MOSSBAUER EFFECT IN A DISPERSE SYSTEM

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It is shown that recoilless resonant absorption of gamma quanta by a nucleon contained in a Brownian particle is well described with the aid of a correlation function derivable from the Langevin equation for Brownian motion.

THE width of the Mossbauer line in a liquid is a function of the diffusion coefficient of the Mossbauer atoms [1,2]. In a disperse system (suspension) Brownian motion likewise leads to a line broadening [3-5] that has a nonlinear dependence on the diffusion coefficient.

Singwi and Sjolander [1] proposed to use the correlation-function formalism to describe resonant absorption of γ quanta by atoms of a liquid. The absorption cross section is determined in such an approach by the expression

$$\sigma_a(E)=\frac{\sigma_0\Gamma}{4\hbar}\int \exp\left[-i\omega t+i\varkappa\mathbf{r}-\frac{\Gamma}{2\hbar}|t|\right]G(\mathbf{r},t)\,d\mathbf{r}\,dt, \tag{1}$$

where κ is the γ-quantum momentum, $\omega=(E-E_0)/\hbar$, and σ_0, E_0, and Γ are the cross section at resonance, the resonance energy, and the level width; $G(\mathbf{r}, t)$ is the autocorrelation function of the absorbing nucleus.

If the motion of the atom is described by the simple diffusion equation, then we get, by substituting the self-diffusion function in (1) in lieu of $G(\mathbf{r}, t)$,

$$\sigma_a(E)=\frac{\sigma_0\Gamma}{4}\exp\left[-\frac{\hbar^2\varkappa^2}{8\mu kT}\right]\times\frac{\Gamma+2\hbar\varkappa^2D}{(E-E_0)^2+(\Gamma+2\hbar\varkappa^2D)^2/4}, \tag{2}$$

where μ is the mass of the atom and D is the diffusion coefficient.

It is more correct to use a correlation function G_L constructed on the basis of a solution of the Langevin equation for Brownian motion. For atomic motions, such a correlation function leads again to expression (2), as shown by Singwi and Sjolander [6] in an analysis of neutron scattering by a liquid. We shall show that in the case of Brownian particles the self-diffusion correlation function G_D and the function G_L give different dependences of the line width on the diffusion coefficient.

The motion of a Mossbauer atom contained in a Brownian particle can be resolved into the following components: motion of the particle center of gravity, rotation of the particle about the center of gravity, and vibrational motion of the atom relative to the lattice site (if the particles have a crystalline structure). Then the correlation function of the atom under consideration can be represented in the form of the product

$$G(\mathbf{r},t)=G_1(\mathbf{R},t)G_2(\mathbf{x},t)G_3(\boldsymbol{\xi},t), \tag{3}$$

where R is the coordinate of the particle center of gravity in the stationary system, ξ the displacement of the atom from the equilibrium position, and $\mathbf{x}+\boldsymbol{\xi}$ the radius vector of the atom in a moving system with origin at the center of gravity of the Brownian particle and with axes parallel to the axes of the stationary coordinate system.

Relation (3) follows from the treatment of the correlation function as being the probability of finding the particle at a certain point at an instant time t, if the particle was at the origin at the initial instant of time. The function $G_3(\xi, t)$ contained in (3) is the correlation function for the crystal. In the Gaussian approximation, the correlation function can be expressed in terms of the width function $\Gamma(t)$, which has the meaning of the mean-square displacement of the particle during the time t:

$$G(\mathbf{r},t)=(2\pi)^{-3/2}[\Gamma_1(t)\Gamma_2(t)\Gamma_3(t)]^{-3/2}\times\exp\left[-\frac{R^2}{2\Gamma_1(t)}\right]\exp\left[-\frac{x^2}{2\Gamma_2(t)}\right]\exp\left[-\frac{\xi^2}{2\Gamma_3(t)}\right]. \tag{4}$$

The function $G(\mathbf{r}, t)$ can also be represented in this approximation in the form

$$G(\mathbf{r},t)=(2\pi)^{-3/2}[\Gamma_1(t)+\Gamma_2(t)+\Gamma_3(t)]^{-3/2}\times\exp\left\{-\frac{r^2}{2[\Gamma_1(t)+\Gamma_2(t)+\Gamma_3(t)]}\right\}, \tag{5}$$

since $\overline{r^2}=\overline{R^2}+\overline{x^2}+\overline{\xi^2}$ by virtue of the independence of the components of the motion. It is easy

to verify by direct calculation the functions (4) and (5) lead to the same expression for the cross section.

For a thin absorber, the experimental spectrum is given by the expression

$$\sigma_e'(s)=\int_0^\infty \sigma_a(E)\,w_e(E)\,dE, \tag{6}$$

where $s = vE_0/c$, v the velocity of the source, and $w_e(E)$ is the emission spectrum:

$$w_e(E)=\frac{1}{2\pi\hbar}\int \exp\left[i(\varkappa r'-\omega t')-\frac{\Gamma}{2\hbar}\right]G_e(r',-t')\,dr'\,dt'. \tag{7}$$

Substituting (1) and (7) in (6) we have

$$\sigma_e'(s)=\frac{\sigma_0\Gamma}{4\hbar}\int \exp\left[-i\frac{t}{\hbar}s-\frac{\Gamma}{\hbar}|t|\right]$$
$$\times\exp\left\{-\frac{\varkappa^2}{2}[\Gamma_1(t)+\Gamma_2(t)+\Gamma_3(t)+\Gamma_e(t)]\right\}dt, \tag{8}$$

where $\Gamma_e(t)$ is the width function of the source. The Mossbauer part of the spectrum (8) is of the form

$$\sigma_e(s)=\frac{\sigma_0\Gamma}{4\hbar}\exp[-2W_e-2W_a]$$
$$\times\int \exp\left\{-i\frac{t}{\hbar}s-\frac{\Gamma}{\hbar}|t|-\frac{\varkappa^2}{2}[\Gamma_1(t)+\Gamma_2(t)]\right\}dt, \tag{9}$$

where $2W_e$ and $2W_a$ are the Debye-Waller factors of the source and of the substance of the Brownian particles.

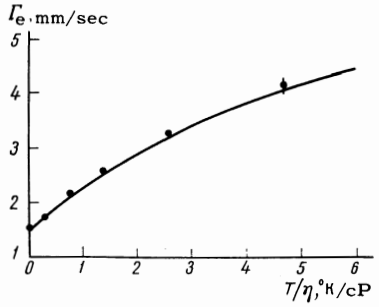
Using for the width functions $\Gamma_1(t)$ and $\Gamma_2(t)$ an expression that follows from the Langevin equation

$$\Gamma(t)=2D[t+g^{-1}(e^{-gt}-1)]+\hbar^2/4MkT,$$

where M is the mass of the Brownian particle and $g = kT/MD$, we get from (9)

$$\sigma_e(s)=\frac{\sigma_0 l}{2\hbar}\exp(-2W_e-2W_a)\exp\left(-\frac{\varkappa^2\hbar^2}{8MkT}\right)$$
$$\times e^{\beta}\sum_{n=0}^{\infty}\frac{(-1)^n}{n!}\beta^n\frac{n+\beta+l}{(\hbar^{-1}g^{-1}s)^2+(n+\beta+l)^2}. \tag{10}$$

Here $\beta=\kappa^2Dg^{-1}$ and $l=\hbar^{-1}g^{-1}\Gamma$.



In the case of atomic motions ($M \approx \mu$) we have $\beta \ll 1$, so that we can neglect all the series terms with $n \geq 1$. We get the results of Singwi and Sjolander for a liquid absorber: the line broadening is proportional to the diffusion coefficient. For Brownian particles $\beta \sim 1$, as a result of which the Mossbauer spectrum differs noticeably from the spectrum obtained with the simple-diffusion model.

From expression (10) we can determine the integral width of the spectrum:

$$\Gamma_e=\frac{1}{\pi\sigma_0/2}\int\sigma_e(s)\,ds$$
$$=g\hbar\exp[-2W_e-2W_a]$$
$$\times e^{-\beta}\left[\sum_{n=0}^{\infty}\frac{(-1)^n}{n!}\beta^n(n+\beta+l)^{-1}\right]^{-1} \tag{11}$$

The solid curve in the figure shows the calculated width of the spectrum in accord with formula (11) as a function of the diffusion coefficient, for tin-dioxide particles of 2.5×10^{-5} cm radius. The points show the experimental results. The absorber was a suspension of tin dioxide in glycerine or castor oil, and the source was $Sn^{119m}O_2$. The measurements were made with a constant-speed setup at different absorber temperatures.

Starting from the good agreement between the experimental and the calculated data, we can conclude that the influence of the Brownian motion on the resonant recoilless absorption of γ quanta can be satisfactorily described with the aid of a correlation function obtained from the Langevin equation.

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