Nuclear resonant absorption in long-lived isomeric transitions

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The temporal behavior of γ-ray emission and absorption from long-lived ($\tau > 1$ ms) nuclear states is modified qualitatively by interaction with the lattice, leading to homogeneous broadening. In contrast, for the usual short-lived ($\tau < 0.1$ ms) states, such nucleus-lattice interaction invariably leads to inhomogeneous broadening. It is shown that resonant emission and absorption with long-lived states can be observed provided the homogeneously broadened width, rather than the natural width, exceeds the solid-state-induced inhomogeneous width.

I. INTRODUCTION

There is general recognition in Mössbauer spectroscopy that when the lifetime τ of a nuclear state exceeds 10^{-3} s, the natural linewidth Γ_n of the corresponding Mössbauer transition becomes rather narrow ($\Gamma_n = \hbar/\tau = 10^{-12}$ eV) and the observation of the resonance absorption rather difficult on account of inhomogeneous broadening. A survey[1] of the literature indicates that typical values of inhomogeneous line broadening in the best crystals amounts to 10^{-12} eV or more. Consequently, if one attempts to observe the Mössbauer effect with a γ ray that has a natural linewidth of, for instance $\Gamma_n = 10^{-17}$ eV, crystalline inhomogeneities will broaden the line by six orders of magnitude, making the observation of the effect impossible. It was therefore a real surprise when two groups independently attempted to observe the Mössbauer effect with the 88-keV γ ray in ^{109}Ag ($\Gamma_n \approx 10^{-17}$ eV) diffused in Ag metal and claimed evidence for a small but positive effect.[2,3] This is indeed unexpected. In the present work we examine the issue at hand theoretically and identify the conditions under which resonant absorption from long-lived isomers can be possible.

II. DESCRIPTION OF LONG-LIVED NUCLEAR STATES IN A LATTICE

In considering the absorption or emission of γ rays, one must recognize that, when the lifetime is long, the interaction of the nuclei with the radiation field actually becomes weaker than the interaction of the nuclei with the crystalline lattice (consisting of electrons and nuclei). In the case of ^{109}Ag we can roughly estimate an upper limit to the interaction energy with the radiation field as the inverse of the lifetime (or the natural width) $\hbar/\tau_n$: 10^{-17} eV, while the interaction energy with the lattice is at least 10^{-13} eV. It is therefore not possible to handle the coupling of the nuclei with the radiation field as if the nuclei are free objects. In fact, we must consider the system of the nuclei plus lattice as a whole interacting with the radiation field. It is clear that this particular situation occurs only because of the extremely long lifetime or the extremely weak coupling to the radiation field. The most general procedure to treat the problem consists of solving the Hamiltonian of the total system, i.e., the observed quantum system plus its environment and the mutual interaction. When an observable on the selected or observed quantum system is needed, one must perform a partial trace over all reservoir observables. This procedure is, however, of purely theoretical interest, since it is difficult to take into account the nearly infinite number of reservoir degrees of freedom.

We assume the coupling between the surrounding and the nuclei to be weak enough so that the surrounding will induce small but frequent changes on the nuclei, which on average will have a small effect. When averaged over a time scale, long compared with a typical correlation time for the lattice, the effect of fluctuations on the nuclei can be smoothed out and described by a continuous function $f(t)$, with a mean fluctuation time T_R that is much longer than the correlation time of the lattice.

When we describe the interaction of the total system with the radiation field, the following approximation will be used. In calculating the emission and absorption amplitudes, we will replace the wave functions of the initial and/or final states by a nonstationary wave function of the type

$$\psi(\mathbf{x},t) = \psi(\mathbf{x},0) f(t) e^{-\Gamma_n t/2} e^{-i\omega_0 t}, \tag{1}$$

which includes the damping on account of nuclear decay and the function $f(t)$ displaying the effect of the surroundings on the nucleus. Here an analogy can be made. The function $f(t)$ describes the effect of fluctuations due to the surroundings, while the damping factor takes into account the effect of vacuum fluctuations on the nuclei. The damping factor leads to a transition of the nuclei from the excited to the ground state, while $f(t)$ does nothing of that kind. Even without a detailed knowledge of the function $f(t)$, reasonable assumptions lead to

specific bounds on this function. For example, for the nuclei and the fluctuating surroundings to be in equilibrium, $f(t)$ can only make random excursions around its averaged unit value, a point we shall return to later.

Conceptually, it is important to recognize that in considering usual Mössbauer transitions with short-lived states (10^{-6} s or less), one expects the damping factor $e^{-\Gamma_n t/2}$ to decrease much faster than variations of the function $f(t)$. This implies that for a particular nucleus that emits or absorbs a γ ray, an instantaneous value of $f(t)$ would apply. For the ensemble of nuclei, this variation in $f(t)$ from nuclei to nuclei introduces inhomogeneities and thus line broadening and a consequent loss in resonant absorption. For long-lived nuclear states on the other hand, the temporal behavior of a transition process is continuously being perturbed by the fluctuations $f(t)$ and the transition amplitude is modulated.

In a variety of experiments the temporal behavior of γ-ray emission and absorption from the usual short-lived Mössbauer states has been investigated.[4-6] One finds that one can qualitatively modify the emission and absorption amplitude by a controlled frequency change or phase change of either the emitter or the absorber, induced by the experimentalist in the laboratory. In long-lived systems, the lattice will have ample opportunity to induce such changes. As such changes are not under the direct control of the experimentalist and are more or less at random, the effect will not be as spectacular but nevertheless important.

At this stage it would be appropriate to specify mathematically the specific assumptions we make about the fluctuation function $f(t)$. We will realize a condition of equilibrium by requiring that the Fourier content of the fluctuations does not depend on the chosen time interval ΔT, if the interval is taken long enough compared to the mean fluctuation time T_R. We can assume that this time average is invariant with respect to a translation of the time interval ΔT, by requiring

$$f^{\Delta T}(\omega)=\frac{1}{\sqrt{2\pi}}\int_0^{\Delta T} f(t)e^{-i\omega t}dt = \frac{1}{\sqrt{2\pi}}\int_T^{T+\Delta T} f(t)e^{-i\omega t}dt ,$$

and also

$$f^{\Delta T}(\omega)=\frac{1}{\sqrt{2\pi}}\int_0^{\Delta T} f(t-t_0)e^{-i\omega t}dt . \quad (2)$$

Because of the equilibrium, we require that the system fluctuates around its free nuclear state and, consequently, the probability, averaged over a time interval ΔT, is normalized. The interval ΔT is chosen to be long compared to the mean fluctuation time, due to the lattice, but short compared to the lifetime of the long-lived state. This normalization procedure finally results in the total averaged power remaining constant, as shown below:

$$\frac{1}{\Delta T}\int_0^{\Delta T}\langle\psi(\mathbf{x},t)|\psi(\mathbf{x},t)\rangle dt = \langle\psi(\mathbf{x},0)|\psi(\mathbf{x},0)\rangle\frac{1}{\Delta T}\int_0^{\Delta T}|f(t)|^2dt=1$$

or

$$\frac{1}{\Delta T}\int_0^{\Delta T}|f(t)|^2dt=\frac{1}{\Delta T}\frac{1}{2\pi}\int_{\omega=0}^{\infty}|f^{\Delta T}(\omega)|^2d\omega=1 . \quad (3)$$

III. TRANSITION PROBABILITIES

In first-order perturbation theory, the transition amplitude as a function of the detuning $(\omega_0-\omega_0')$ is given as a Fourier transform of the transition matrix element[7]

$$A^T(\omega_0-\omega_0')=-\frac{i}{\hbar}\int_0^T\langle\lambda|V|\alpha\rangle e^{-i(\omega_0-\omega_0')t}dt . \quad (4)$$

The initial state $|\alpha\rangle$ with total energy $\hbar\omega_0$ consists of a photon, the nucleus in its ground state and the reservoir. The final state $|\lambda\rangle$ with total energy $\hbar\omega_0'$ consists of the excited nucleus plus the reservoir.

We also want to include in the formalism the effect of the fluctuations in the emitting system as well as the absorbing one. In the source, fluctuations will occur during emission, and the EM field associated with the photon will display these fluctuations, as considered earlier for absorption. Thus the emitted photon will appear as a superposition of free photonic states. This is homogeneous broadening.

The fluctuations on the photon are not correlated to the fluctuations on the absorbing nucleus. Therefore, we will accept that the matrix element will contain two uncorrelated fluctuation functions, given as $f(t)$ and $g(t)$, one referring to relaxation mechanism, intrinsic to the lattice of the emitter, while the other corresponds to the lattice of the absorber.

With fluctuations, the matrix element in Eq. (4), can be written as an unperturbed matrix element $\langle f|V|i\rangle$ and a time-dependent factor $f(t)g^*(t)e^{-\Gamma_n t}$

$$A^T(\omega_0-\omega_0')=-\frac{i}{\hbar}\langle f|V|i\rangle \times\int_0^T f(t)g^*(t)e^{-\Gamma_n t}e^{-i(\omega_0-\omega_0')t}dt . \quad (5)$$

The decay factor reflects the nuclear decay. Using Parcivals theorem we can write

$$A^T(\omega_0-\omega_0')=-\frac{i}{\hbar}\langle f|V|i\rangle \times\mathrm{Re}\int_0^{\infty} f^T(\omega)g^T(\omega-\omega_0+\omega_0')d\omega , \quad (6)$$

in which $f^T(\omega)$ and $g^T(\omega)$ are the Fourier transform of $f(t)e^{-(\Gamma_n/2)t}$ and $g(t)e^{-(\Gamma_n/2)t}$. We can shift the distribution $g(\omega)$ and $f(\omega)$ to obtain

$$\int_0^{\infty} f^T(\omega)g^T(\omega-\omega_0+\omega_0')d\omega = \int_0^{\infty} f^T(\omega-\omega_0')g^T(\omega-\omega_0)d\omega . \quad (7)$$

We recognize now that the amplitude for absorption is a convolution of the distribution $f^T(\omega-\omega_0')$ and $g^T(\omega-\omega_0)$. Such a convolution measures the overlap of both functions. Therefore one obtains a maximum when both have the same shape and the same position

$(\omega_0-\omega_0'=0)$. We thus conclude that Mössbauer absorption will be optimal when relaxation mechanisms in the emitter and the absorber are the same. This is the case for the ^{109}Ag self-absorption experiments.

We define the probability that a transition has taken place at time T as

$$P^T(\omega_0-\omega_0')=|A^T(\omega_0-\omega_0')|^2 \tag{8}$$

and the absorption rate at time T as

$$R^T(\omega_0-\omega_0')=\frac{P^{T+\Delta T}(\omega_0-\omega_0')-P^T(\omega_0-\omega_0')}{\Delta T}\ , \tag{9}$$

where ΔT is short compared to the nuclear lifetime but long compared to the fluctuations of the amplitude $A^T(\omega_0-\omega_0')$ introduced by the fluctuation process. This condition can only be fulfilled for long-lived nuclear states. We thus obtain

$$P^T(\omega_0-\omega_0')=\frac{1}{\hbar^2}|\langle f|V|i\rangle|^2\left|\int_0^T f(t)g^*(t)e^{-\Gamma_n t}e^{i(\omega_0-\omega_0')t}dt\right|^2\ , \tag{10}$$

which we can rewrite

$$P^T(\omega_0-\omega_0')=\frac{1}{\hbar^2}|\langle f|V|i\rangle|^2\int_0^T e^{-2\Gamma_n t'}g(t')f^*(t')\left[\int_0^T f(t)g^*(t)e^{-\Gamma_n(t-t')}e^{i(\omega_0-\omega_0')(t-t')}dt\right]dt'\ . \tag{11}$$

Because of the equilibrium condition we assume that at each t' value we can replace $f(t)g^*(t)$ in the integral over t by $f(t-t')g^*(t-t')$, which then becomes

$$\int_0^T f(\tau)g^*(\tau)e^{-\Gamma_n\tau}e^{i(\omega_0-\omega_0')\tau}d\tau,\quad \text{where } \tau=t-t'\ . \tag{12}$$

In that way the probability becomes a product of two integrals:

$$\frac{1}{\hbar^2}|\langle f|V|i\rangle|^2\int_0^T e^{-2\Gamma_n t'}g(t')f^*(t')dt'\int_0^T f(\tau)g^*(\tau)e^{-\Gamma_n\tau}e^{i(\omega_0-\omega_0')\tau}d\tau\ . \tag{13}$$

In the first integral the exponential decay is very slow compared to the fluctuation rate of $f^*(t')g(t')$, and therefore a replacement of $f^*(t')g(t')$ by its mean value, which is unity, will not introduce any serious error

$$\int_0^T e^{-2\Gamma_n t'}g(t')f^*(t')dt'\approx\int_0^T e^{-2\Gamma_n t'}dt'\ . \tag{14}$$

Then the absorption rate can be given as

$$R^T(\omega_0-\omega_0')=\frac{1}{\Delta T}\frac{1}{\hbar^2}|\langle f|V|i\rangle|^2\left[\int_0^{T+\Delta T}e^{-2\Gamma_n t'}dt'\int_0^{T+\Delta T}\cdots e^{i(\omega_0-\omega_0')\tau}d\tau-\int_0^T e^{-2\Gamma_n t'}dt'\int_0^T\cdots e^{i(\omega_0-\omega_0')\tau}d\tau\right]\ . \tag{15}$$

In the factor

$$\int_0^{T+\Delta T}e^{-2\Gamma_n t'}dt'=\int_0^T e^{-2\Gamma_n t'}dt'+\int_T^{T+\Delta T}e^{-2\Gamma_n t'}dt'\ , \tag{16}$$

we can neglect the last term when T is long compared to ΔT and ΔT is short compared to the nuclear lifetime, conditions that can be fulfilled for long-lived states only. This approximation allows us to reduce the expression in square brackets of Eq. (15) as follows:

$$[\ ]=\int_0^T e^{-2\Gamma_n t'}dt'\left[\int_0^{T+\Delta T}\cdots e^{i(\omega_0-\omega_0')\tau}d\tau-\int_0^T\cdots e^{i(\omega_0-\omega_0')\tau}d\tau\right]\ , \tag{17}$$

and the transition rate at time T simplifies to

$$R^T(\omega_0-\omega_0')=\frac{1}{\hbar^2}|\langle f|V|i\rangle|^2\frac{1}{2\Gamma_n}(1-e^{-2\Gamma_n T})\frac{1}{\Delta T}\int_T^{T+\Delta T}f(\tau)g^*(\tau)e^{-\Gamma_n\tau}e^{i(\omega_0-\omega_0')\tau}d\tau\ . \tag{18}$$

Using the time translation invariance, we can replace the integration from T to $T+\Delta T$ by an integration from 0 to ΔT. Using Parcivals theorem we then obtain

$$R^T(\omega_0-\omega_0')=\frac{1}{\hbar^2}|\langle f|V|i\rangle|^2\frac{1}{2\Gamma_n}(1-e^{-2\Gamma_n T})\frac{1}{\Delta T}\frac{1}{2\pi}\int_{\omega=0}^{\infty}f^{\Delta T}(\omega-\omega_0)g^{\Delta T}(\omega-\omega_0')^*d\omega\ . \tag{19}$$

The interesting feature of this particular formula appears when we consider the case in which the fluctuations in the emitter and the absorber, while uncorrelated, are assumed to have the same Fourier spectrum. Then the absorption rate is proportional to the average power,

$$R^T(\omega_0-\omega_0')\sim\frac{1}{\Delta T}\frac{1}{2\pi}\int_{\omega=0}^{\infty}f^{\Delta T}(\omega-\omega_0)^*f^{\Delta T}(\omega-\omega_0')d\omega\ , \tag{20}$$

which for zero detuning gives us

$$R^{T}(\omega_0-\omega_0'=0)\sim\frac{1}{\Delta T}\frac{1}{2\pi}\int_{\omega=0}^{\infty}|f^{\Delta T}(\omega-\omega_0)|^2 d\omega=\frac{1}{\Delta T}\int_0^{\Delta T}|f(t)|^2 dt\ . \tag{21}$$

We had previously assumed that the fluctuations are such that the time-averaged value of $|f(t)|^2$ is unity if the averaging is performed over a time interval that is long enough. We conclude that under such conditions, the zero detuning transition rate becomes insensitive to the width and the shape of the Fourier spectrum of the fluctuations. Furthermore when the detuning is small compared to the width of $f(\omega)$, but large compared to the natural width, we still obtain a large absorption rate. In the absence of fluctuations the absorption rate would be negligibly small as we recognized earlier.

IV. CONCLUSIONS

We conclude that in the ^{109}Ag self-absorption experiments, to observe the effect, it is merely sufficient for the inhomogeneous width (or detuning distribution) to be small compared with the width of the Fourier spectrum of the fluctuations, rather than with the natural linewidth. In ^{109}Ag, the T_2 relaxation time is of the order of milliseconds or less and we thus expect the "Fourier width" to be of the order of at least 10^{-13} eV. It is thus sufficient to reduce the distribution of static fields below that level and not below the more stringent level of 10^{-17} eV (which corresponds to the nuclear decay width) to see a positive Mössbauer effect.

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[1]P. Boolchand, J. Quant. Spectrosc. Radiat. Transfer **40**, 777 (1988).
[2]W. Wildner and U. Gonser, J. Phys. **40**, C2 47 (1979).
[3]G. Hoy and R. Taylor, J. Quant. Spectrosc. Radiat. Transfer **40**, 763 (1988).
[4]P. Helistö, E. Ikonen, T. Katila, and K. Riski, Phys. Rev. Lett. **49**, 1209 (1982).
[5]P. Helistö, I. Tittonen, H. Lippmaa, and T. Katila, Phys. Rev. Lett. **66**, 2037 (1991).
[6]S. L. Ruby and D. I. Bolef, Phys. Rev. Lett. **5**, 5 (1960).
[7]C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, *Processus d'Interaction Entre Photons et Atomes* (Inter Editions, Editions du CNRS, Paris, 1988).