obtained by restricting the calculation to coherent forward scattering. The solution provides a unified microscopic picture of nuclear-resonant scattering processes in which the radiation undergoes sequential scattering from one nucleus to another before reaching the detector. For recoil-free processes the various 'paths' to the detector contribute coherently. The solution for this case gives results identical to the classical optical model. The one-dimensional model shows that the 'speed-up' and 'dynamical beating' effects are primarily a consequence of the fact that the single-nuclear scattering processes are 180° out of phase with the incident radiation while the double-nuclear-scattering processes are in phase with the incident radiation. All multiple-scattering paths are, and must be, included. The model can treat the incoherent processes, i.e. processes involving gamma emission with recoil or conversion-electron emission. The results show that a correction may be needed when analysing time-differential Mössbauer spectroscopic data due to incoherent processes that occur in the absorber.

1. Introduction

The Mössbauer effect [l, 2] is a consequence of the recoil-free emission and/or recoil-free absorption of gamma radiation that can happen in solids. The experimental technique relevant for this paper is time-differential Mössbauer spectroscopy (TDMS) [3]. The resulting 'lifetime curve' does not, in general, show the expected exponential behaviour. This effect has been called 'time filtering'. Hamermesh [3] analysed the recoil-free process using a classical optical model. (The classical optical model has also been applied to the transmission of radiation through an electronic resonant medium [4].) Subsequently Harris [5], using methods developed by Heitler [6], was able to show that the quantum mechanical treatment gives the same result. In this paper, using the same approach, a one-dimensional model is taken to represent the absorber and a different closed-form solution is found.

The studies by Hamermesh and Harris considered the recoil-free coherent nuclear radiation. Until recently there has been no experimental TDMS study of the incoherent channels. In such an experiment [7] the conversion electrons or the resulting x-rays, i.e. the inelastic channel, are detected instead of the second gamma ray. The results presented here show that the incoherent channel also exhibits unusual time-dependent effects. (In experiments using synchrotron radiation, the incoherent channel shows up more dramatically: see below.)

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In recent years there has been a renewed interest in time-domain nuclear-resonant scattering because of the availability of synchrotron-radiation facilities. A recent review [8] of this exciting field contains a summary of experimental and theoretical results. The one-dimensional model developed here does *not* apply directly to the synchrotron-radiation case. However, the model has been applied to the case of synchrotron radiation [9]. Therefore only a few remarks will be made here regarding synchrotron radiation.

The problem addressed is the one in which a radioactive source nucleus emits gamma radiation and this radiation interacts with a polycrystalline absorber containing resonant nuclei which are initially in the ground state. The time at which the source nucleus is in its first excited state is determined by a precursor gamma ray. The one-dimensional model predicts the shape of the lifetime curve, i.e. the time-dependent intensity of radiation from that state, when the radiation passes through resonant matter.

Section 2 gives a brief review of the quantum mechanical approach used to solve this problem. The model for nuclear-resonant coherent forward scattering is developed in section 3. In section 4 the connection between the model and its application to real physical situations is established. The model applied to the recoil-free processes gives an answer identical to that obtained using the classical optical model. One simply needs to establish the relationship between N, the only parameter in the model, representing the effective number of resonant nuclei, and the usual nuclear-resonant thickness parameter of the sample which is needed in the classical optical theory.

The details of the solution are put in an appendix. The result for the recoil-free processes when the source and absorber are in resonance is given in equation (12) and in the appendix equation (A22). The result when the source and absorber are not in resonance can be found by using (A17)–(A21). These equations are more complicated and will not be discussed further. The results for the processes with recoil are given in (13)–(15) and (A28)–(A34), while those for the inelastic channel are found in (16), (17) and (A35)–(A39).

2. Review of the quantum mechanical approach

The general method used in this paper is discussed by Harris [5] and Heitler [6]. The method applies quantum mechanical time-dependent perturbation theory in the frequency domain to obtain a set of coupled linear equations. The Hamiltonian of the system is divided into two parts; H_0 corresponding, in this case, to the nuclear states and the free radiation field, taken as plane waves, and H which is responsible for making transitions between the states $|\phi_p\rangle$ of H_0 by allowing the nuclei to absorb and emit radiation. In the standard approach the true state of the system $|\psi(t)\rangle$ can be expressed as

$$|\psi(t)\rangle = \sum_p a_p(t)\,\mathrm{e}^{-\mathrm{i}(E_p t/\hbar)}|\phi_p(0)\rangle \tag{1}$$

where $|\phi_p(0)\rangle$ is an eigenstate of H_0. Solving the Schrödinger equation in the usual way one arrives at a set of coupled differential equations relating the expansion coefficients $a_p(t)$.

$$\mathrm{i}\hbar\frac{\mathrm{d}a_p}{\mathrm{d}t} = \sum_m a_m(t)\,\mathrm{e}^{\mathrm{i}(\omega_p-\omega_m)t}\langle\phi_p(0)|H|\phi_m(0)\rangle + \mathrm{i}\hbar\delta_{pl}\delta(t) \tag{2}$$

The Kronecker delta and the delta function on the right-hand side in (2) are needed to denote that at time $t = 0$ the system is in the state where $p = l$ (i.e. for our case only the source nucleus is excited).

Now introducing the Fourier transform

$$a_p(t) = -\frac{1}{2\pi\,\mathrm{i}}\int_{-\infty}^{\infty}\mathrm{d}\omega\,A_p(\omega)\,\mathrm{e}^{\mathrm{i}(\omega_p-\omega)t} \tag{3}$$

into (2) and writing $\delta(t)$ in an integral representation gives

$$(\omega-\omega_p)A_p(\omega)=\sum_m A_m(\omega)\frac{\langle\phi_p(0)|H|\phi_m(0)\rangle}{\hbar}+\delta_{pl}. \tag{4}$$

However, in order that $a_p(t)=0$ for all p when $t<0$, $A_p(\omega)$ must have a pole only in the lower half of the complex plane. To ensure this (4) is re-written

$$(\omega-\omega_p+\mathrm{i}\varepsilon)A_p(\omega)=\sum_m A_m(\omega)\frac{\langle\phi_p(0)|H|\phi_m(0)\rangle}{\hbar}+\delta_{pl}. \tag{5}$$

3. The *N*-nucleus scattering problem

This formalism can be applied to study the problem where at $t=0$ there is an excited source nucleus, and N resonant 'absorber' nuclei, in the ground state, located between the source and the detector. This problem has a closed-form solution if one restricts the calculation to forward scattering. The solution has a simple form and a clear physical interpretation.

For this case there are five amplitudes: $A(\omega)$, the source nucleus located at the origin is excited (energy $\hbar\omega_0$), all absorber nuclei are in the ground state, and no photons or conversion electrons are present; $B_k(\omega)$, all nuclei are in the ground state and a photon is present of wave number k and energy $\hbar\omega_k$; $C_m(\omega)$, only the absorber nucleus located at $x=x_m$ is excited (energy $\hbar\omega_0'$) and no photons or conversion electrons are present; $D_p(\omega)$, a conversion electron from the source nucleus is present having momentum p and all nuclei are in the ground state; $E_{mp}(\omega)$, a conversion electron is present from the absorber nucleus located at $x=x_m$, and all nuclei are in the ground state. I neglect the photon polarization and the electron spin which would only obscure the relatively simple form of the final solution. Assuming that at time $t=0$ the source nucleus is excited, and substituting these amplitudes into (5), gives the following set of coupled linear equations:

$$(\omega-\omega_0+\mathrm{i}\varepsilon)A(\omega)=1+\sum_k\frac{B_k(\omega)H_k}{\hbar}+\sum_p\frac{D_p(\omega)H_p}{\hbar} \tag{6}$$

$$(\omega-\omega_k+\mathrm{i}\varepsilon)B_k(\omega)=\frac{A(\omega)H_k^*}{\hbar}+\sum_m\frac{C_m(\omega)H_k^*}{\hbar}\,\mathrm{e}^{-\mathrm{i}kx_m} \tag{7}$$

$$(\omega-\omega_0'+\mathrm{i}\varepsilon)C_m(\omega)=\sum_k\frac{B_k(\omega)H_k}{\hbar}\,\mathrm{e}^{\mathrm{i}kx_m}+\sum_p\frac{E_{mp}(\omega)H_p}{\hbar}\,\mathrm{e}^{\mathrm{i}(p/\hbar)x_m} \tag{8}$$

$$(\omega-\omega_p+\mathrm{i}\varepsilon)D_p(\omega)=\frac{A(\omega)H_p^*}{\hbar} \tag{9}$$

$$(\omega-\omega_p+\mathrm{i}\varepsilon)E_{mp}(\omega)=\frac{C_m(\omega)H_p^*}{\hbar}\,\mathrm{e}^{-\mathrm{i}(p/\hbar)x_m} \tag{10}$$

where H_k and H_k^* are the matrix elements corresponding to absorption and emission of a photon, respectively. Also H_p and H_p^* are the matrix elements corresponding to absorption and emission of a conversion electron, respectively.

The meaning of these equations can be made clear by considering, for example, (6) and (7). Equation (6) governs the amplitude for finding the source nucleus excited, $A(\omega)$. Since this is the case at $t=0$, that accounts for the '1' on the right-hand side. The source can also reach the excited state, when in the ground state, by absorbing a photon that is present. This is the meaning of the second term on the right-hand side. Similarly, when the source nucleus is in the ground state, it can be excited by absorbing its own conversion electron. Since the source nucleus is at the origin of our coordinates, there are no spatial

phase factors needed. On the other hand consider (7). This is the equation describing the situation in which all nuclei are in the ground state and there is only a photon present, $B_k(\omega)$. How can this happen? The source can emit a photon; that is the meaning of the first term on the right-hand side. Also one absorber nucleus, located at x_m, can emit a photon. Now I must put in the phase factor representing the fact that this photon appears at $x = x_m$. Furthermore one must allow any other absorber nucleus to do the same thing: the summation over all absorber nuclei is needed. The other three equations can be understood in the same way. The solution to the problem is obtained by solving this set of coupled linear equations. Some of the mathematical details are given in the appendix.

3.1. Summary of the solution

The wave function describing the transmitted intensity, when the source and absorber nuclei are in exact resonance, is expressed by (A22) from the appendix.

$$\psi_r(t') = \psi_r^{source}(t')\left[1 + \sum_{n=1}^{N}\binom{N}{n}\left(-\frac{\Gamma_r t'}{2\hbar}\right)^n \frac{1}{n!}\right] \tag{11}$$

where $\psi_r^{source}(t')$ is due to the source alone, Γ_r is the radiative width, and t' is the time measured from the time of formation of the first excited nuclear level in the source. Equation (11) contains all the amplitudes contributing to the forward scattering. This solution can be expressed in the following way by identifying the various amplitudes. There is an amplitude for the source radiation to reach the detector without interacting with any absorber nuclei: the first term on the right-hand side. There is another amplitude for the 'path' in which the source radiation is absorbed and re-emitted by one absorber nucleus before reaching the detector. This single-scattering process I term the 'one-hop' process. This amplitude contributes N times since there are N absorber nuclei: the first term involving the summation sign. There is another amplitude or 'path' where the source radiation makes two 'hops' (double scattering) on absorber nuclei before reaching the detector. This amplitude can occur according to the number of ways two objects can be selected from N objects, i.e., the binomial coefficient N over 2: the second term under the summation sign. The other 'paths' involving more hops (multiple scattering) are of a similar nature. It is important to take note that for each hop (single scattering) there is an 180° phase shift, the minus sign, and a probability given by the radiative width $\Gamma_r/2$. Next consider the amplitudes for different processes and how each contributes to the total.

3.1.1. Recoil-free processes. The source emits recoil-free radiation and, when the absorber nuclei do the same, it is impossible to distinguish which 'path' was taken for each photon that reaches the detector. Therefore all paths must be added coherently. All amplitudes must be added before taking the absolute value squared to obtain the time-dependent intensity of gamma radiation reaching the detector. The only modification needed in adapting (11) to this case is to realize that the probability for gamma-ray absorption or emission is not given by the radiative width Γ_r alone. Now the radiative width must be multiplied by the recoil-free fraction (f). Thus the time-dependent intensity of radiation reaching the detector when the source nucleus and absorber nuclei are in resonance is given by modifying (11) ((A27)) accordingly.

$$I_{fr}(t') = \frac{f\Gamma_r}{2\hbar}\,\mathrm{e}^{-(\Gamma/\hbar)t'}\left[1 + \sum_{n=1}^{N}\binom{N}{n}\left(\frac{-f\Gamma_r t'}{2\hbar}\right)^n \frac{1}{n!}\right]^2 \tag{12}$$

where Γ is the total decay width. Using (12) one can see 'speed-up' and 'dynamical beat' effects familiar [3, 8] from experimental results and classical optical model calculations. It appears that the source nucleus is decaying more rapidly than normal, i.e. the speed-up effect. Also the time-dependent intensity using thick absorbers shows local maxima at times different from zero, i.e. dynamical beats.

The one-dimensional model results for the recoil-free processes are identical to those obtained using the classical optical model when N is correlated with the nuclear-resonant thickness parameter β. The thickness parameter β is equal to $N_0 f \sigma_0 d$, where N_0 is the number of resonant nuclei per cubic centimetre, f is the recoil-free fraction, σ_0 is the maximum cross section evaluated on resonance, and d is the thickness of the sample. This agreement is shown in figures 1 and 2.

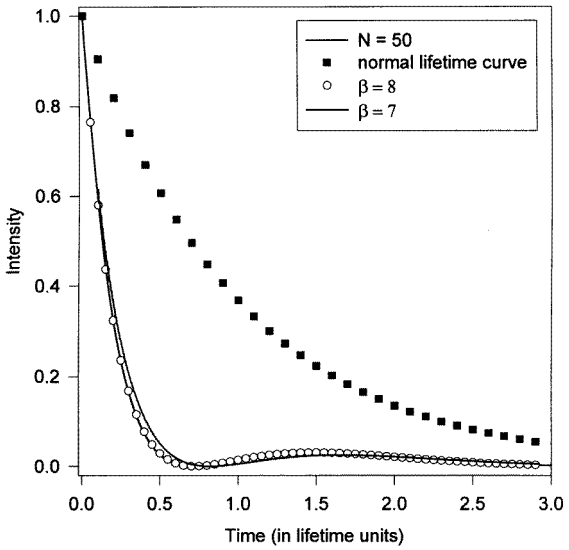

Figure 1. A comparison of the one-dimensional model with the classical optical model assuming recoil-free processes for ^{57}Fe. The one-dimensional model with $N = 50$ agrees with the classical optical model for $\beta = 8$. The result for $\beta = 7$ does not agree. The result for $\beta = 9$ does not agree either, but this is not shown to keep the figure legible. Notice the speed-up effect and dynamical beat.

In the one-dimensional model the origin of the speed-up and the dynamical beat effects can be seen by looking at the phase of each term in (12). The factors inside the square brackets of (12) are the terms to be studied. Each case n corresponds to an n-hop path. The total number of different n-hop processes is determined by the number N, the effective 'thickness' of the 'absorber'. As an example consider an 'absorber' whose resonant thickness can be correlated with $N = 50$, as shown in figure 1. In this case we will have 50 one-hop processes, 1225 two-hop processes, ... up to and including one 50-hop process where all absorber nuclei participate. A convenient way to show the various contributions is to plot each individual term in the sum, i.e. the result for each n multiplied, for convenience, by the exponential function. Figure 3 shows the no-hop, the one-hop, and the two-hop amplitudes each multiplied by the exponential factor for the case when the absorber 'contains' $N = 50$ effective nuclei. Notice that the one-hop amplitude is 180° out of phase with respect to the no-hop and the two-hop amplitudes. When these amplitudes

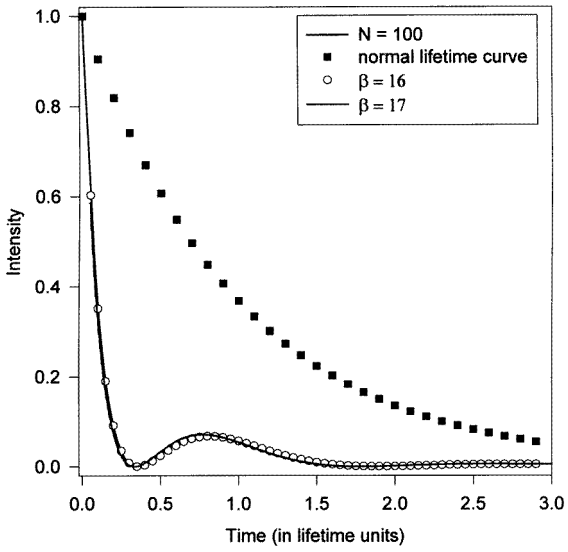

Figure 2. A comparison of the one-dimensional model with the classical optical model assuming recoil-free processes for ^{57}Fe. The one-dimensional model with $N = 100$ agrees with the classical optical model for $\beta = 16$. The result for $\beta = 17$ does not agree. The result for $\beta = 15$ does not agree either, but this is not shown to keep the figure legible. Notice the increase in the speed-up effect and an additional dynamical beat. Compare with figure 1.

are added and then squared, the one-hop amplitude causes the resulting curve to decay more rapidly near $t = 0$, the speed-up effect. At later times the contribution from the two-hop processes helps produce the local maximum near time $t = 1.5\tau$ seen in figure 1. The exact result must be computed by considering all such 'paths'.

3.1.2. Processes with recoil. Now consider the processes that occur *with* recoil in the source and absorber. If the source emits radiation, and in the process recoils, the radiation will not be in resonance with the absorber nuclei. There is an amplitude for such radiation to reach the detector. This process is distinguishable from the other processes discussed, so one only needs to consider its intensity. That intensity is given by (13) ((A28))

$$I_{(1-f)source}(t') = \frac{(1-f)\Gamma_r}{2\overline{h}}\,\mathrm{e}^{-(\Gamma/\overline{h})t'} \tag{13}$$

where $(1 - f)$ is the recoil fraction. This term, along with the recoil-free term, has been used when analysing TDMS data as discussed by Hamermesh [3].

There are other terms normally not considered when analysing TDMS data. These other processes are ones in which the recoil processes occur in the absorber. The source nucleus emits recoil-free radiation and as a result any absorber nucleus can absorb and re-emit the radiation recoil free or with recoil. Recoil-free radiation 'hops' along through the absorber but finally one nucleus may emit radiation with recoil. Given an absorber of some thickness, i.e. having an effective number of nuclei N, we must consider each absorber nucleus separately. Consider N to represent an effective number of 'absorber' layers. Start with the 'nucleus' or 'layer' closest to the source nucleus and then consider each layer out to the farthest or last one. In evaluating the contribution from each 'absorber' layer, one must include all 'paths' that lead to *excitation* of that particular 'nucleus' or 'layer'. The

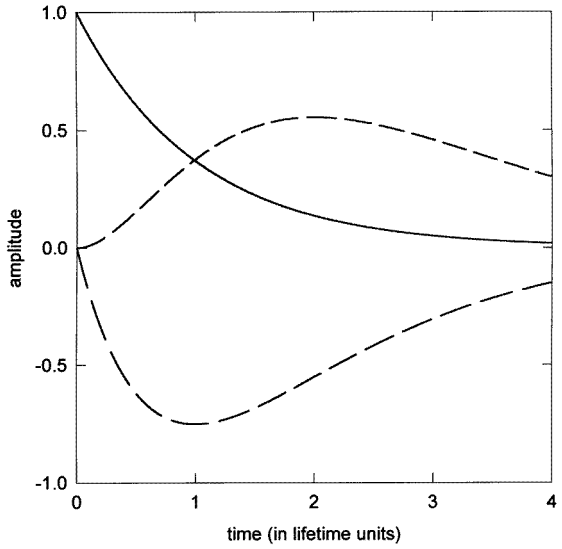

Figure 3. The 'amplitudes' (see the text for explanation) for the no-hop (solid line), one-hop (longer-dashed line), and two-hop (shorter-dashed line) multiple-scattering processes are shown multiplied by the decaying exponential. The case chosen is for $N = 50$. Notice that the one-hop amplitude is negative, while the no-hop and two-hop amplitudes are both positive. For coherent processes one must add amplitudes before squaring to obtain an intensity.

result for the total contribution of incoherent radiation from the source and all layers is given by (A33),

$$I_{(1-f)}(t') = I_{(1-f)source}(t') + \sum_{n=1}^{N} |\psi_{nk'}(t')|^2 \tag{14}$$

where k' is the wave number for a photon from the 'absorber'. Evaluating $\psi_{nk'}(t')$ for the condition when the source and absorber nuclei are in exact resonance gives (A34)

$$\psi_{nk'}(t') = \sqrt{\frac{(1-f)\Gamma_r}{2\bar{h}}}\, \mathrm{e}^{-\mathrm{i}(\omega_0 - \mathrm{i}(\Gamma/2\bar{h}))t'} \sum_{m=0}^{n-1} \binom{n-1}{m} \left(\frac{f\Gamma_r t'}{2\bar{h}}\right)^{m+1} \frac{(-1)^{m+1}}{(m+1)!}. \tag{15}$$

The first term in (14) is just the intensity from the recoiling source nucleus itself, while the terms in the summation are due to emission with recoil from different 'absorber' layers. For example for $n = 1$ the calculation gives the contribution to the intensity from the 'first' absorber 'nucleus' or 'layer'. This layer absorbs recoil-free radiation from the source nucleus and then recoils, emitting radiation. This can only happen one way. On the other hand, consider the tenth-farthest layer from the source nucleus. There are many ways in which recoil-free radiation from the source nucleus can 'hop' along recoillessly to arrive at this 'nucleus' or 'layer'. The amplitudes for these processes are given by (15).

Using the parameters for the first excited state of ^{57}Fe some results are shown in figure 4 for those processes in which emission with recoil occurs from nuclei (or layers) *in* the absorber. Figure 4 could be taken to represent the case when the detector is placed in a non-forward position and the direct radiation from the source is shielded from reaching the detector. (In figure 4 the prompt Rayleigh contribution is ignored.) In figure 4(a) three curves are given that are labelled by the location 'n' of each absorber layer. So $n = 1$

corresponds to the 'first' absorber layer, i.e. closest to the source, while $n = 10$ corresponds to the tenth absorber layer farther 'downstream'. Notice these contributions occur after $t = 0$. Because of the 180° phase change in each hop, there is a cancellation (speed-up) effect for layers downstream from the source. Figure 4(b) gives the total contribution from all the layers in the absorber assuming each 'n' layer contribution is weighted equally and the detector is shielded from the direct source radiation. (In general it is not strictly true that each contribution should be added equally. Such radiation must, in general, traverse different path lengths in the absorber before reaching the detector.) Such a time-dependent intensity should be verifiable by placing the detector in a non-forward position.

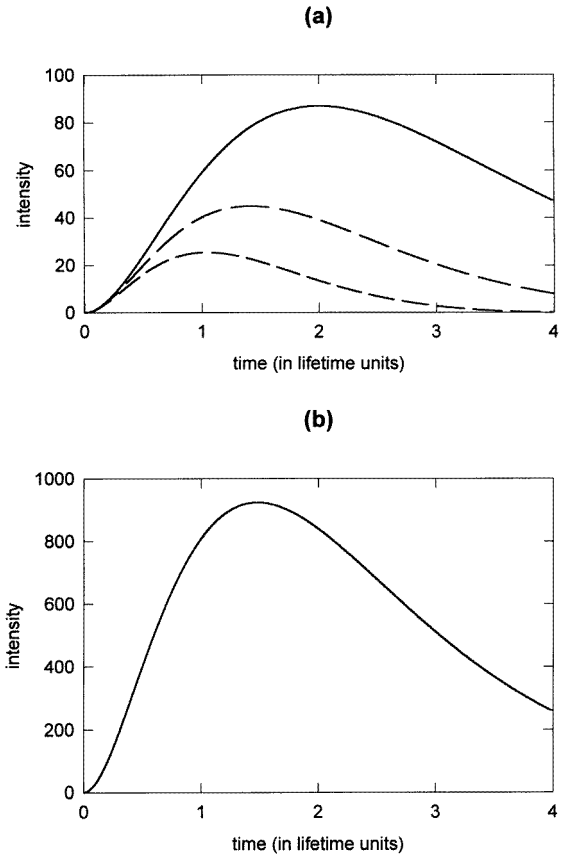

Figure 4. Calculated results for the processes in which there is emission of radiation, with recoil from resonant ^{57}Fe nuclei in the 'absorber'. (a) Three contributions to the total. The solid curve is for the 'first' layer (nucleus $n = 1$), the long-dashed curve is for the 'tenth' layer (nucleus $n = 10$), and the shorter-dashed curve is for the 'last' layer (nucleus) assuming the total effective number of resonant nuclei (layers) is $N = 20$. (b) The total contribution to the time-dependent intensity from all layers (nuclei) in the absorber which decay with recoil.

3.1.3. Processes involving the inelastic channel. Next consider the inelastic channel. That is, instead of detecting the gamma ray, detect the conversion electron or, perhaps more practically, the resulting x-ray. The processes that need to be considered are those in

which the source emits radiation without recoil and this radiation reaches the nth absorber nucleus where the radiation is absorbed but the decay takes place by internal conversion. This process is incoherent with the same process occurring at the pth absorber nucleus. All such processes ending in internal conversion are incoherent since these 'paths' are distinguishable. In order to calculate the amplitude for such a process one must consider all the indistinguishable paths that lead to excitation of the nth absorber nucleus. Notice that in the prior calculation, involving an absorber nucleus emitting radiation with recoil, the form of the process is essentially identical to that treated in this case. So now it is easy to treat the inelastic channel. One simply needs to incorporate the conversion-electron channel width (Γ_c) into the analogous equations above, and remove the factor $(1 - f)$.

The time-dependent intensity due to the inelastic channel processes in the absorber is (A38)

$$I_{(ie)}(t') = \sum_{n=1}^{N} |\psi_{n(ie)}(t')|^2 \tag{16}$$

where the 'n' corresponds to the nth nucleus in the 'absorber' in exactly the same fashion as for the processes with recoil discussed above. Evaluating $\psi_{n(ie)}(t')$ for the condition when the source and absorber nuclei are in exact resonance gives (A39)

$$\psi_{n(ie)}(t') = \sqrt{\frac{\Gamma_c}{2\hbar}}\, \mathrm{e}^{-\mathrm{i}(\omega_0 - \mathrm{i}(\Gamma/2\hbar))t'} \sum_{m=0}^{n-1} \binom{n-1}{m} \left(\frac{f\Gamma_r t'}{2\hbar}\right)^{m+1} \frac{(-1)^{m+1}}{(m+1)!}. \tag{17}$$

This result is nearly identical to (15). In an actual comparison with experiment additional care is required here because, for each internal-conversion process, the emitted conversion electron or x-ray must pass through the rest of the absorber to reach the detector. Each 'n' contribution must be weighted properly. In detecting the conversion electron itself this is particularly important since a conversion electron usually cannot travel far in the sample. As in ordinary conversion-electron Mössbauer spectroscopy, one would observe only a surface layer effect.

4. Application of the theory

The model contains several physical parameters: the lifetime of the nuclear-resonant level, and the radiative and conversion-electron widths of that level. There is an additional parameter N, the effective number of resonant absorber nuclei, representing the absorber thickness. If the theory is to be useful, this effective number of nuclei (or layers) N in the 'absorber' must be related to the actual resonant thickness parameter of the sample. As seen in figures 1 and 2 the one-dimensional model and the classical optical model give identical results. Noting this observation, one can determine the relationship (see the acknowledgments) between N in the one-dimensional model and β in the classical optical model. To do this consider the thin-absorber limit. In the classical optical model, when the source and absorber are in resonance, this amounts to expanding the J_0 Bessel function. In the one-dimensional model the thin 'absorber' limit is set by putting $N = 1$. Comparing the results gives

$$N = \frac{\beta\Gamma}{2f\Gamma_r} \tag{18}$$

where all the terms have been previously defined. Once N is determined for a given sample, all processes can be calculated.

Consider the analysis of TDMS data using both the classical optical and one-dimensional models. In the classical optical model calculation [3] one simply gives weights to two components: the recoil-free component, exhibiting speed-up, and a normal lifetime component due to recoil events in the source. The component that may be missing is due to recoil processes that take place in the *absorber* itself. This component can be accounted for in the one-dimensional model. Notice from figure 4 that the effect occurs at times well after $t = 0$. (This is not true for nuclear-resonant scattering using synchrotron radiation. The incoherent scattering in non-forward directions is evident [10].)

Furthermore, and most importantly, the size of this contribution in TDMS depends strongly on the geometry of the experimental configuration. In order to make some comparison, figure 5 gives simulations of TDMS results using both types of calculation and two different geometrical configurations. The absorber has a nuclear-resonant thickness $\beta = 3.2$ ($N = 20$). The coherent forward scattering goes forward but the incoherent scattering goes into all 4π steradians. The relative contribution from the incoherent scattering depends on the solid angle subtended by the detector relative to the source and absorber. In figure 5(a) the solid angle subtended by the detector relative to the source is $\Omega_{source} = 0.2$ steradians and relative to the absorber $\Omega_{absorber} = 2\pi$ steradians. The difference between the predictions of the two models is large in such a case. For figure 5(b) the solid angles are $\Omega_{source} = 0.004$ steradians and $\Omega_{absorber} = 0.4$ steradians. The difference now is quite small.

5. Summary

Dr Stan Ruby has, for many years, been advocating the idea of tracing multiple-scattering paths. He gave a discussion of his ideas at *ICAME'93*. The present paper is a step in that direction. A one-dimensional model for nuclear-resonant forward scattering of gamma radiation is developed which gives a unified physical picture of the scattering processes involved in time-domain nuclear-resonant spectroscopy. For the recoil-free processes the model gives a result identical to that obtained using the classical optical model. One simply needs to establish the relationship between the effective number of resonant nuclei N in the model and the resonant thickness parameter β used in the classical optical model (see (18)). The calculations using this new model are not difficult and the physical picture of the processes is clear. The model shows that the speed-up effect is a consequence of the 180° phase shift that occurs for each recoil-free scattering (hopping) event. Dynamical beats, the additional bumps that appear in time-domain spectra when using a thick sample, are also due to this same interference effect. In this case the double-scattering (two-hop) processes dominate the single-scattering (one-hop) processes at later times.

In TDMS the contribution of the incoherent scattering in the absorber is usually small. However, the first observation of incoherent scattering using synchrotron radiation [10] showed obvious speed-up effects. Furthermore, in order to interpret the results of [10] using the classical optical model, the authors were forced to divide the sample into layers to obtain an averaged speed-up component which was then added to a normal lifetime component. Similar synchrotron radiation calculations and discussion have been performed by others [11–13]. The one-dimensional model is able to handle the incoherent processes as a natural extension.

During the past ten years there have been interesting extensions of the TDMS method. These include time chopping [14] and phase modulation (gamma echo) [15, 16] of the source beam. These techniques can be analysed using the one-dimensional model but this will be done in a later paper.

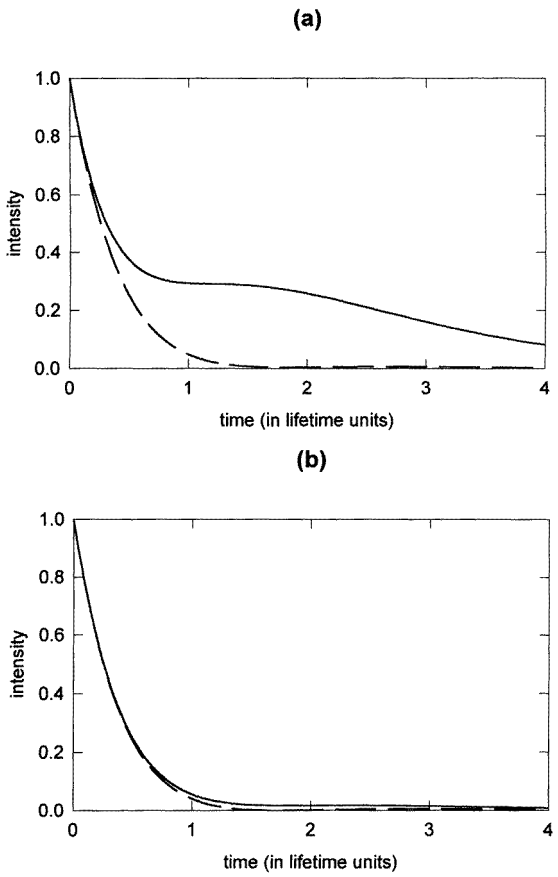

Figure 5. A calculated comparison of TDMS spectra using the one-dimensional model (the solid curves) and the classical optical model (the dashed curves) neglecting the electronic time resolution of an actual experimental apparatus. These calculations are for the ^{57}Fe case assuming an effective number of resonant nuclei $N = 20$ for the one-dimensional model, and the corresponding $\beta = 3.2$ for the classical optical model. (a) The solid angle subtended by the detector relative to the source is $\Omega_{source} = 0.2$ steradians and relative to the absorber $\Omega_{absorber} = 2\pi$ steradians. (b) The solid angles are $\Omega_{source} = 0.004$ steradians and $\Omega_{absorber} = 0.4$ steradians.

The source–matter (source–absorber) system has a unique feature in which there is complete phase memory when considering nuclear-resonant recoil-free scattering processes. The result is completely independent of the wavelength of the radiation assuming only the resonance condition. Furthermore the positions and spacings between the N effective nuclei do not enter the result, so questions such as raised by Dicke [17] when discussing superradiance are irrelevant for the recoil-free processes.

Consider a radioactive source which contains resonant nuclei in the ground state. In such a case the nuclear excitation may 'wander'. The excitation that was originally on one nucleus may spread out over many nuclei. The 'speed-up' effect is due to this collective behaviour [18, 19]. Thus it appears that the radiative transition rate has been enhanced. Does this mean, for example, that under certain conditions the cross section for stimulated emission can be larger than that normally assumed?

Acknowledgments

I would like to thank Professor J Odeurs for discussions and a careful reading of the manuscript and Professor Romain Coussement for his discussions and enthusiastic encouragement. Thanks also to Professor S M Harris for sending a copy of his technical report. Several very helpful discussions were held with Professor Rocco Schiavilla. Furthermore, the author would like to acknowledge the initial support from Dr Jack Davis of the Naval Research Laboratory. Finally I would like to thank the unknown referees for their helpful comments. In particular, one referee's remark led to (18).

Appendix

Starting from (6)–(10), solve (9) for $D_p(\omega)$ and substitute into (6); the summation over p can be converted into an integral which can be evaluated according to the usual prescription

$$\sum_p \frac{A(\omega)|H_p|^2}{(\omega-\omega_p+\mathrm{i}\varepsilon)\overline{h}^2} \Rightarrow \frac{L}{2\pi\overline{h}}\int \mathrm{d}p \frac{A(\omega)|H_p|^2}{(\omega-\omega_p+\mathrm{i}\varepsilon)\overline{h}^2} \tag{A1}$$

using the symbolic relation

$$\frac{1}{(x-a)\mp\mathrm{i}\varepsilon} = P\frac{1}{(x-a)} \pm \mathrm{i}\pi\delta(x-a) \tag{A2}$$

where P indicates the principal value. The principal value part of the integral gives rise to an energy shift, which we will neglect, while the imaginary part produces the conversion-electron width. The result is

$$\left(\omega-\omega_0+\mathrm{i}\frac{\Gamma_c}{2\overline{h}}\right)A(\omega) = 1+\sum_k \frac{B_k(\omega)H_k}{\overline{h}}. \tag{A3}$$

Solving (10) for $E_{mp}(\omega)$ and substituting into (8) gives, after using the same procedure,

$$\left(\omega-\omega_0'+\mathrm{i}\frac{\Gamma_c}{2\overline{h}}\right)C_m(\omega) = \sum_k \frac{B_k(\omega)H_k}{\overline{h}}\,\mathrm{e}^{\mathrm{i}kx_m} \tag{A4}$$

again neglecting the frequency shift. Solving (7) for $B_k(\omega)$ and substituting into (A4) gives

$$\left(\omega-\omega_0'+\mathrm{i}\frac{\Gamma_c}{2\overline{h}}\right)C_m(\omega) = \frac{A(\omega)}{\overline{h}^2}\sum_k \frac{|H_k|^2\,\mathrm{e}^{\mathrm{i}kx_m}}{(\omega-\omega_k+\mathrm{i}\varepsilon)} + \sum_{m'}\frac{C_{m'}(\omega)}{\overline{h}^2}\sum_k \frac{|H_k|^2\,\mathrm{e}^{\mathrm{i}k(x_m-x_{m'})}}{(\omega-\omega_k+\mathrm{i}\varepsilon)}. \tag{A5}$$

Converting the sums on k to integrals and evaluating the integrals in the same manner as described above gives

$$\left(\omega-\omega_0'+\mathrm{i}\frac{\Gamma}{2\overline{h}}\right)C_m(\omega) = -\mathrm{i}\frac{\Gamma_r}{2\overline{h}}\,\mathrm{e}^{\mathrm{i}(\omega/c)x_m}A(\omega) - \mathrm{i}\frac{\Gamma_r}{2\overline{h}}\sum_{m'=1}^{m-1} C_{m'}\,\mathrm{e}^{\mathrm{i}(\omega/c)(x_m-x_{m'})}\Theta(x_m-x_{m'}) \tag{A6}$$

where Γ_r is the radiative width, Γ (the total width) $=\Gamma_r+\Gamma_c$ and $\Theta(x)$ is the Heaviside step function defined to be zero when the argument is less than zero and one when it is greater than or equal to zero. Using an iteration procedure it is not difficult to show that

$$C_m(\omega) = -\mathrm{i}\frac{\Gamma_r}{2\overline{h}}\frac{\mathrm{e}^{\mathrm{i}(\omega/c)x_m}A(\omega)}{(\omega-\omega_0'+\mathrm{i}(\Gamma/2\overline{h}))}\left[1-\mathrm{i}\frac{\Gamma_r}{2\overline{h}}\frac{1}{(\omega-\omega_0'+\mathrm{i}(\Gamma/2\overline{h}))}\right]^{m-1}. \tag{A7}$$

Substituting the expression for $C_m(\omega)$ into (7), one can find $B_k(\omega)$

$$B_k(\omega) = \frac{H_k^*}{\hbar}\frac{1}{(\omega-\omega_0+\mathrm{i}(\Gamma/2\hbar))(\omega-\omega_k+\mathrm{i}\varepsilon)}\left[1+\sum_{m=0}^{N-1}\mathrm{e}^{\mathrm{i}((\omega/c)-k)x_{m+1}}\alpha(1+\alpha)^m\right] \tag{A8}$$

where

$$\alpha = -\mathrm{i}\frac{\Gamma_r}{2\hbar}\frac{1}{(\omega-\omega_0'+\mathrm{i}(\Gamma/2\hbar))}. \tag{A9}$$

Before taking the Fourier transform, one can express $B_k(\omega)$ in a symbolic form according to (A8).

$$B_k = B_k^{source} + \sum_{m=0}^{N-1} B_{k,m}^{absorber}. \tag{A10}$$

The first term is due to the source alone while the summation term describes the effect due to each absorber nucleus according to the value of m, which not only labels the location of that particular absorber nucleus, but also includes the effects of other absorber nuclei that are positioned 'in front' of the one located at $x = x_m$. In this context, 'in front' means that the source radiation passes those nuclei before reaching the one located at $x = x_m$. Now take the Fourier transform of $B_k(\omega)$ to obtain $b_k(t)$ according to (3). Representing the radiation as a plane wave allows one to write

$$\psi_r(x,t) = \sum_k \frac{\mathrm{e}^{\mathrm{i}(kx-\omega_k t)}}{\sqrt{L}} b_k(t) = \frac{\sqrt{L}}{2\pi}\int_{-\infty}^{\infty}\mathrm{d}k\,\mathrm{e}^{\mathrm{i}(kx-\omega_k t)}b_k(t). \tag{A11}$$

The plane wave due to the source alone is

$$\psi_r^{source}(t) = -\mathrm{i}\sqrt{L}\frac{H_k^*}{\hbar c}\,\mathrm{e}^{-\mathrm{i}(\omega_0-\mathrm{i}(\Gamma/2\hbar)(t-(x/c))}\Theta\left(t-\frac{x}{c}\right)\Theta(x). \tag{A12}$$

These step functions indicate that if the detector is located at x, there must be sufficient time elapsed in order for the radiation to reach the detector. The other terms in (A10) are more complicated and related to the effect due to the absorber nuclei. The general term in the Fourier transform looks like

$$b_{k,m}(t) = -\frac{1}{2\pi\,\mathrm{i}}\frac{H_k^*}{\hbar}\int_{-\infty}^{\infty}\mathrm{d}\omega\,\mathrm{e}^{\mathrm{i}(\omega_k-\omega)t}\,\mathrm{e}^{\mathrm{i}((\omega/c)-k)x_{m+1}}\frac{\alpha(1+\alpha)^m}{(\omega-\omega_0+\mathrm{i}(\Gamma/2\hbar))(\omega-\omega_k+\mathrm{i}\varepsilon)} \tag{A13}$$

where α is defined above. It turns out that the only pole in this integral that contributes to the final answer is the one where $\omega = \omega_k - \mathrm{i}\varepsilon$. This condition arises because the detector position at x must be greater than the position of the last absorber nucleus. Notice that the positions of the absorber nuclei do not appear in the answer. This can be seen, physically, by noting that the path from the source to the detector is fixed so the specific locations of the absorbing nuclei are irrelevant: the only thing that is important is how many nuclei are encountered in a given process. Evaluating the general term gives

$$b_{k,m}(t) = \frac{H_k^*}{\hbar}\left(-\mathrm{i}\frac{\Gamma_r}{2\hbar}\right)\frac{1}{(\omega_k-\omega_0+\mathrm{i}(\Gamma/2\hbar))(\omega_k-\omega_0'+\mathrm{i}(\Gamma/2\hbar))}\sum_{n=0}^{m}\binom{m}{n}\alpha^n(\omega_k) \tag{A14}$$

where now

$$\alpha(\omega_k) = -\mathrm{i}\frac{\Gamma_r}{2\hbar}\frac{1}{(\omega_k-\omega_0'+\mathrm{i}(\Gamma/2\hbar))}$$

and the factor m over n is the binomial coefficient. Grouping another $\alpha(\omega_k)$ factor, one can write

$$b_{k,m}(t) = \frac{H_k^*}{\hbar} \frac{1}{(\omega_k - \omega_0 + \mathrm{i}(\Gamma/2\hbar))} \sum_{n=0}^{m} \binom{m}{n} \alpha^{n+1}(\omega_k). \tag{A15}$$

Thus,

$$b_k(t) = b_k^{source}(t) + \sum_{m=0}^{N-1} b_{k,m}(t). \tag{A16}$$

Next, one needs to calculate the plane-wave amplitude according to the prescription used above. (It is at this point where the terms omitted in the solution of (A13) do not contribute.) One can identify terms in an obvious fashion

$$\psi_r(t) = \psi_r^{source}(t) + \sum_{m=0}^{N-1} \sum_{n=0}^{m} \psi_{r,m}^{(n)}(t). \tag{A17}$$

The general term that needs evaluation is

$$\psi_{r,m}^{(n)}(t) = \frac{\sqrt{L}}{2\pi} \frac{H_k^*}{\hbar c} \int_{-\infty}^{\infty} \mathrm{d}\omega_k \frac{\mathrm{e}^{-\mathrm{i}\omega_k(t-(x/c))}}{(\omega_k - \omega_0 + \mathrm{i}(\Gamma/2\hbar))} \binom{m}{n} \alpha^{n+1}(\omega_k). \tag{A18}$$

As one works through the details an interesting and physically meaningful pattern develops. If one re-expresses the result in terms of amplitudes depending on how many absorber nuclei participate in a particular process, the following form emerges:

$$\psi_r(t') = \psi_r^{source}(t') + N a_1(t) + \binom{N}{2} a_2(t') + \binom{N}{3} a_3(t') + \cdots + \binom{N}{N} a_N(t') \tag{A19}$$

where for $n = 1$

$$a_1(t') = -\mathrm{i}\sqrt{L} \frac{H_k^*}{\hbar c} \mathrm{e}^{-\mathrm{i}(\omega_0 - \mathrm{i}(\Gamma/2\hbar))t'} \left(-\mathrm{i} \frac{\Gamma_r}{2\hbar} \right) \left[\frac{1 - \mathrm{e}^{-\mathrm{i}(\omega_0' - \omega_0)t'}}{(\omega_0 - \omega_0')} \right] \tag{A20}$$

and for $n = 2, 3, \ldots, N$

$$a_n(t') = -\mathrm{i}\sqrt{L} \frac{H_k^*}{\hbar c} \mathrm{e}^{-\mathrm{i}(\omega_0 - \mathrm{i}(\Gamma/2\hbar))t'} \left(-\mathrm{i} \frac{\Gamma_r}{2\hbar} \right)^n \times \left[\left(1 - \mathrm{e}^{-\mathrm{i}(\omega_0' - \omega_0)t'} - \mathrm{e}^{-\mathrm{i}(\omega_0' - \omega_0)t'} \sum_{p=1}^{n-1} \frac{(-\mathrm{i}t')^p (\omega_0 - \omega_0')^p}{p!} \right) (\omega_0 - \omega_0')^{-n} \right]. \tag{A21}$$

If one evaluates these expressions *on* resonance, i.e., when ω_0 equals ω_0', the total amplitude takes on the following simple form:

$$\psi_r(t') = \psi_r^{source}(t') \left[1 + \sum_{n=1}^{N} \binom{N}{n} \left(-\frac{\Gamma_r t'}{2\hbar} \right)^n \frac{1}{n!} \right] \tag{A22}$$

and the intensity reaching the detector as a function of time is

$$I_r(t') = c|\psi_r(t')|^2. \tag{A23}$$

A.1. Recoil-free processes

I can immediately write down the result for the completely recoil-free process as

$$\psi_{fr}(t') = \sqrt{\frac{f\Gamma_r}{2\overline{h}}}\,\mathrm{e}^{-\mathrm{i}(\omega_0-\mathrm{i}(\Gamma/2)\overline{h})t'}\left[1+\sum_{n=1}^{N}\binom{N}{n}a_{fn}(t')\right] \tag{A24}$$

where for $n = l$

$$a_{f1}(t') = \sqrt{\frac{f\Gamma_r}{2\overline{h}}}\,\mathrm{e}^{-\mathrm{i}(\omega_0-\mathrm{i}(\Gamma/2)\overline{h})t'}\left(-\mathrm{i}\frac{f\Gamma_r}{2\overline{h}}\right)\left[\frac{1-\mathrm{e}^{-\mathrm{i}(\omega_0'-\omega_0)t'}}{(\omega_0-\omega_0')}\right]. \tag{A25}$$

and for $n = 2, 3, \ldots, N$

$$a_{fn}(t') = \sqrt{\frac{f\Gamma_r}{2\overline{h}}}\,\mathrm{e}^{-\mathrm{i}(\omega_0-\mathrm{i}(\Gamma/2)\overline{h})t'}\left(-\mathrm{i}\frac{f\Gamma_r}{2\overline{h}}\right)^n \times\left[\left(1-\mathrm{e}^{-\mathrm{i}(\omega_0'-\omega_0)t'}-\mathrm{e}^{-\mathrm{i}(\omega_0'-\omega_0)t'}\sum_{p=1}^{n-1}\frac{(-\mathrm{i}t')^p(\omega_0-\omega_0')^p}{p!}\right)(\omega_0-\omega_0')^{-n}\right]. \tag{A26}$$

The time-dependent intensity of radiation reaching the detector when the source nucleus and absorber nuclei are in resonance, and one considers only the recoil-free processes, is

$$I_{fr}(t') = \frac{f\Gamma_r}{2\overline{h}}\,\mathrm{e}^{-(\Gamma/\overline{h})t'}\left[1+\sum_{n=1}^{N}\binom{N}{n}\left(\frac{-f\Gamma_r t'}{2\overline{h}}\right)^n\frac{1}{n!}\right]^2. \tag{A27}$$

A.2. Processes with recoil

There is an amplitude for the source to emit radiation with recoil and the resulting radiation to reach the detector. This process is incoherent with the other processes discussed, so compute its intensity.

$$I_{(1-f)source}(t') = \frac{(1-f)\Gamma_r}{2\overline{h}}\,\mathrm{e}^{-(\Gamma/\overline{h})t'}. \tag{A28}$$

Furthermore, any absorber nucleus can absorb radiation without recoil and then emit radiation with recoil. To calculate this amplitude one must consider all the processes that lead to excitation of that particular absorber nucleus which can then emit with recoil. In order to do this I add a sixth amplitude to the set of equations (6)–(10)

$$(\omega-\omega_{k'}+\mathrm{i}\varepsilon)F_{mk'}(\omega) = \frac{C_m(\omega)H_{k'}^*\,\mathrm{e}^{-\mathrm{i}kx_m}}{\overline{h}} \tag{A29}$$

where $H_{k'}^*$ corresponds to the operator for emission with recoil. Proceeding in the same fashion as above one arrives at the following amplitude for the process resulting in emission with recoil from the nth absorber nucleus,

$$\psi_{nk'}(t') = \sum_{m=0}^{n-1}\binom{n-1}{m}a_{(1-f)m+1}(t') \tag{A30}$$

where for $n = 1$

$$a_{(1-f)1}(t') = \sqrt{\frac{(1-f)\Gamma_r}{2\overline{h}}}\,\mathrm{e}^{-\mathrm{i}(\omega_0-\mathrm{i}(\Gamma/2)\overline{h})t'}\left(-i\frac{f\Gamma_r}{2\overline{h}}\right)\left[\frac{1-\mathrm{e}^{-\mathrm{i}(\omega_0'-\omega_0)t'}}{(\omega_0-\omega_0')}\right]. \tag{A31}$$

and for $n = 2, 3, \ldots, N$

$$a_{(1-f)n}(t') = \sqrt{\frac{(1-f)\Gamma_r}{2\overline{h}}}\,\mathrm{e}^{-\mathrm{i}(\omega_0-\mathrm{i}(\Gamma/2)\overline{h})t'}\left(-\mathrm{i}\frac{f\Gamma_r}{2\overline{h}}\right)^n \times\left[\left(1-\mathrm{e}^{-\mathrm{i}(\omega_0'-\omega_0)t'}-\mathrm{e}^{-\mathrm{i}(\omega_0'-\omega_0)t'}\sum_{p=1}^{n-1}\frac{(-\mathrm{i}t)^p(\omega_0-\omega_0')^p}{p!}\right)(\omega_0-\omega_0')^{-n}\right]. \tag{A32}$$

Since these recoil processes are incoherent with each other and with the recoil-free processes, i.e. they correspond to distinguishable 'paths', the intensity for each process must be calculated separately. Thus, the final intensity for the processes with recoil is

$$I_{(1-f)}(t') = I_{(1-f)source}(t') + \sum_{n=1}^{N}|\psi_{nk'}(t')|^2. \tag{A33}$$

Evaluating $\psi_{nk'}(t')$ for the condition when the source and absorber are in exact resonance gives

$$\psi_{nk'}(t') = \sqrt{\frac{(1-f)\Gamma_r}{2\overline{h}}}\,\mathrm{e}^{-\mathrm{i}(\omega_0-\mathrm{i}(\Gamma/2)\overline{h})t'}\sum_{m=0}^{n-1}\binom{n-1}{m}\left(\frac{f\Gamma_r t'}{2\overline{h}}\right)^{m+1}\frac{(-1)^{m+1}}{(m+1)!}. \tag{A34}$$

A.3. Processes involving the inelastic channel

The processes that need to be considered are those in which the source emits radiation without recoil and this radiation reaches the mth absorber nucleus where the radiation is absorbed without recoil but the decay takes place by internal conversion. All such processes are incoherent. To calculate these amplitudes consider all the indistinguishable paths that lead to excitation of the mth absorber nucleus. One needs to incorporate the conversion-electron channel width into the analogous equations from the recoil case above and remove the factor $(1-f)$. This gives

$$\psi_{n(ie)}(t') = \sum_{m=0}^{n-1}\binom{n-1}{m}a_{(ie)m+1}(t') \tag{A35}$$

where for $n = 1$

$$a_{(ie)1}(t') = \sqrt{\frac{\Gamma_c}{2\overline{h}}}\,\mathrm{e}^{-\mathrm{i}(\omega_0-\mathrm{i}(\Gamma/2)\overline{h})t'}\left(-\mathrm{i}\frac{f\Gamma_r}{2\overline{h}}\right)\left[\frac{1-\mathrm{e}^{-\mathrm{i}(\omega_0'-\omega_0)t'}}{(\omega_0-\omega_0')}\right] \tag{A36}$$

and for $n = 2, 3, \ldots, N$

$$a_{(ie)n}(t') = \sqrt{\frac{\Gamma_c}{2\overline{h}}}\,\mathrm{e}^{-\mathrm{i}(\omega_0-\mathrm{i}(\Gamma/2)\overline{h})t'}\left(-\mathrm{i}\frac{f\Gamma_r}{2\overline{h}}\right)^n \times\left[\left(1-\mathrm{e}^{-\mathrm{i}(\omega_0'-\omega_0)t'}-\mathrm{e}^{-\mathrm{i}(\omega_0'-\omega_0)t'}\sum_{p=1}^{n-1}\frac{(-\mathrm{i}t')^p(\omega_0-\omega_0')^p}{p!}\right)(\omega_0-\omega_0')^{-n}\right]. \tag{A37}$$

These inelastic channel processes are incoherent with each other so the final intensity for the inelastic channel processes is

$$I_{(ie)}(t') = \sum_{n=1}^{N}|\psi_{n(ie)}(t')|^2. \tag{A38}$$

Evaluating $\psi_{n(ie)}(t')$ for the condition when the source and absorber are in exact resonance gives

$$\psi_{n(ie)}(t') = \sqrt{\frac{\Gamma_c}{2\overline{h}}}\, \mathrm{e}^{-\mathrm{i}(\omega_0 - \mathrm{i}(\Gamma/2)\overline{h})t'} \sum_{m=0}^{n-1} \binom{n-1}{m} \left(\frac{f\Gamma_r t'}{2\overline{h}}\right)^{m+1} \frac{(-1)^{m+1}}{(m+1)!}. \tag{A39}$$

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