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Integral and Differential Moessbauer Spectra Taking Account of Metastable Atomic States

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Abstract

Integral and time-differential Moessbauer spectra taking account of metastable atomic states are calculated properly. The results are consistent with the calculations by Hamermesh which are applicable to a system having no metastable atomic state.

Recently some workers are interested in the study of unusual metastable atomic or chemical configurations of the sample atom by Moessbauer emission spectroscopy (see [1-5], for example). Their results are well explained by assuming some metastable states with lifetimes comparable to the life of the Moessbauer nuclear state. In these cases, owing to interference between the transitions of metastable atomic and nuclear states, it is not reasonable to assume that the spectrum consists of a superposition of some isolated Lorentzians or time-differential lines. In order to analyze the experimental data correctly, it is important to introduce a Moessbauer spectrum by taking account of the interference.

Kankeleit [6] calculated the integral and time-differential spectra influenced by metastable atomic states using a simple model, which involves four states and a decay by two transitions, as shown in Fig.1.

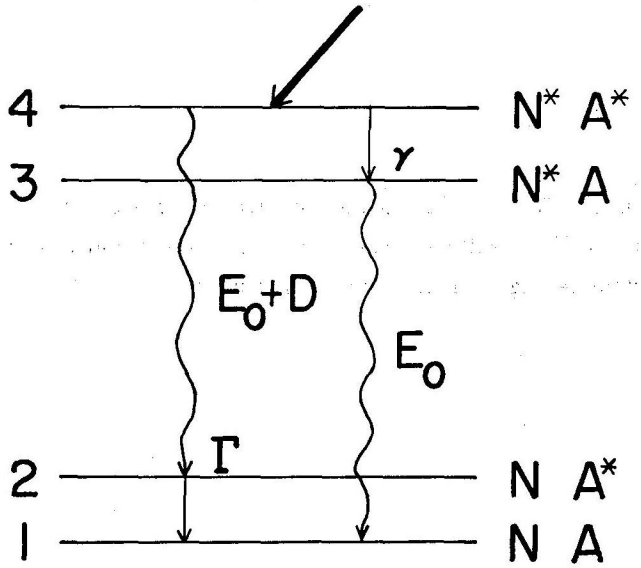

Fig.1 Decay scheme of Moessbauer state. N and A mean the ground state of a nucleus and atom, respectively. The symbol * indicates an excited state of the nucleus or atom. Energies of the 3-1 and 4-2 transitions are E_0 and E_0+D, respectively. Γ and γ represent decay rates of the nuclear and atomic system, respectively.

Upon reexamining the calculations, we find some incorrect procedures in the formulation which results in inconsistency with the results by Hamermesh [7] for a system with no metastable state. The amplitude of a gamma quantum can be written by help of the step function

$$\theta(t)=\begin{cases} 0 & t<0 \\ 1 & t\geqq 0 \end{cases} \qquad (1)$$

as

$$a(t,T) = \exp\left(i(E_0+D+i\Gamma/2)t\right)\left[\theta(t)-\theta(t-T)\right] + \exp\left(i(E_0+i\Gamma/2)t\right)\theta(t-T). \tag{2}$$

Here T is the time at which the excited atomic state decays and i is the imaginary unit. The amplitude of the gamma quantum at energy E becomes then

$$\begin{aligned} a(E,T) &= \int_{-\infty}^{\infty} \exp(-iET)\, a(t,T)\, dt \\ &= \frac{1}{i(E-E_0-D)+\Gamma/2}\left[1-\exp\left\{-i(E-E_0-D)T+\Gamma T/2\right\}\right] \\ &+ \frac{\exp\left\{-i(E-E_0)T+\Gamma T/2\right\}}{i(E-E_0)+\Gamma/2} \end{aligned} \tag{3}$$

Using Eq.(3), we obtain the emission spectrum,

$$\begin{aligned} W(E-E_0) = \frac{1}{N}\Bigg[& \frac{\gamma}{\Gamma+\gamma}\,\frac{1}{(E-E_0)^2+(\Gamma/2)^2} \\ & + \frac{\Gamma+2\gamma}{\Gamma+\gamma}\,\frac{1}{(E-E_0-D)^2+(\Gamma/2+\gamma)^2} \\ & - \frac{2(\Gamma+\gamma)\gamma}{D^2+(\Gamma+\gamma)^2}\,\frac{(E-E_0)(E-E_0+D)-\Gamma/2(\Gamma/2+\gamma)}{\left\{(E-E_0)^2+(\Gamma/2)^2\right\}\left\{(E-E_0)^2+(\Gamma/2+\gamma)^2\right\}}\Bigg], \end{aligned} \tag{4}$$

where N is the normalization factor. When we use a single line absorber of width Γ and of effective thickness t_a, an incident quantum with energy between E and $E+dE$ has the following probability to pass through the absorber :

$$\exp\left[-\frac{t_a(\Gamma/2)^2}{(E-E_0')^2+(\Gamma/2)^2}\right] dE,$$

where E_0' is the transition energy of the absorber, and we ignore non-resonant absorption. The transmission spectrum is then described by

$$M(V) = \int W(E-E_0)\exp\left[-\frac{t_a}{(\Gamma/2)^2(E-E_0'-V)^2+(\Gamma/2)^2}\right] dE, \tag{5}$$

where V is a Doppler velocity applied to the absorber. Taking the energy as the deviation from E_0', we obtain

$$M(V) = \int_{-\infty}^{\infty} W(E-d)\exp\left[-\frac{t_a(\Gamma/2)^2}{(E-V)^2+(\Gamma/2)^2}\right] dE, \tag{6}$$

where $d = E_o - E_o'$ is the isomer shift of the source nucleus of which atomic system is at the ground state. We can perform the intergration in Eq. (6) analytically when the absorber is thin enough to neglect the terms higher than t_a^2. The result is

$$M(V) = 1 - \frac{1}{N} P(V), \tag{7}$$

where the first term comes from the non-absorbed gamma quanta, and $P(V)$ shows the Moessbauer spectrum which is described as

$$P(V) = \frac{t_a \pi}{\Gamma} \left[\frac{\gamma}{\Gamma+\gamma} \frac{1}{V^2+\Gamma^2} + \frac{1}{(V-D)^2+(\Gamma+\gamma)^2} - \frac{\gamma}{D^2+(\Gamma+\gamma)^2} \frac{[(\Gamma+\gamma)V^2+(2\Gamma+\gamma)DV-\Gamma(\Gamma+\gamma)^2]}{(V^2+\Gamma^2)[V^2+(\Gamma+\gamma)^2]} \right] \tag{8}$$

The first and second terms of Eq.(8) correspond to the transition 3-1 and 4-2, respectively, in Fig.1. The transition 4-2 has the effective width $2(\Gamma+\gamma)$ instead of 2Γ. The last term comes from interference of these two transitions.

We try to calculate a time-differential Moessbauer spectrum in the following way. The energy spectrum of gamma rays is changed by passing through a thin resonance absorber of the line-width Γ, and then the amplitude $a(E, T)$ is modulated as follows [7] :

$$a'(E, T, V) = \exp\left[\frac{i\, t_a\, \Gamma/4}{E - E_o' - V - i\Gamma/2} \right] a(E, T). \tag{9}$$

By the inverse Fourier transformation, we can obtain the amplitude of the transmission gamma ray at a time t as

$$a'(t, T, V) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \exp(iEt)\, a'(E, T, V)\, dE. \tag{10}$$

We can perform this integration by a similar way as described in Appendix of Ref.[7], or by using the Laplace transformation. The result is, when $0 \leqq T \leqq t$

$$a'(t, T, V) = \exp(-\Gamma t/2) \sum_{n=0}^{\infty} \left\{ \left[-i \frac{2(V-d-D)t}{(t_a \Gamma t)^{1/2}} \right]^n Jn((t_a \Gamma t)^{1/2}) - \left[\exp(-i(V-d-D)T) \left[-i \frac{2(V-d-D)(t-T)}{(t_a \Gamma (t-T))^{1/2}} \right]^n - \exp(-i(V-d)T) \left[-i \frac{2(V-d)(t-T)}{(t_a \Gamma (t-T))^{1/2}} \right]^n Jn((t_a \Gamma (t-T))^{1/2}) \right\}, \tag{11}$$

and when $T > t$

$$a'(t, T, V) = \exp(-\Gamma t/2) \sum_{n=0}^{\infty} \left[-i \frac{2(V-d-D)t}{(t_a \Gamma t)^{1/2}}\right]^n Jn((t_a \Gamma t)^{1/2}). \quad (11)'$$

Here $Jn((t_a \Gamma t)^{1/2})$ is the Bessel function of order n. We can write Eqs. (11) and (11)' in more simple forms by using a function, $C(t, V)$, defined as

$$C(t, V) = \sum_{n=0}^{\infty} \left[i \frac{2Vt}{(t_a \Gamma t)^{1/2}}\right]^n Jn((t_a \Gamma t)^{1/2}). \quad (12)$$

Then Eqs. (11) and (11)' become

$$\begin{aligned} a'(t, T, V) = & \exp(-\Gamma t/2) \{ C(t, V-d-D) \\ & - \exp(-i(V-d-D)T)\, C(t-T, V-d-D) \\ & + \exp(-i(V-d)T)\, C(t-T, V-d) \}, \end{aligned} \quad (13)$$

when $0 \leqq T \leqq t$, and

$$a'(t, T, V) = \exp(-\Gamma t/2)\, C(t, V-d-D) \quad (13)'$$

when $T > t$. Here $\overline{C(t,V)}$ represents the complex conjugate (c.c.) of $C(t,V)$. The time-differential spectrum at the detector is given by

$$\begin{aligned} I(t, V) = & \int_0^{\infty} \gamma \exp(-\gamma T) \left| a'(t, T, V) \right|^2 dT \\ = & \exp(-\Gamma t) \left| C(t, V-d-D) \right|^2 \\ & + \exp(-\Gamma t) \int_0^t \gamma \exp(-\gamma T) \left[\left| \exp(-iDT)\, C(t-T, V-d) \right|^2 \right. \\ & - C(t, V-d-D) \{ \exp(i(V-d-D)T)\, C(t-T, V-d-D) \\ & \left. - \exp(i(V-d)T)\, C(t-T, V-d) \} - \text{c.c.} \right] dT. \end{aligned} \quad (14)$$

For extreme values of γ, Eqs. (8) and (14) approach to reasonable forms. That is, when $\gamma \to 0$,

$$P(V) \to \frac{t_a \Gamma}{4} \frac{1}{(V-d-D)^2 + \Gamma^2}, \quad (15)$$

and

$$I(t, V) \to \exp(-\Gamma t) \left| C(t, V-d-D) \right|^2, \quad (16)$$

and when $\gamma \to \infty$,

$$P(V) \to \frac{t_a \Gamma}{4} \frac{1}{(V-d)^2 + \Gamma^2}, \quad (15)'$$

and

$$I(t, V) \to \exp(-\Gamma t) \left| C(t, V-d) \right|^2, \tag{16}'$$

These agree well with the results of Hamermesh [7], which are applicable to a system having no metastable atomic state.

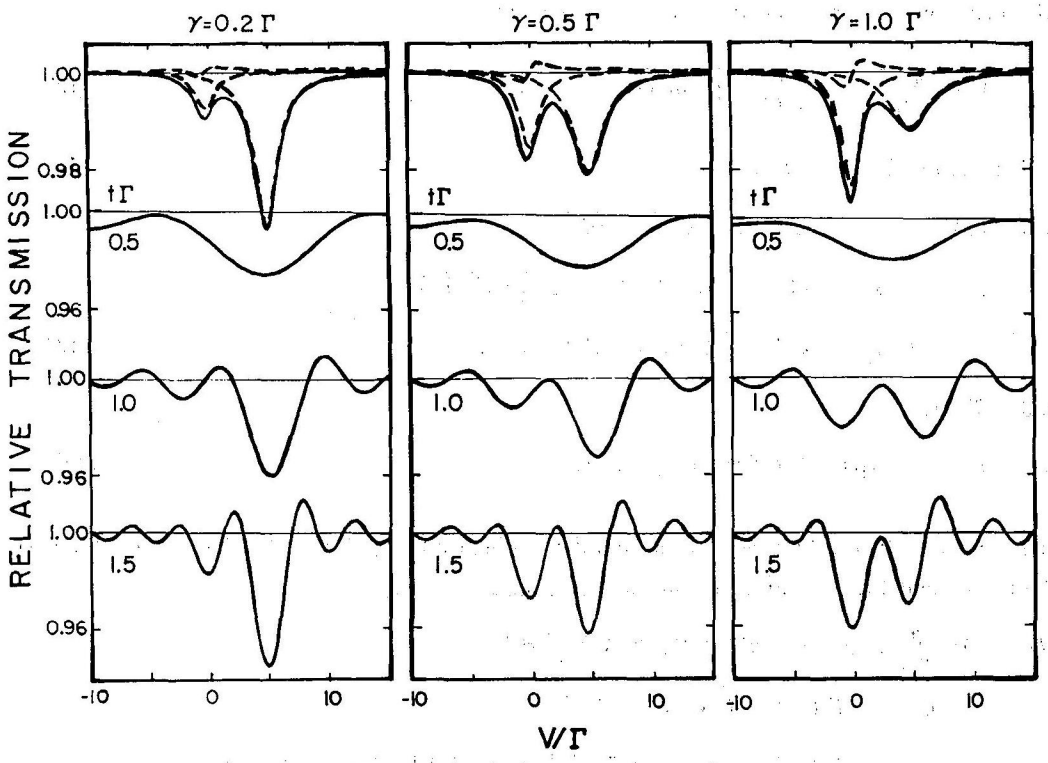

Fig.2 Integral and time-differential spectra. $d = 0$ and $t_a = 0.1$. In the integral spectrum, each term in Eq. (8) is indicated by a dashed line.

We show the results of the numerical calculations of Eqs.(8) and (14) in Fig.2 for the cases that $t_a = 0.1$, $d = 0$, $D = 5\ \Gamma$, $\gamma = 0.2\ \Gamma$, $0.5\ \Gamma$ and Γ, and $\Gamma\ t = 0.5$, 1, and 1.5. When we estimate the value of Eq. (14), we use the series

$$C(t, V) = \exp\left(i\left(Vt + \frac{t_a \Gamma}{4V}\right)\right) - \sum_{n=1}^{\infty} \left[i \frac{(t_a \Gamma t)^{1/2}}{2Vt}\right]^n Jn\left((t_a \Gamma t)^{1/2}\right) \tag{17}$$

for

$$\frac{(t_a \Gamma t)^{1/2}}{2Vt} < 1. \tag{18}$$

Equation (17) converges more rapidly than Eq. (12) when Eq. (18) is satisfied [7]. Evaluation of Eq. (14) is performed by disregarding the factor $\exp(-\Gamma t)$. In the integral spectra, each consists of two Lorentzians and a relatively small interference term. Accordingly, in an experimental situation, it is difficult to make clear the effect of metastable atomic states in the integral spectrum. In the time-differential spectra, we can see clearly that the peaks shift towards $V=0$ from D/Γ, particularly in the case of large γ. The peaks are not sometimes noticed at $V=0$ and/or at D/Γ owing to the oscillating feature of the time-filtering spectrum [8-10]. Numerical integration of Eq. (14) in regard to t from 0 to $10/\Gamma$ gives a good agreement with Eq. (8). Even in cases where such a simple model is applicable, a high speed computer is indispensable in order to analyze experimental spectra.

The general treatment applicable to complex cases, involving more complicated combinations of metastable nuclear and atomic states with different populations and lifetimes, hyperfine interactions, etc., can be developed as an extension of the present work.

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