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Enhanced resolution in Mössbauer spectroscopy

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Abstract. Using a resonant detector in Mössbauer spectroscopy can result in a spectral linewidth that is 1.46Γ, where Γ is the linewidth of the excited-state nuclear level. As is well known, the minimum linewidth obtained in conventional Mössbauer experiments is 2Γ. The quantum mechanical calculation using a nuclear resonant detector, which predicts this result, is presented. The fundamental equations describing the system are solved by means of perturbation theory in the frequency domain. The model system is taken to consist of a source nucleus, an absorber nucleus, and the resonant-detector nucleus. As noted, the minimum linewidth obtained in a Mössbauer spectrum taken under these conditions is found to be appreciably smaller than the linewidth obtained in a conventional Mössbauer set-up. Thus the conversion-electron, resonant-detector scheme may be used to advantage in experiments requiring the highest possible energy resolution.

1. Introduction

A conventional Mössbauer-effect apparatus [1] consists of a radioactive source, an absorber containing the same type of nuclei in the ground state, and a radiation detector such as a proportional counter. One measures the transmitted gamma radiation as a function of the relative velocity of the source with respect to the absorber. It is well known that the minimum linewidth obtainable in such a conventional transmission Mössbauer experiment is twice the natural linewidth of the excited state. In this paper it will be shown that another experimental set-up will lead to a linewidth that is actually smaller. In experiments requiring the highest possible energy resolution, this approach may prove useful. The difference, between the proposed experimental set-up and a conventional Mössbauer set-up, is due to the nature of the detector. Instead of a conventional radiation detector, such as a proportional counter or a NaI detector, one uses a nuclear 'resonant' detector. The resonant detector contains ground-state nuclei, which are in resonance with the stationary excited-state source nuclei. In this experimental configuration, as also often used in a conventional set-up, the absorber is moved with respect to the source and the detector which are both stationary. The experiment proceeds by detecting the conversion electrons generated in the resonant detector as a function of the velocity of the absorber.

A quantum mechanical model system consisting of a source nucleus, an absorber nucleus, and a resonant-detector nucleus, along with the associated resonant photons and conversion electrons, will be used to analyse the results from the proposed experimental configuration. Since we are interested in a minimum linewidth, it is reasonable to go to the thin absorber limit. In our model this corresponds to describing the absorber as having one resonant nucleus.

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2. Quantum mechanical model

The general method used in this paper is discussed in Heitler [2], Harris [3], and in a more recent article [4]. The method applies quantum mechanical perturbation theory in the frequency domain to obtain a set of coupled equations. The Hamiltonian of the system is divided into two parts. H_0 is the unperturbed part which describes the evolution of the nuclear states, the free radiation field and conversion electrons in the absence of coupling between the nuclear states, the radiation field and the conversion electrons. The eigenstates of H_0 correspond to nuclear states, the states of the free radiation field, and the states of the conversion electrons. The perturbing part of the Hamiltonian is denoted by V and is responsible for making transitions between the nuclear levels.

The actual state of the system is then expressed as

$$|\Psi(t)\rangle = \sum_l a_l(t)\, \mathrm{e}^{-\mathrm{i}E_l t/\hbar}|\varphi_i(0)\rangle \tag{1}$$

where $|\varphi_l(0)\rangle$ is an eigenstate of H_0 and E_l the corresponding energy. Solving the Schrödinger equation leads to a set of coupled differential equations relating the expansion coefficients $a_l(t)$. Heitler [2] has shown that in order to satisfy the boundary conditions at $t = 0$, one has the following set of coupled differential equations.

$$\mathrm{i}\hbar\frac{\mathrm{d}a_l}{\mathrm{d}t} = \sum_q a_q(t)\, \mathrm{e}^{\mathrm{i}(\omega_l-\omega_q)t}\langle\varphi_l(0)|V|\varphi_q(0)\rangle + \mathrm{i}\hbar\delta_{1n}\delta(t) \tag{2}$$

where $\omega_l - \omega_q = (E_l - E_q)/\hbar$, δ_{1n}, is the Kronecker delta and $\delta(t)$ the Dirac delta function. Introducing the Fourier transform [3] and inserting the causality condition [2] allows (2) to be re-written in the frequency domain

$$(\omega - \omega_l + \mathrm{i}\varepsilon)A_l(\omega) = \sum_q A_q(\omega)\frac{V_{lq}}{\hbar} + \delta_{1n} \tag{3}$$

where V_{lq} is the matrix element inducing a transition from the qth unperturbed state to the lth unperturbed state

$$V_{lq} = \langle\varphi_l(0)|V|\varphi_q(0)\rangle. \tag{4}$$

The advantage of the set of equations (3), is that we now have only a linear, coupled system.

Consider a system whose initial condition is as follows. We have an excited nucleus, having energy $\hbar\omega_0$, at the origin of a coordinate system, an absorber nucleus, whose excited-state energy is $\hbar\omega_0'$, in the ground state (nucleus 1) situated at position $\vec{r}_1$, and another ground-state nucleus, having an excited-state energy $\hbar\omega_0$, at position $\vec{r}_2$ (nucleus 2). The last nucleus, nucleus 2, represents the resonant detector. The absorber nucleus is situated between the source nucleus and the resonant-detector nucleus. The evolution of the quantum system composed of the three nuclei, the radiation field and the conversion electrons will be investigated below. In the following, we will assume that the recoilless fractions [1] of the emission and absorption processes are one, which is however, not an essential hypothesis.

In applying the mathematical formalism to our system, the following amplitudes can be defined:

- $A(\omega)$ is the amplitude corresponding to the source nucleus excited ($\hbar\omega_0$), the other two nuclei in the ground state, and no photons or conversion electrons present;
- $B_k(\omega)$ is the amplitude corresponding to all nuclei in ground state, a photon of wavenumber $\vec{k}$ and energy $\hbar\omega_{\vec{k}}$ present, and no conversion electrons present;
- $C_i(\omega)$ ($i = 1, 2$) is the amplitude corresponding to nucleus at position $\vec{r}_i$ excited, all other nuclei in ground state, and no photons or conversion electrons present;

- $D_{\vec{p}}(\omega)$ is the amplitude corresponding to having a conversion electron from the source nucleus present, having momentum $\vec{p}$, all nuclei in the ground state, and no photons present;
- $E_{i\vec{p}}(\omega)$ is the amplitude corresponding to having a conversion electron from nucleus i ($i = 1, 2$) present, all nuclei in ground state, and no photons present.

At $t = 0$, only the source nucleus is excited, so we have the following set of coupled equations

$$(\omega - \omega_0 + \mathrm{i}\varepsilon)A(\omega) = 1 + \sum_{\vec{k}} \frac{H_{\vec{k}}}{\hbar} B_{\vec{k}}(\omega) + \sum_{\vec{p}} \frac{H_{\vec{p}}}{\hbar} D_{\vec{p}}(\omega) \tag{5}$$

$$(\omega - \omega_{\vec{k}} + \mathrm{i}\varepsilon)B_{\vec{k}}(\omega) = \frac{H^*_{\vec{k}}}{\hbar} A(\omega) + \frac{H^*_{\vec{p}}}{\hbar} \mathrm{e}^{-\mathrm{i}\vec{k}\cdot\vec{r}_1} C_1(\omega) + \frac{H^*_{\vec{k}}}{\hbar} \mathrm{e}^{-\mathrm{i}\vec{k}\cdot\vec{r}_2} C_2(\omega) \tag{6}$$

$$(\omega - \omega_0' + \mathrm{i}\varepsilon)C_1(\omega) = \sum_{\vec{k}} \frac{H_{\vec{k}}}{\hbar} \mathrm{e}^{\mathrm{i}\vec{k}\cdot\vec{r}_1} B_{\vec{k}}(\omega) + \sum_{\vec{p}} \frac{H_{\vec{p}}}{\hbar} \mathrm{e}^{\mathrm{i}\vec{p}\cdot\vec{r}_1/\hbar} E_{1\vec{p}}(\omega) \tag{7}$$

$$(\omega - \omega_0 + \mathrm{i}\varepsilon)C_2(\omega) = \sum_{\vec{k}} \frac{H_{\vec{k}}}{\hbar} \mathrm{e}^{\mathrm{i}\vec{k}\cdot\vec{r}_2} B_{\vec{k}}(\omega) + \sum_{\vec{p}} \frac{H_{\vec{p}}}{\hbar} \mathrm{e}^{\mathrm{i}\vec{p}\cdot\vec{r}_2/\hbar} E_{2\vec{p}}(\omega) \tag{8}$$

$$(\omega - \omega_{\vec{p}} + \mathrm{i}\varepsilon)D_{\vec{p}}(\omega) = \frac{H^*_{\vec{p}}}{\hbar} A(\omega) \tag{9}$$

$$(\omega - \omega_{\vec{p}} + \mathrm{i}\varepsilon)A(\omega)E_{i\vec{p}}(\omega) = \frac{H^*_{\vec{p}}}{\hbar} \mathrm{e}^{-\mathrm{i}\vec{p}\cdot\vec{r}_1/\hbar} C_i(\omega) \qquad i = 1,\ 2 \tag{10}$$

where $H_{\vec{k}}$ and $H^*_{\vec{k}}$ are the matrix element corresponding to absorption and emission, respectively, of a photon having wave vector $\vec{k}$. $H_{\vec{p}}$ and $H^*_{\vec{p}}$ are similarly defined for the conversion electron. Equations (5)–(10) can be interpreted in a straightforward manner. For example, equation (6) describes the amplitude $B_k(\omega)$ for production of a photon having wave vector $\vec{k}$. This can occur when the source nucleus emits a photon as described by the first term on the right-hand side of equation (6), or when either nucleus at position $\vec{r}_i$ ($i = 1$ or 2) emits a photon, the second and third terms on the right-hand side equation (6). The functions $\mathrm{e}^{-\mathrm{i}\vec{k}\cdot\vec{r}_i}$ ($i = 1,\ 2$) designate the position where the photon emission takes place. The other equations can be interpreted in a similar manner.

3. Solution of the equations

One can proceed to solve these equations. After making various substitutions, the resulting summations can be converted to integrals and evaluated using well-known prescriptions [2–4]. In [4] the problem corresponding to an initial condition of an excited source nucleus and N resonant absorber nuclei in the ground state has been treated. It has been shown [4] that this problem has a closed-form solution if one restricts the calculation to forward scattering.

The fundamental equation for the source radiation is

$$\left(\omega - \omega_0 + \mathrm{i}\frac{\Gamma}{2\hbar}\right) A(\omega) = 1 \tag{11}$$

where the total width of the excited state, Γ, is the sum of the conversion-electron and radiative widths

$$\Gamma = \gamma_c + \gamma_r. \tag{12}$$

The radiative width in this one-dimensional problem is

$$\gamma_r = \frac{2L}{\hbar c} |H_k(\omega)|^2. \tag{13}$$

The simple form of equation (11) is due to the fact that, when the radiation goes from the source nucleus to one of the ground state nuclei, we do not expect radiation to be re-radiated back to the source nucleus. Macroscopic distances separate the absorber and detector nuclei from the source nucleus thus any such processes are extremely rare.

Going back to the time domain, it can be easily shown using equation (11) that

$$a(t) = \mathrm{e}^{\Gamma t/2\hbar}. \tag{14}$$

The source nucleus decays in the normal exponential fashion uninfluenced by the absorber and detector 'nuclei'. The remaining equations become

$$\left(\omega - \omega_0' + \mathrm{i}\frac{\Gamma}{2\hbar}\right) C_1(\omega) = -\frac{\mathrm{i}\gamma_r}{2\hbar}\mathrm{e}^{\mathrm{i}(\omega/c)x_1} A(\omega) \tag{15}$$

$$\left(\omega - \omega_0 + \mathrm{i}\frac{\Gamma}{2\hbar}\right) C_2(\omega) = -\frac{\mathrm{i}\gamma_r}{2\hbar}\mathrm{e}^{\mathrm{i}(\omega/c)x_2} A(\omega) - \frac{\mathrm{i}\gamma_r}{2\hbar}\mathrm{e}^{\mathrm{i}(\omega/c)(x_2-x_1)} C_1(\omega). \tag{16}$$

The physical reason why equation (15) is different from equation (16) is due to the locations of the absorber nucleus and the detector nucleus relative to the source nucleus. The radiation coming from the detector nucleus, which is 'downstream' with respect to the positive direction of the x axis from the absorber nucleus, does not 'return' to re-excite the absorber nucleus. The conclusion of a careful analysis shows that only radiation coming from nuclei 'upstream' from a given nucleus can give a contribution to the excitation of that nucleus, i.e. the radiation effectively only goes forward. When considering an absorber having many nuclei, the same conclusion holds [4]. The fact, noted above, that radiation coming from the absorber nuclei does not get back to the source nucleus is consistent with this conclusion.

Summarizing our results, we have

$$A(\omega) = \frac{1}{\omega - \omega_0 + \mathrm{i}(\Gamma/2\hbar)} \tag{17}$$

$$C_1(\omega) = -\frac{\mathrm{i}\gamma_r}{2\hbar}\frac{1}{\omega - \omega_0' + \mathrm{i}(\Gamma/2\hbar)}\mathrm{e}^{\mathrm{i}(\omega/c)(x_1)} A(\omega) \tag{18}$$

$$C_2(\omega) = -\frac{\mathrm{i}\gamma_r}{2\hbar}\frac{1}{\omega - \omega_0 + \mathrm{i}(\Gamma/2\hbar)}\mathrm{e}^{\mathrm{i}(\omega/c)(x_2)} A(\omega) - \frac{\mathrm{i}\gamma_r}{2\hbar}\frac{1}{\omega - \omega_0 + \mathrm{i}(\Gamma/2\hbar)}\mathrm{e}^{\mathrm{i}(\omega/c)(x_2-x_1)} C_1(\omega). \tag{19}$$

Substituting equations (17) and (18) into equation (19) gives

$$C_2(\omega) = -\frac{\mathrm{i}\gamma_r}{2\hbar}\frac{1}{(\omega - \omega_0 + \mathrm{i}(\Gamma/2\hbar))^2}\mathrm{e}^{\mathrm{i}(\omega/c)x_2}\left(1 - \frac{\mathrm{i}\gamma_r}{2\hbar}\frac{1}{\omega - \omega_0' + \mathrm{i}(\Gamma/2\hbar)}\right). \tag{20}$$

The probability of having nucleus 2, the 'detector nucleus', excited is found from equation (20).

$$|C_2(\omega)|^2 = \frac{\gamma_r^2}{4\hbar^2}\frac{1}{((\omega - \omega_0)^2 + (\Gamma^2/4\hbar^2))^2} + \frac{\gamma_r^3(\gamma_r - 2\Gamma)}{16\hbar^4}\frac{1}{((\omega - \omega_0)^2 + (\Gamma^2/4\hbar^2))^2}\frac{1}{((\omega - \omega_0')^2 + (\Gamma^2/4\hbar^2))}. \tag{21}$$

The probability of having a conversion electron produced in the 'detector' is

$$\int_{-\infty}^{+\infty} |C_2(\omega)|^2\,\mathrm{d}\omega = \frac{\gamma_r^2}{4\hbar^2}\int_{-\infty}^{+\infty}\left((\omega - \omega_0)^2 + \frac{\Gamma^2}{4\hbar^2}\right)^{-2}\mathrm{d}\omega + \frac{\gamma_r^3(\gamma_r - 2\Gamma)}{16\hbar^4}\int_{-\infty}^{+\infty}\left((\omega - \omega_0)^2 + \frac{\Gamma^2}{4\hbar^2}\right)^{-2}\left((\omega - \omega_0')^2 + \frac{\Gamma^2}{4\hbar^2}\right)^{-1}\mathrm{d}\omega. \tag{22}$$

The first integral is a standard integral. One finds for the first integral a constant value $\pi\hbar\gamma_r^2/\Gamma^3$. This constant value does not depend on ω_0' the frequency corresponding to the absorber nucleus. This contribution corresponds to the conversion-electron production due to radiation coming directly from the source to the detector nucleus without interacting with the absorber. In the recorded spectrum this will give a constant background analogous to the background observed in a conventional Mössbauer experimental result. The second integral, which will be denoted $I(\omega_0, \omega_0')$, can be calculated by means of contour integration. One finds

$$I(\omega_0, \omega_0') = \frac{\pi\gamma_r^3(\gamma_r - 2\Gamma)}{4\hbar\Gamma^3}\frac{12a^2 + \Delta\omega_0^2}{(4a^2 + \Delta\omega_0^2)^2} \tag{23}$$

where a and $\Delta\omega_0$ are defined by the following expressions

$$a = \frac{\Gamma}{2\hbar} \tag{24}$$

$$\Delta\omega_0 = \omega_0 - \omega_0'. \tag{25}$$

Equation (23) gives the conversion-electron distribution as a function of the relative frequency of the absorber nucleus relative to the source (and resonant detector) nucleus. This distribution looks like a Lorentzian, although it is not a real one. The full width at half maximum of this distribution can be calculated easily. One finds a value 1.463Γ. This value is close to 1.47Γ, advanced in [5], where a heuristic approach has been given based the calculation of the transmission integral. Also in [5], experiments are presented where this narrowing was confirmed using ^{119}Sn. More recently this narrowing has been observed [6] again with ^{119}Sn.

In order to check the consistency of our model, one can calculate the resulting linewidth in a conventional Mössbauer procedure using this approach. To do this, consider only the source and absorber nucleus. Then imagine doing the conventional experiment by detecting the conversion electrons produced in the absorber as a function of ω_0'. The result is obtained by solving equation (18) for $C_1(\omega)$, finding the absolute value squared and integrating over ω, as done above for $C_2(\omega)$. The full width at half maximum is found to be 2Γ as expected.

4. Summary and conclusions

We can summarize our results as follows. A quantum mechanical theory of a particular Mössbauer-effect set-up, making use of a conventional source, a conventional absorber and a resonant detector, consisting of ground-state nuclei with the same environment as the source nuclei, has been developed. The model is based on perturbation theory in frequency domain. The equations for the complete system of resonant nuclei, gamma radiation, and conversion electrons have been solved. When counting-conversion electrons, produced by the resonant detector nuclei, as a function of the Doppler velocity of the absorber with respect to the source and resonant detector, the minimum linewidth is 1.463Γ. This linewidth is appreciably less than the minimum linewidth of 2Γ obtained in a conventional Mössbauer-effect experiment. Thus, for those experiments that profit from obtaining the highest possible energy resolution, the proposed experimental configuration can be used to advantage. The theory can be extended to the case where both the absorber and detector have an arbitrary thickness. Under such conditions line broadening will naturally occur, as in conventional Mössbauer spectroscopy, but the advantage of relatively narrower lines, compared to the conventional set-up, will remain.

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