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Semiclassical approach to nuclear gamma resonance in crystals

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Abstract. The emission of phonons produced under the absorption or emission of a γ quantum (or the scattering of an electron or a neutron) by an atomic nucleus in a crystal lattice is a Poisson process from the probability point of view. This approach enables, in particular, an expression for the Mössbauer effect probability to be derived in a simple and transparent manner without the cumbersome mathematical (quantum-mechanical) computations being involved.

1. Introduction

In just a few years following its discovery over four decades ago, nuclear gamma resonance, owing to Rudolf Mössbauer's pioneering work [1, 2] and the work of others, evolved into one of the most widely accepted nuclear method for investigating both the fundamental physical problems and applied problems in many areas of chemistry, geology, biology, medicine, metallurgy, etc. While the early research efforts mostly focused on various aspects of the phenomenon itself [3 – 7], the subsequent work in this field was mainly of applied nature [8 – 10]. Today, along with long-lived radioactive nuclei traditionally used as a source of γ radiation for the Mössbauer effect, alternative approaches such as synchrotron radiation, the use of neutron capture reactions, reactions with charged particles and the Coulomb excitation to directly excite Mössbauer transitions are receiving ever wider attention. Significant improvements have been made in the methodology of research. In fact, the Mössbauer effect has already become a mature curriculum subject for laboratory courses at highly-specialized university departments as well as at general physics laboratories [11, 12].

The aim the authors pursue in offering yet another theoretical approach to a well-known effect is a purely methodological one, namely, to show that many aspects of the processes with the absorption and emission of phonons can be described to our opinion with simpler and more physical concepts than they usually are. In presenting the material we attempted to avoid complicated quantum mechanical derivations but rather relied on more vivid, if less rigorous, general concepts. Unavoidably, some derivations due to other authors are repeated in the paper.

The scientific literature on the Mössbauer effect is enormous, as witnessed by the publication rate growth from about 1,000 per year in the early 70s to 2,000 per year by the early 90s. Since the modern electronic database allows, in principle, to obtain any required information from this huge source, only review papers and those original works needed for the discussion of the basic theoretical and experimental aspects of the effect are included in our reference list.

2. Basic features of nuclear gamma resonance

We begin by discussing the simplest properties of nuclear gamma resonance. If the energy of a photon hitting an atomic nucleus equals the energy difference between the nuclear ground state and one of its excited states, then the nucleus can absorb the photon thus making a transition into corresponding excited state. This process is possible only for γ rays of certain energies and is therefore of a resonant nature.

The energy dependence of the resonant γ-quantum absorption cross section is determined by the γ wavelength, the spins of the nuclei in their ground (I_o) and excited (I_e)

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states, and the probability of internal conversion:

$$\sigma_a(E) = \frac{2I_e + 1}{2I_o + 1} \frac{2\pi\hat{\lambda}^2}{1 + K} \frac{(\Gamma/2)^2}{(E - E_0)^2 + (\Gamma/2)^2}, \quad (1)$$

In this equation (known as the Lorentz formula or, in nuclear physics, as the Breit – Wigner formula), $K = \Gamma_e/\Gamma_\gamma$ is the internal conversion coefficient [$\Gamma_e/\Gamma = K/(1 + K)$ determining the probability that the nucleus having absorbed a photon falls into its ground state after giving energy to the atomic electrons], the parameter $\Gamma = \Gamma_e + \Gamma_\gamma$ is the full width of distribution at half maximum, E_0 is the nuclear transition energy, and $\hat{\lambda} = \lambda/2\pi$ is the reduced wavelength of the γ radiation.

It is well known that the width of an excited level Γ is related to its mean lifetime $\tau = T_{1/2}/\ln 2$ ($T_{1/2}$ being the half-life of the state) by the uncertainty relation

$$\Gamma\tau = \hbar. \quad (2)$$

Numerically, the natural linewidth is expressed in terms of the average lifetime and the decay period of the excited state as follows

$$\Gamma(\text{eV}) = 6.58 \times \frac{10^{-16}}{\tau(\text{s})} = 4.55 \times \frac{10^{-16}}{T_{1/2}(\text{s})}. \quad (3)$$

The natural width of a level is determined only by decay processes and characterizes both the width of the energy distribution of incident γ quanta and that of the nuclear absorption level. Only in the ideal case have the emission and absorption lines an ideal shape. As indicated previously [5], if the emission and absorption lines are Lorentzian in shape and have a width Γ, then the experimental curve is also a Lorentzian, and for a not very thick absorber the instrumental width is

$$\frac{\Gamma_{ins}}{\Gamma} = 2.00 + 0.27l, \quad 0 \leqslant l \leqslant 5. \quad (4)$$

The effective thickness of the absorber is $l = f'na\sigma_0 t$, where f' is the fraction of γ rays absorbed recoil-free, n is the atomic concentration, a is the relative abundance of nuclei absorbing γ rays resonantly, t is the absorber thickness, and the absorption cross section at resonance is $\sigma_0 = 2\pi\hat{\lambda}^2(1 + K)^{-1}(2I_e + 1)/(2I_0 + 1)$.

If other factors cause the level to broaden, the line shape is not necessarily Lorentzian. The experimentally determined resonance linewidth Γ_{exp} results from the superposition of the source and absorber lines. If the absorbers and sources are thin enough then, in the absence of vibrations, it is seen from Eqn (4) that the linewidth is practically twice the natural width, 2Γ. Increasing the absorber thickness broadens the resonance line considerably. This is because the quanta whose energy is close to the line maximum are strongly absorbed even in a thin absorber, and for such quanta increasing the thickness of the absorber has a smaller effect than for those at the wings of the line. Another possible reason for line broadening is the self-absorption of quanta in the source provided this latter contains resonantly absorbing nuclei. A significant contribution to the linewidth comes from the instrumental broadening related to the imperfection of measuring devices, in particular the vibrations of the source and the absorber (Doppler broadening) and the absorber's nonuniform motion with respect to the source.

At first sight, the resonant absorption of γ rays seems to be a widespread, easy-to-observe phenomenon. Indeed, one would expect that all one needs to detect it is to pass a beam of γ rays from a radioactive source through an absorber containing the same nuclei in an unexcited state. This is not the case, however, because the energy E_γ carried away by a γ quantum turns out to be less than E_0, the energy of the transition between the levels. A small but quite noticeable fraction of the energy is carried away by the nucleus which, as a result of recoil, starts moving in the opposite direction to that of the γ quantum that flies out.

Let us make some simple estimates. A nucleus emitting a γ quantum acquires a recoil momentum equal in magnitude to the momentum of the γ quantum. If the nucleus is free and initially at rest, the recoil energy R of the nucleus of mass M_n is

$$R = \frac{p^2}{2M_n} = \frac{E_\gamma^2}{2M_n c^2}. \quad (5)$$

Consider as an example the nucleus of lead, ^{119}Sn, in which the energy separation between the ground and the first excited levels is $E_0 = 23.8$ keV. The recoil energy in this case amounts to

$$R = \frac{E_\gamma^2}{2M_n c^2} \simeq \frac{E_0^2}{2M_n c^2} \simeq \frac{(2.38 \times 10^4)^2}{2 \times 119 \times 10^8} = 2.5 \times 10^{-3}\,\text{eV}. \quad (6)$$

The energy spent on the recoil of the nucleus that absorbs a γ quantum turns out to be exactly the same, so that the emission line is shifted by an amount R to the left, while the absorption line is shifted by the same amount to the right of E_0.

In discussing the effect of the shift R on the resonant absorption of γ rays, it should be noted that the quantity R is of little interest in itself. What is of interest is the relation between R and the width Γ of the corresponding resonance line. Resonant absorption is possible only if the emission and absorption spectra overlap, i.e. when one finds

$$2R \leqslant \Gamma. \quad (7)$$

This condition is almost never obeyed for γ transitions in free nuclei. Thus, for the ^{119}Sn nucleus considered above, $\Gamma \simeq 3 \times 10^{-8}$ eV, i.e. orders of magnitude less than R. Note that for optical transitions in atoms, the relation between R and Γ is markedly different. The transition energies in this case turn out to be four, and hence R eight, orders of magnitude less than for γ radiation, and the level widths in these processes are of the same order of magnitude. Resonant absorption is easily observed in this situation (R Wood, 1904).

In principle, the energy shift $2R$ can be compensated using the Doppler effect. For this, the emitting and absorbing nuclei must move with a velocity

$$v = \frac{c \cdot 2R}{E_\gamma} \quad (8)$$

relative to each other. For ^{119}Sn nuclei, the velocity required is $v \simeq 60$ m s^{-1}.

In real processes, the width of an emission (absorption) line is composed of the natural linewidth plus its Doppler width, of which the latter, due to the thermal motion of the

atoms, is dominant. Let us make necessary estimates. The Doppler shift of a level, Δ, may be calculated from non-relativistic formulas because u, the atomic thermal velocity, is much less than the speed of light. Thus we have

$$\Delta = \frac{u}{c} E_\gamma \simeq \frac{u}{c} E_0 \,. \tag{9}$$

To estimate the quantity u, recall that the average kinetic energy per degree of freedom (for motion towards or away from the absorber) is $(1/2)k_B T$. This gives $(1/2)M_n u^2 = (1/2)k_B T$, so that

$$u = \sqrt{\frac{k_B T}{M_n}}, \tag{10}$$

which when substituted to Eqn (9) and using Eqn (6) yields

$$\Delta = \sqrt{2Rk_B T}. \tag{11}$$

A more accurate calculation (see below) gives

$$\Delta = 2\sqrt{Rk_B T}. \tag{12}$$

At room temperatures $k_B T \simeq 1/40$ eV, so that for ^{119}Sn one finds

$$\Delta = 2\sqrt{2.3 \times 10^{-3} \times 2.5 \times 10^{-2}} = 1.5 \times 10^{-2} \text{ eV}$$

showing that the Doppler linewidth greatly exceeds the natural linewidth and in some cases — in ^{119}Sn, for example — it is found to be more than R. As a result of the Doppler broadening, there is a partial overlap between the emission and absorption lines, implying that a certain number of γ quanta exist for which the recoil R is compensated and resonant absorption can in principle be observed, albeit with very small probability.

The above picture of gamma emission and absorption processes involved in nuclear transitions is valid if the binding energy in the material is negligible, i.e. *if atomic nuclei may be considered free*. The characteristic features of nuclear resonant fluorescence we have described above were at the basis of experimental techniques in the pre-Mössbauer period.

We proceed now to the absorption and emission of γ quanta by nuclei making up a crystal lattice. The simplest — and least interesting — case is that in which the recoil energy exceeds the binding energy of a nucleus in the lattice. The binding of the nucleus is then of little significance, and no new phenomena occur. The energy required to displace a nucleus is quite large, ranging from 10 to 30 eV. Formula (6) shows that this situation is only possible for highly energetic γ quanta.

If the energy of the emitted γ quanta is $E_\gamma < 1$ MeV, the recoil energy is not sufficiently large to eject a nucleus from the crystal lattice, and the recoil momentum is, in one form or another, transferred to the entire crystal. The recoil energy most often is converted to sound vibrations of the lattice — an obvious transformation both from the quantum mechanical and classical points of view (in the former context, the recoil energy is transferred to sound vibration quanta, i.e. phonons).

Phonons are generated — and hence the recoil energy transforms to sound waves — more easily when more phonons are already available, i.e. at sufficiently high temperatures. The reason is that phonons obey Einstein–Bose statistics, and the number of phonons increases rapidly with temperature. According to quantum statistics, the probability for the generation of bosons of frequency ω is proportional to the number of such bosons that already exist (induced emission). At low temperatures, this 'boson amplification' process is unlikely, and if this mechanism is the one masking the phononless process, then the greater fraction of events can be described by Eqn (1). Indeed, in considering the energy and momentum balance for these cases, the mass of the nucleus in the recoil energy equation (5) should be replaced by the mass of the entire crystal, with the result that the recoil energy is reduced by 10 to 20 orders of magnitude and becomes small enough to be considered zero. It is commonly said that in such processes the recoil momentum is transferred to the entire crystal. This statement, even though it is widely used in the scientific literature and has in fact become part of scientific jargon in the field, is however incorrect because the momentum lost by a particle (γ quantum) interacting with one of the components of a composite target is always transferred to the entire target.

While elastic scattering does not excite the internal degrees of freedom of the target, strictly speaking the energy E_f of the particle that flies out is less than initial energy E_i by the recoil energy R, which is received by the entire target. The elastic (recoil-free) emission and absorption of γ quanta in solids has come to be known as the *Mössbauer effect*.

We now turn to discuss the absorption and emission of γ quanta in more detail.

3. Emission from nuclei in a gas

If a free emitting nucleus moves with a velocity $\mathbf{v}$ prior to the emission of a γ quantum, then the recoil energy turns out to be other than indicated above because the kinetic energy change in one reference frame differs from that in a frame of reference moving with a velocity $\mathbf{v}$ relative to the first, viz.

$$\Delta E_2 = \Delta E_1 + \Delta \mathbf{p}_1 \mathbf{v} \,, \tag{13}$$

where $\Delta \mathbf{p}_1$ is the momentum change in the first reference frame.

In an ensemble of chaotically moving, noninteracting with each other emitting nuclei (i.e. when the emitter is a classical ideal gas), the emitted γ quanta have a broad energy distribution with the first two moments given by

$$\langle E_\gamma \rangle_T = E_0 - R + \left\langle E_0 \frac{\mathbf{vp}}{c} \right\rangle_T = E_0 - R \,, \tag{14}$$

$$\langle E_\gamma^2 \rangle_T = \left\langle \left[E_0 - R + E_0 \frac{\mathbf{v}\hat{\mathbf{p}}}{c} \right]^2 \right\rangle_T \simeq \langle E_\gamma \rangle_T^2 \left(1 + \frac{\langle v^2 \rangle_T}{c^2} \right). \tag{15}$$

The symbol $\langle \ldots \rangle_T$ in the above equations denotes the averaging over the nuclear velocity distribution in the emitting gas at temperature T, and $\hat{\mathbf{p}} = \mathbf{p}_\gamma / |\mathbf{p}_\gamma|$. From Eqns (14) and (15) it follows that

$$\langle E_\gamma^2 \rangle_T - \langle E_\gamma \rangle_T^2 = \langle E_\gamma^2 \rangle_T \frac{\langle v^2 \rangle_T}{c^2} \,. \tag{16}$$

The spectrum of emitted γ quanta centers at the energy value $E_0 - R$, the width of the distribution being determined by the mean square of the velocity the nucleus had prior to emission.

An explicit expression for this distribution is obtained easily by noting that the velocity distribution of ideal gas

atoms has a Maxwellian form

$$P(E_\gamma)\,\mathrm{d}E_\gamma = \frac{1}{\sqrt{\pi}}\exp\left[-\frac{E_\gamma - E_0 \pm R}{\Delta^2}\right]\frac{\mathrm{d}E_\gamma}{\Delta}\,, \tag{17}$$

where $\Delta \simeq 2\sqrt{Rk_\mathrm{B}T}$ (for $R \ll E_\gamma$) is the Doppler width of the emission (+) or absorption (−) line.

We have, in Eqn (17), made use of the fact that

$$\overline{v_x^2} = \frac{2}{M}\,\overline{\mathcal{E}}_x = \frac{2}{M}\,\frac{kT}{2} = \frac{kT}{M}\,, \tag{18}$$

where $\overline{\mathcal{E}}_x$ is the average kinetic energy per degree of freedom of an atom moving in ideal gas (along the momentum of the emitted quantum).

Strictly speaking, the energy distribution curve exhibits this Gaussian form in its central portion provided $\Delta \gg \Gamma_0$. For $\Gamma \gg \Delta$, the Doppler broadening of line is of no significance. For arbitrary Γ/Δ, the distribution function for $|E_\gamma - (E_0 \pm R)| \gg \Gamma$ decreases according to the Breit–Wigner formula.

4. The Lipkin sum rule

Although the qualitative analysis of data on γ quanta emission from a moving nucleus (including the chaotic thermal motion of atoms in a gaseous phase) shows γ-quantum energies both greater than and smaller than $E_0 - R$ to be possible, the spectrum-averaged recoil energy of the nucleus is independent of the precise type of nuclear motion and is equal to R. This statement follows from a sum rule first obtained by Lipkin in 1960 [13].

Sum rules fix the value of the sum (integral) of the matrix elements for transitions between the states of a system under study, and take their origin in the probabilistic nature of quantum-mechanical predictions. The simplest and the most fundamental sum rule states that the total probability of finding the system in one of its possible state is unity.

What interests us here is to what extent the absorption or emission of a γ quantum changes the state of the crystal lattice.

The line shape for the absorption (emission) of a γ quantum with momentum $\mathbf{p}$ is given by

$$I(E_\gamma - E_R + E_{n_0}) = \sum_n F(E_\gamma - E_R + E_{n_0} - E_n) \times \left|\left\langle n\right|\exp\left(\frac{\mathrm{i}\mathbf{pR}}{\hbar}\right)\left|n_0\right\rangle\right|^2, \tag{19}$$

where $|n\rangle$ ($|n_0\rangle$) and E_n (E_{n_0}) are the wave function and energy defining the motion of the center of mass of the absorbing (emitting) nucleus in the final (initial) state, and $\mathbf{R}$ is the radius vector of the nuclear center of mass. The function $F(E_\gamma - E_R + E_{n_0} - E_n)$ is nothing but the resonance factor in the Breit–Wigner formula (1), and $|\langle n|\exp(\mathrm{i}\mathbf{pR}/\hbar)|n_0\rangle|^2$ determines the probability of the transition from the initial state $|n_0\rangle$ to one of the final states $|n\rangle$.

One important property of the function $I(E)$ can be obtained in a straightforward manner. Expanding the function $F(E_\gamma - E_R + E_{n_0} - E_n)$ formally in a power series in $(E_n - E_{n_0})$ (the energy transfer involved in the transition $n \to n_0$), we obtain

$$I(E_\gamma - E_R + E_{n_0}) \simeq F(E_\gamma - E_R) + F'(E_\gamma - E_R) \times \sum_n (E_n - E_{n_0})\left|\left\langle n\right|\exp\left(\frac{\mathrm{i}\mathbf{pR}}{\hbar}\right)\left|n_0\right\rangle\right|^2 + \ldots\,, \tag{20}$$

where we have used the fact that since the system is closed the wave functions $|n\rangle$ must satisfy the condition

$$\sum_n \left|\left\langle n\right|\exp\left(\frac{\mathrm{i}\mathbf{pR}}{\hbar}\right)\left|n_0\right\rangle\right|^2 = 1\,. \tag{21}$$

This condition reflects the fact that the sum of the probabilities for the transition from the initial state to any other state is unity — a fact which, as indicated above, can also be considered as a sum rule.

Direct calculation shows that

$$\sum_n (E_n - E_{n_0})\left|\left\langle n\right|\exp\left(\frac{\mathrm{i}\mathbf{pR}}{\hbar}\right)\left|n_0\right\rangle\right|^2 = \frac{p^2}{2M}\,, \tag{22}$$

where M is the mass of the absorbing (emitting) nucleus. Thus, what the Lipkin sum rule states is that the average energy transferred to all the final states of the system during the emission of a γ quantum by a nucleus is exactly the recoil energy of the nucleus.

Based on this universal result, an important qualitative prediction can be made about when a transition without energy transfer to the crystal lattice may occur with substantial probability. If a system can make transitions with a large energy transfer (exceeding even the recoil energy of a free nucleus), i.e. if hard phonon excitations are important in the case of a nucleus in a crystal, then (i) nearly all terms in the sum (22) correspond to such transitions, (ii) virtually no long-wavelength phonons are excited in the system, and (iii) the probability for a zero-energy-change transition (or, formally, for the excitation of a $\lambda = \infty$ phonon) increases considerably. In other words, in a polyatomic lattice the probability of Mössbauer effect is generally enhanced.

Thus, in particular, the presence of optical modes can drastically change the temperature dependence of the Mössbauer effect in the crystal. The reason is that optical phonons start to be excited at higher temperatures than acoustic phonons, so if optical modes are actively involved in the vibrations of the emitting atom, the probability of a phononless γ emission event decreases with temperature much more slowly compared with the case of a monatomic lattice with the same Debye temperature.

5. A nucleus in a crystal lattice

If a γ emitting nucleus is incorporated into a molecule or a crystal, the laws of conservation of energy and momentum must be obeyed by the isolated system composed of the γ quantum plus the molecule (or crystal) containing the nucleus. In considering the system γ-quantum + the emitting nucleus, the laws of conservation of energy and momentum turn out 'to be separated', thus permitting γ emission, processes in which the momentum is transferred to the entire massive emitter, so that the recoil energy is virtually zero (i.e. leaves the radiation energy unchanged).

It should be noted that, as F L Shapiro [3] has shown, while a full description of the phenomenon requires a complete quantum-mechanical treatment, some characteristic features of nuclear γ resonance in condensed media can be vividly described in classical terms. Because of the thermal motion of the lattice atoms, the emitting nucleus is in continuous vibrational motion which, due to the Doppler effect, results in the γ emission being frequency modulated, with a large number of side lines appearing in its spectrum.

The higher the temperature, the larger the vibration amplitudes, the larger the degree of modulation, and the lower the intensity of the carrier frequency (unshifted line). In other words, the Mössbauer line is rather strong if, as will be shown below, the vibration amplitude of lattice atoms is small compared with the γ radiation wavelength.

Before discussing the probability of the Mössbauer effect, we first consider the characteristic time scales determining various aspects of the process. As pointed out by A B Migdal [14], the radiative lifetime is defined as the time interval $\tau_0 \sim \lambda_\gamma/c$, where λ_γ is the emission wavelength. For a γ quantum with energy $E_\gamma = 100$ MeV, one obtains

$$\lambda_\gamma = \frac{2\pi}{k_\gamma} = \frac{2\pi\hbar}{p_\gamma} = \frac{2\pi\hbar c}{p_\gamma c} = \frac{2\pi\hbar c}{E_\gamma} = 10^{-12}\ \text{cm}\,, \tag{23}$$

giving

$$\tau_0 = \frac{10^{-12}}{3\times 10^{10}} \simeq 0.3\times 10^{-22}\ \text{s}\,. \tag{24}$$

For $E_\gamma = 100$ keV, the wavelength is 10^{-9} cm and hence $\tau_0 \simeq 3\times 10^{-20}$ s. Strictly speaking, our estimate holds good for $\lambda > R_\text{n} \sim 10^{-12}$ cm, when $\tau_0 \sim R_\text{n}/c$. Our first example puts a limit on these two estimates. In practically interesting cases $E_\gamma = 10-100$ keV and $\tau_0 \sim (1.5-0.3)\times 10^{-19}$ s.

It is therefore reasonable to assume that the whole emission process proceeds in two stages: the emission proper, with a characteristic time τ_0, and the final stage, when slow (molecular and solid-state) degrees of freedom with characteristic times $\tau_1 \sim 10^{-13}$ s come into play. Thus, during the γ quantum emission process the emitting nucleus instantly receives a recoil momentum $\mathbf{p}_\gamma$, but it practically does not move from its original position for a time $\tau_0 \ll \tau_1$.

Since a γ quantum may be emitted at any time during the lifetime $\sim \hbar/\Gamma$ of the excited state of the nucleus, it is on the passage of this time interval that the process of emission should be considered completed. Only thereafter we can speak about the fixed energy of the emitted quantum and a certain excitation of the phonon subsystem of the crystal, and the final result therefore depends on the value of the parameter $\tau_1\Gamma/\hbar$. For $\tau_1\Gamma/\hbar \gg 1$, a nucleus in a crystal emits as a free particle, and for $\tau_1\Gamma/\hbar \ll 1$, the energy distribution of γ quanta is strongly affected by the way the nucleus is bound to the crystal (molecule).

Thus, in the first case the nucleus should be expected to emit as a free particle moving with a momentum $\mathbf{p}_0$, the probability of finding a particular value of $\mathbf{p}_0$ being determined by the nuclear momentum distribution in the crystal. In the second limiting case, in accord with the general rules of quantum mechanics, the excitation of the original motion of the nuclear center-of-mass breaks down into a set of independent proper motions of a harmonic crystal — normal modes of vibration with the excitation of various numbers of their quanta, i.e. phonons. At the semiclassical level this transformation may be interpreted as the emission of various numbers of phonons. We can analyze such a process based on probability-theoretic hypotheses that underlie the Poisson random process. The very application of a semiclassical treatment of this process relies on the properties of the motion of a harmonic oscillator which suddenly acquires a momentum $\mathbf{p}$; these properties admit such a nonquantum description.

Thus, the subsequent motion of a nucleus incorporating into a molecule or a crystal requires, strictly speaking, an exact quantum-mechanical treatment. To reduce the problem to its essentials, let us model the center-of-mass motion of an emitting nucleus by a one-dimensional harmonic oscillator (potential energy $U(x) = kx^2/2$) originally occupying the ground state $|0\rangle$.

Immediately after the emission of a γ quantum the center-of-mass wave function of the nucleus is of the form

$$|f\rangle = \exp\left(\mathrm{i}\frac{p_\gamma}{\hbar}x\right)|0\rangle\,. \tag{25}$$

Such a sudden shaking of the slow subsystem (the center-of-mass motion of the nucleus) enables the excitation of oscillator states with $n \geqslant 1$ [n being the quantum number of the excited state with energy $\mathcal{E}_n = \hbar\omega(n+1/2)$, where ω is the oscillator frequency]. Along with this, there is a probability that the oscillator will remain in the unexcited state $|0\rangle$. The wave function $|f\rangle$ is a linear combination of the oscillator eigenstates, viz.

$$|f\rangle = \sum_{n=0}^{\infty} C_n|n\rangle\,. \tag{26}$$

According to the laws of quantum mechanics, the probability $\widetilde{W}_n$ of the system being excited to the state $|n\rangle$ is determined by $|C_n|^2$. Since the acts of excitation of a particular state $|n_0\rangle$ are independent of the excitation of other states, probability-theoretic arguments show that $\widetilde{W}_n$ obeys the Poisson distribution

$$\widetilde{W}_f \equiv \widetilde{W}_n = \frac{\langle n\rangle^n}{n!}\exp\left(-\langle n\rangle\right), \qquad \sum_{n=0}^{\infty}\widetilde{W}_n = 1\,, \tag{27}$$

where angle brackets $\langle\ldots\rangle$ denote the averaging over the distribution $\widetilde{W}_n$.

This form of $\widetilde{W}_n$ is confirmed by a rigorous quantum-mechanical calculation. From the foregoing arguments, the expression for $\langle n\rangle$ follows clearly as

$$\langle n\rangle = \frac{R}{\hbar\omega} = \frac{E_\gamma^2}{2Mc^2\hbar\omega}\,. \tag{28}$$

The quantity we are concerned with — the probability of the oscillator remaining in its original state $|0\rangle$ despite the momentum transfer p_γ — is given by

$$\widetilde{W}_0 = \exp\left(-\langle n\rangle\right). \tag{29}$$

This indicates the probability of detecting an emission line with a natural width Γ, i.e.

$$P(E_0) \propto \frac{\widetilde{W}_0}{(E_\gamma - E_0)^2 + \Gamma^2/4}\,. \tag{30}$$

Similarly, with probability $\widetilde{W}_1 = \langle n\rangle\exp(-\langle n\rangle)$ there appears an emission line corresponding to the excitation of a single phonon, of the form

$$\frac{1}{(E_\gamma - E_0 + \hbar\omega)^2 + \Gamma^2/4}\,, \tag{31}$$

and so forth.

This result (we limit the discussion to an unshifted emission line) can be extended in a straightforward manner

to the case of a harmonic crystal involving N atoms. The probability $\widetilde{W}_f$ is then the product of $S = 3N - 6 \simeq 3N$ probabilities $\widetilde{W}_{n_i}$ for each of the independent normal modes of vibration, viz.

$$\widetilde{W}_f = \prod_{i=1}^{S} \widetilde{W}_{n_i} . \tag{32}$$

This model of localized crystal vibrations is known as the Einstein model. In a more realistic approach the dynamics of a crystal are modelled by a system of independent, travelling collective motions (phonons). In this case the structure of $\widetilde{W}_f$ remains the same, because the individual modes of the collective motion are independent, and n_i must be understood as the number of excited phonons, i.e. of the quanta of the ith vibrational mode.

The probability of a recoil-free transition is defined as

$$\widetilde{W}_0 = \prod_{i=1}^{S} \exp\left[-\frac{(\mathbf{p}_\gamma \mathbf{e}_i)^2}{2M\hbar\omega_i} \right] = \exp\left[-\sum_{i=1}^{S} \frac{(\mathbf{p}_\gamma \mathbf{e}_i)^2}{2M\hbar\omega_i} \right]$$

$$= \exp[-2W_0] . \tag{33}$$

Here ω_i and $\mathbf{e}_i$ are the frequency and the polarization vector of the ith normal mode in a crystal.

The quantity $\exp[-2W_0]$ is the so-called Debye – Waller factor familiar from the theory of scattering of X-rays and thermal neutrons in crystals (for the crystal target assumed to be at the absolute zero of temperature, $T = 0$). For the case of a simple cubic lattice relation (28) derived above becomes

$$\sum_i \frac{(\mathbf{p}_\gamma \mathbf{e}_i)^2}{2M\hbar\omega_i} = \frac{p^2}{2M} \frac{1}{3N} \sum_{i=1}^{3N} \frac{1}{\hbar\omega_i} = \frac{p^2}{2M} \left\langle \frac{1}{\hbar\omega} \right\rangle . \tag{34}$$

Here $\langle \ldots \rangle$ denotes the averaging over the frequency spectrum of the normal modes of the crystal, namely, one finds

$$\left\langle \frac{1}{\hbar\omega} \right\rangle = \int_0^{\omega_{\max}} \frac{g(\omega)}{\hbar\omega} \, \mathrm{d}\omega , \quad \int_0^{\omega_{\max}} g(\omega) \, \mathrm{d}\omega = 1 . \tag{35}$$

The number of vibrational modes in the interval $(\omega + \mathrm{d}\omega, \omega)$ is $3Ng(\omega) \, \mathrm{d}\omega$.

In the case of the Debye model of a crystal, we have

$$g(\omega) \, \mathrm{d}\omega = \begin{cases} \dfrac{3\omega^2}{\omega_{\max}^3} \, \mathrm{d}\omega & \text{for} \quad \omega \leqslant \omega_{\max} = \omega_{\mathrm{D}} , \\ 0 & \text{for} \quad \omega \geqslant \omega_{\max} , \end{cases} \tag{36}$$

where ω_{D} is the Debye frequency, and the Debye temperature is given by $\Theta = \hbar\omega_{\max}/k$.

Consequently, in this model one obtains

$$\left\langle \frac{1}{\hbar\omega} \right\rangle = \frac{3}{\omega_{\max}^3} \int_0^{\omega_{\max}} \frac{\omega' \, \mathrm{d}\omega}{\hbar\omega} = \frac{3}{2} \frac{\omega_0^2}{\omega_{\mathrm{D}}^3} = \frac{3}{2\hbar\omega_{\mathrm{D}}} , \tag{37}$$

$$\langle n \rangle = \frac{E_\gamma^2}{2Mc^2} \left\langle \frac{1}{\hbar\omega} \right\rangle = \frac{3E_\gamma^2}{4Mc^2} \frac{1}{\hbar\omega_{\mathrm{D}}} = \frac{3}{4} \frac{E_\gamma^2}{Mc^2} \frac{1}{k\Theta} . \tag{38}$$

The probability of finding an unshifted line is larger, the higher the Debye temperature of the crystal and the lower the energy of the nuclear γ transition (in accord with the Lipkin sum rule).

The Debye – Waller factor is thus

$$\exp[-2W_0] = \exp\left[-\langle n \rangle \right] = \exp\left[-R \left\langle \frac{1}{\hbar\omega} \right\rangle \right] . \tag{39}$$

We can put the Debye – Waller factor into a form allowing a different interpretation qualitatively. From the virial theorem we have

$$\frac{1}{2} M\omega^2 \langle x^2 \rangle = \frac{\langle p^2 \rangle}{2M} = \frac{E_{\mathrm{tot}}}{2} = \frac{\hbar\omega}{4} , \qquad T = 0 . \tag{40}$$

From this it follows

$$\langle x^2 \rangle = \frac{\hbar}{2M\omega} , \qquad \frac{1}{\hbar\omega} = \langle x^2 \rangle \frac{2M}{\hbar^2} , \tag{41}$$

$$\langle p^2 \rangle = \frac{M\hbar\omega}{2} , \qquad \frac{1}{\hbar\omega} = \frac{1}{\langle p^2 \rangle} \frac{M}{2} . \tag{42}$$

It is readily seen that

$$\langle x^2 \rangle \langle p^2 \rangle = \frac{\hbar^2}{4} \tag{43}$$

in response to the uncertainty relation.

Using the above relations, the Debye – Waller factor can be put in the form

$$\exp[-2W_0] = \exp\left[-\frac{R}{\hbar\omega} \right] = \exp\left[-R \frac{2M}{\hbar^2} \langle x^2 \rangle \right]$$

$$= \exp\left[-\frac{E_\gamma^2}{\hbar^2 c^2} \langle x^2 \rangle \right] = \exp\left[-k_\gamma^2 \langle x^2 \rangle \right]$$

$$= \exp\left[-\frac{4\pi^2 \langle x^2 \rangle}{\lambda_\gamma^2} \right] . \tag{44}$$

Written in this way, the Debye – Waller factor shows that waves generated at various points in the crystal add up with random phase factors $\exp[\mathrm{i}\varphi(\mathbf{R}_0, \mathbf{x})] = \exp(\mathrm{i}k_\gamma x)$ at the point of observation, $\mathbf{R}_0$. If the variance of $\varphi(\mathbf{R}_0, \mathbf{x})$ is weak and if various displacements are independent of each other, then averaging over the Gaussian distribution yields

$$\left\langle \exp\left[\mathrm{i}\varphi(R_0, x) \right] \right\rangle = \exp\left(-\frac{4\pi^2 \langle x^2 \rangle}{\lambda^2} \right) . \tag{45}$$

The decoherence of the added waves can be neglected if $\sqrt{\langle x^2 \rangle} \ll \lambda/2\pi$.

A totally different physical interpretation complementary to the previous one can be obtained by writing

$$\exp(-2W_0) = \exp\left(-\frac{\hbar^2 k_\gamma^2}{4 \langle p^2 \rangle} \right) . \tag{46}$$

The radiation stays coherent if the recoil momentum is small compared to the momentum spread of the vibrating nucleus, $\sqrt{\langle p^2 \rangle}$. The transition from the wave to the corpuscular picture of the process occurs in accord with the uncertainty relation (43).

6. Nonzero temperatures

Arguments based on the assumption of phonons being excited independently during the emission of a γ quantum are not valid if the emitter is found itself at nonzero

temperature. The quanta of harmonic vibrational modes — phonons in the case of a crystal — are quasi-particles obeying Bose – Einstein statistics and they cannot be treated as independent because of there being specific statistical correlations between them (induced emission of phonons). In the case of a crystal emitter, however, the expression for the Debye – Waller thermal factor valid for any temperature (see above) can be employed and one arrives at

$$\exp(-2W_f)_T = \exp\left(-\frac{4\pi^2\langle u^2\rangle_T}{\lambda^2}\right). \tag{47}$$

Here $\langle u^2\rangle_T$ is the mean square of the displacement of the emitting nucleus in the harmonic crystal. Using now the relation between $\langle u^2\rangle_T$ and the total energy, i.e.

$$\frac{M\omega^2\langle u^2\rangle_T}{2} = \hbar\omega\left[\langle n_\omega\rangle_T + \frac{1}{2}\right], \quad \langle n_\omega\rangle_T = \frac{1}{\exp(\hbar\omega/kT)-1}, \tag{48}$$

and again limiting ourselves to the one-degree-of-freedom case, we have

$$\langle u^2\rangle_T = \frac{2\hbar}{M\omega}\left[\langle n_\omega\rangle_T + \frac{1}{2}\right]. \tag{49}$$

For a monatomic cubic lattice, one finds

$$\begin{aligned} 2W_T &= \frac{\hbar k_\gamma^2}{2M}\,\frac{1}{3N}\sum_{s=1}^{3N}\frac{2\langle n_s\rangle_T+1}{\omega_s} \\ &= \frac{\hbar k_\gamma^2}{2M}\int_0^{\omega_{\max}}\frac{2\langle n_s\rangle_T+1}{\omega}\,g(\omega)\,\mathrm{d}\omega \\ &= \frac{R}{\hbar}\int_0^{\omega_{\max}}\left[\frac{2}{\exp(\hbar\omega/kT)-1}+1\right]\frac{g(\omega)\,\mathrm{d}\omega}{\omega}, \end{aligned} \tag{50}$$

which in the Debye approximation becomes

$$2W_T = \frac{6R}{k\Theta}\left[\frac{1}{4}+\frac{T}{\Theta}\Phi\left(\frac{\Theta}{T}\right)\right], \quad \Phi(x) = \frac{1}{x}\int_0^x\frac{t\,\mathrm{d}t}{\exp t - 1}. \tag{51}$$

In the limiting cases of temperatures T far above and far below the Debye temperature, we have

$$2W_T \simeq \frac{3R}{2k\Theta}\left[1+\frac{2\pi^2}{3}\left(\frac{T}{\Theta}\right)^2\right], \qquad T \ll \Theta, \tag{52}$$

and

$$2W_T \simeq \frac{6R}{k\Theta}\left(\frac{T}{\Theta}\right)^2, \qquad T \gg \Theta, \tag{53}$$

respectively.

In the case of a small number of vibrational degrees of freedom, corrections are needed to the above results because our analysis ignored certain correlations between the quanta of one and the same vibrational mode. In large systems such as a crystal ($3N \gg 1$), the ignored corrections are small. In one dimension, however, an exact quantum-mechanical calculation shows that the thermal exponential should be multiplied by an additional factor, giving

$$\widetilde{W}_0 = \exp(-2W_T) I_0\left(2W_T\ \mathrm{sh}^{-1}\frac{\hbar\omega}{2kT}\right). \tag{54}$$

Here $I_0(x)$ is a zero-order Bessel function in the imaginary argument. For $T \to 0$, it is known that $I_0\big(2W_T\ \mathrm{sh}^{-1}(\hbar\omega/2kT)\big) \to I_0(0) = 1$, and we retrieve the result $\exp(-2W_0)$ obtained above. If the number of normal vibrational modes of the system is large, we have

$$\widetilde{W}_0 = \exp(-2W_T)\prod_{s=1}^{3N} I_0\left(2W_{Ts}\ \mathrm{sh}^{-1}\frac{\hbar\omega_s}{2kT}\right), \tag{55}$$

where $2W_{T_s} \sim 2W/N$ is the amount each of the modes contributes to $2W_T$.

The above correction to the previous result $\widetilde{W}_0 = \exp(-2W_T)$ is significant when, along with the large number of vibrational degrees of freedom which are generally on the same footing with one another, there is a small group which is special in some respect. Such a situation occurs when the emitter is placed within a crystal of dissimilar atoms and therefore represents an impurity.

7. Excitation of a large number of phonons

We now turn to that portion of the emission line that governs the excitation (absorption) of a large number of phonons. For $T = 0$, only transitions that excite quanta of elastic vibrations are possible. We take as our starting point the Poisson distribution

$$\widetilde{W}_n = \frac{\langle n\rangle^n}{n!}\exp(-\langle n\rangle). \tag{56}$$

If $n = \langle n\rangle(1+\varepsilon)$, then for $\langle n\rangle \gg 1$ and $\varepsilon \ll 1$, the Poisson distribution reduces to the Gaussian one

$$\widetilde{W}_n \simeq \frac{1}{\sqrt{2\pi\langle n\rangle}}\exp\left[-\frac{(n-\langle n\rangle)^2}{2\langle n\rangle}\right], \quad \left\langle (n-\langle n\rangle)^2\right\rangle = \langle n\rangle,$$

$$\sigma = \sqrt{\left\langle (n-\langle n\rangle)^2\right\rangle} = \sqrt{\langle n\rangle}. \tag{57}$$

For large n ($n \gg 1$) and small deviations from $\langle n\rangle$ [$n = \langle n\rangle(1+\varepsilon)$, $\varepsilon \ll 1$] we shall have a Gaussian instead of a Poisson distribution.

Thus, for $T = 0$, in the case of a harmonic oscillator, $\langle n\rangle = 2W_0 = R/\hbar\omega$, we have

$$P(E_\gamma)\,\mathrm{d}E_\gamma = \frac{1}{\sqrt{2\pi R\hbar\omega}}\exp\left[-\frac{(E_\gamma - E_0 \pm R)^2}{2R\hbar\omega}\right]\mathrm{d}E_\gamma. \tag{58}$$

It is interesting to compare this expression with the distribution of E_γ in the case of an emitting nucleus in a gas at temperature T:

$$P(E_\gamma)\,\mathrm{d}E_\gamma = \frac{1}{\sqrt{\pi}}\frac{\mathrm{d}E_\gamma}{\Delta}\exp\left[-\frac{(E_\gamma - E_0 \pm R)^2}{\Delta^2}\right],$$

$$\Delta = 2\sqrt{RkT}. \tag{59}$$

The squares of the widths of these distributions are

$$\Delta^2_{\mathrm{osc}} = 2R\hbar\omega = 4R\mathcal{E}_0 = 8R\langle\mathcal{E}_{\mathrm{к}}\rangle_{T=0}, \quad T = 0,$$

$$\Delta^2_{\mathrm{gas}} = 4RkT = 8R\langle\mathcal{E}_{\mathrm{к}}\rangle. \tag{60}$$

The interpolation formula linking these two extremes is obtained by rewriting the above relations in the form

$$\Delta_{\rm osc}^2 = 8R\langle\mathcal{E}_{\rm к}\rangle_T = 4R\langle\mathcal{E}_{\rm tot}\rangle_T . \tag{61}$$

In the Debye model, one has

$$\langle\mathcal{E}_{\rm tot}\rangle_T = 3kT\left(\frac{T}{\Theta}\right)^3 \int_0^{\Theta/T} \left(\frac{1}{\exp t - 1} + \frac{1}{2}\right) t^3 \, {\rm d}t . \tag{62}$$

For an arbitrary frequency spectrum of the normal vibrational modes of the crystal, we find

$$\langle\mathcal{E}_{\rm tot}\rangle_T = \int_0^{\omega_{\max}} g(\omega)\hbar\omega \left[\langle n_\omega\rangle + \frac{1}{2}\right] {\rm d}\omega . \tag{63}$$

Thus, for arbitrary temperature, the width of the energy distribution of γ radiation accompanied by the emission (and absorption) of a large number of phonons is given by

$$\Delta^2 = 4R\langle\mathcal{E}_{\rm tot}\rangle_T . \tag{64}$$

In the high-temperature range, the center-of-mass motion of the emitter can be treated classically. Then only the velocity distribution is of importance and indeed we arrive at

$$\Delta^2 = 8R\langle\mathcal{E}_{\rm к}\rangle = 4R\langle\mathcal{E}_{\rm tot}\rangle , \qquad T \gg \Theta . \tag{65}$$

At the same time, it follows that

$$\Delta^2 = 4R\mathcal{E}_{\rm tot} = 2R\hbar\omega , \qquad T = 0 . \tag{66}$$

8. Conclusions

Admittedly, many undeniably important topics related to the Mössbauer effect remained untreated in this paper. Among them are the coherent effects which emerge due to the fact that waves corresponding to γ quanta coherently scattered from two scatterers interfere with each other; if resonantly scattering nuclei are introduced in a regular manner into a crystal lattice, resonant nuclear diffraction of γ quanta is possible — and indeed is examined experimentally. Mention should also be made of the suppression of inelastic nuclear reaction channels at high concentrations of resonantly scattering nuclei — an effect theoretically predicted by Yu M Kagan and A M Afanas'ev [15] and then confirmed repeatedly by experiment.

In the present paper we have not even touched on developments in experimental methodology (practical aspects of experiment are reviewed in detail in Ref. [16], and methods for analyzing Mössbauer spectra are discussed in Ref. [17]). Most of the studies related to the use of the Mössbauer effect have and are being conducted in the transmission geometry because of its simplicity, the large magnitude of the effect, and the high counting rate. It should be noted, however, that also experiments on the detection of scattered radiation, secondary X-rays, and conversion electrons are being conducted. In conversion Mössbauer spectroscopy, the resonant absorption of γ quanta is detected by the change in the intensity of the conversion electrons escaping the sample. Emission spectroscopy [18] explores materials containing implanted radioactive nuclei which — as a result of nuclear transformations and a subsequent γ transition cascade — form excited nuclei that emit resonant γ quanta.

As already mentioned, the application spectrum of the Mössbauer effect covers many other fields, including chemistry [19], biology [20], magnetism [21], surface physics [22], the physics of metals, archeology, geology, and medicine. An indisputable advantage of Mössbauer spectroscopy is that it takes only one experiment to determine the probability of the effect, the temperature and chemical shifts, quadrupole and magnetic splittings, and the line shapes of individual components. In addition to that, Mössbauer spectra lend themselves to being affected by a variety of external factors such as temperature, pressure, electric and magnetic fields, ultrasound, etc. The development of the γ laser using the Mössbauer effect is another promising line of research in this field [23].

Much attention is currently being given to the use of synchrotron radiation in γ-resonance spectroscopy [24, 25].

Among recent theoretical studies, those of Refs [26–28] deserve special mention. Lomonosov and Sazonov [26] developed a time-dependent theory of resonant fluorescence and analyzed the wave packets of scattered particles by considering their dynamics of formation and propagation in space. Hoy [27], in his treatment of a one-dimensional quantum-mechanical model of nuclear resonance scattering of γ radiation, takes into account possible resonant re-emissions, which is especially important for the accurate time analysis of spectroscopic data. Of particular interest is the work of Koncharovskaya et al. [28], who emphasize the possibility of using a coherent optical laser field to control a nuclear–electron system. It is shown that the emission (absorption) spectra of isomeric nuclear levels can be changed significantly by exploiting the coupling between the electronic and nuclear degrees of freedom.

Two final remarks are in order. First, although the discussion in this paper has focused on the Mössbauer effect, practically all the conclusions hold for electron or neutron scattering in a lattice — processes which also involve the excitation of the phonon subsystem. Second, an unshifted line can be observed not only in crystals but also in large biological molecules or complex chemical compounds. Separate fragments of such molecules may be in more than one stable equilibrium state, among which random transitions may occur at sufficiently high temperatures. Because the motion of such a fragment is confined spatially, the emission and absorption spectra of their constituent nuclei contain an unshifted line of natural width against the background of an additional Lorentzian line due to diffusion of fragments [29, 30].

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