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Line broadening and line shift of long-lived states of nuclei in a lattice

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Abstract. Long-lived states of nuclei embedded in a lattice are continuously perturbed by the surrounding atoms. The perturbations can be considered as stochastic processes described by a stochastic variable. The first-order correlation function, from which the frequency spectrum radiated by the nuclei can be derived, depends, among other things, on the ensemble average of a particular function of the stochastic variable. This function depends on the actual perturbation mechanisms. Making use of the ergodic theorem allows for the calculation of this ensemble average and, hence, of the radiated frequency spectrum. The spectrum is shown to be Lorentzian and homogeneously broadened. Also a frequency shift with respect to the unperturbed frequency has been found. An order of magnitude of the broadening and shift is given.

1. Introduction

Recently [1–3] we have presented two different descriptions of the behaviour of long-lived nuclei embedded in a solid-state lattice.

In [1] and [2] the continuous modification of the wave trains, emitted by the nuclei in long-lived nuclear states, due to the interactions of the nuclei with the crystalline lattice in which they are incorporated, has been analysed. For long-lived nuclei the stochastic interactions between the lattice and the nuclei are very brief (compared to the nuclear lifetime) uncorrelated Markovian processes. It can be shown that the frequency spectrum radiated by an ensemble of the nuclei is Lorentzian with a full width at half maximum equal to the sum of the natural width (coming from radioactive decay) and a lattice induced width. The line broadening has been shown to be homogeneous for long-lived states.

In [3] a completely quantum mechanical approach is introduced, based on the study of the behaviour of the nuclei themselves, leading also to a homogeneously broadened radiation spectrum for long-lived nuclear states.

The purpose of these studies is, amongst other things, to explain why a positive Mössbauer effect could be observed making use of the 88 keV transition of ^{109}Ag. The natural line width of the long-lived isomeric state, giving rise to the 88 keV Mössbauer transition in ^{109}Ag, is about 10^{-17} eV. Without any homogeneous line broadening it would thus be impossible to observe a Mössbauer effect using very long-lived states. Several groups [4–6] claimed evidence for a small effect in ^{109}Ag.

The first idea of invoking homogeneous line broadening is described qualitatively by Coussement *et al* [7].

In the present article we will present another approach that will lead not only to line broadening but also to a shift in the frequency distribution of the emitted radiation. In

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section 2 the frequency spectrum will be defined. In section 3 the model leading to the line broadening and the shift will be presented. A numerical estimation of the line broadening and shift will be given.

2. Definition of the frequency spectrum

The frequency spectrum $F(\omega)$ of a classical radiated electromagnetic field can be written [8, 9] as

$$F(\omega) = \frac{1}{2\pi}\int_{-\infty}^{+\infty} g^{(1)}(\tau)\,\mathrm{e}^{\mathrm{i}\omega\tau}\,\mathrm{d}\tau = \frac{1}{\pi}\mathrm{Re}\int_{0}^{+\infty} g^{(1)}(\tau)\,\mathrm{e}^{\mathrm{i}\omega\tau}\,\mathrm{d}\tau \tag{1}$$

where the first order correlation function $g^{(1)}(\tau)$ is defined by

$$g^{(1)}(\tau) = \frac{\langle E^*(\boldsymbol{r},\, t)E(\boldsymbol{r},\, t+\tau)\rangle}{\langle E^*(\boldsymbol{r},\, t)E(\boldsymbol{r},\, \tau)\rangle}. \tag{2}$$

$E(t)$ is a component of a classical electric (or magnetic) field. $\langle\rangle$ is the statistical average over all possible field values.

The quantum mechanical definition of the spectrum [8–10], although less obvious than the classical one, is defined in an analogous way with $g^{(1)}(\tau)$ now given by

$$g^{(1)}(\tau) = \frac{\langle E^-(\boldsymbol{r},\, t)E^+(\boldsymbol{r},\, t+\tau)\rangle}{\langle E^-(\boldsymbol{r},\, \tau)E^+(\boldsymbol{r},\, \tau)\rangle} \tag{3}$$

with E^- the quantized electric (or magnetic) field component operator containing the photon creation operators and E^+ the operator containing the photon annihilation operators. The expression $\langle O\rangle$ represents the average value of the operator O. The numerator is a two-time average, which presents a formal difficulty that can be circumvented with the aid of the quantum regression theorem [11]. The denominator of (3) is a one-time average.

The study of the radiated spectrum can be performed classically as well as quantum mechanically. The latter treatment is however much more involved, as shown in [3].

The analysis given in the next section can be applied to both the classical and the quantum mechanical case.

3. Homogeneous line broadening and shift of the frequency spectrum radiated by long-lived nuclei in a lattice

3.1. Stochastic processes in a lattice

In the following nuclei having a long-lived isomeric state and a stable ground state are considered. The unperturbed nuclear Bohr frequency corresponding to these two levels will be denoted ω_n.

When the nuclei are incorporated in a crystalline lattice, their interactions with the atoms surrounding them perturb (continuously or not) the nuclear energy levels. This means that the nuclear Bohr frequency is shifted from the unperturbed value ω_n. The instantaneous Bohr frequency can be written as

$$\omega(t) = \omega_n - x(t). \tag{4}$$

$x(t)$ is the value at time t of a stochastic variable X.

The important expression to be used in both the classical and the quantum mechanical definition of the radiated spectrum is [1–3]

$$f(\tau) = \left\langle \exp\left(\mathrm{i} \int_t^{t+\tau} x(t')\, \mathrm{d}t' \right) \right\rangle. \tag{5}$$

The first-order correlation function from which the spectrum is directly derived according to (1) can be shown [1–3] for both cases to be

$$g^{(1)}(\tau) = \mathrm{e}^{-\mathrm{i}\omega_n \tau} \mathrm{e}^{-(\frac{\gamma}{2})\tau} f(\tau) \tag{6}$$

where γ describes the natural nuclear decay.

The stochastic average $\langle\rangle$ depends on the probability density function governing the stochastic variable X, thus on the nature of the interactions between the radiating nuclei and their surroundings in the lattice.

We will dwell further on the stochastic variable X. The perturbers—the neighbouring atoms—produce at the position of the nucleus a randomly fluctuating microfield. So we have a stochastic process, i.e., a randomly fluctuating function of time $X(t)$, defined by the possible values that the stochastic variable X may take at any instant. These values constitute the sample space [12] of the stochastic variable X. The possible values x that the variable X may take are determined by the probability with which the value x occurs. The stochastic variable X (which is in our case simply related to the energy of the nucleus in the lattice) is subject to uncontrollable actions that render precise predictions impossible.

A process is stationary if all statistical properties are unchanged when all time arguments are shifted by the same amount. In particular one has

$$f(t+\tau) = f(t). \tag{7}$$

In the following we will suppose that the properties of the emitting source (the excited nuclei in the lattice) are stationary, which means that the influence governing the statistical fluctuations does not depend on the starting time of the observations. This implies, among other things, that the observation period must be long compared to the time scale of the fluctuations. For long-lived nuclei this condition is always fulfilled.

A stationary process is ergodic if all statistical properties can be deduced from a single realization of infinite duration [12]. Then the ensemble average $\langle\rangle$ is equal to a time average. For long-lived nuclei interacting with their neighbouring atoms in the lattice in which they are embedded, the fluctuations on the nuclei can be described as an ergodic process.

3.2. Calculation of the first-order correlation function making use of the ergodic theorem. Radiated spectrum

As already has been mentioned the key element for the calculation of the frequency spectrum of the radiation emitted by nuclei incorporated in a lattice is expression (5).

If we define

$$\phi(t) = \int_0^t x(t')\, \mathrm{d}t' \tag{8}$$

it can be shown then that

$$f(\tau) = \left\langle \exp \mathrm{i}\left[\phi(t+\tau) - \phi(t)\right]\right\rangle. \tag{9}$$

If the system is ergodic, the ensemble average can be replaced by a time average. During an interval $\mathrm{d}\tau$ the change of $f(\tau)$ is

$$\begin{aligned} \mathrm{d}f(\tau) &= \left\langle \exp \mathrm{i}\left[\phi(t+\tau+\mathrm{d}\tau) - \phi(t)\right]\right\rangle - \left\langle \exp \mathrm{i}\left[\phi(t+\tau) - \phi(t)\right]\right\rangle \\ &= \left\langle \exp \mathrm{i}\left[\phi(t+\tau) - \phi(t)\right]\left(\mathrm{e}^{\mathrm{i}\alpha} - 1\right)\right\rangle \end{aligned} \tag{10}$$

where α represents the additional phase shift during $\mathrm{d}\tau$.

If during $\mathrm{d}\tau$ there has not been any interaction between the nucleus and the lattice, α is evidently zero. If an interaction takes place during $\mathrm{d}\tau$, then the phase is changed in a random way. So the model consists in assuming that the phase at $t+\tau$ is uncorrelated to the phase $\mathrm{d}\tau$ later if an interaction occurs during $\mathrm{d}\tau$ and that a finite phase jump occurs during $\mathrm{d}\tau$.

This means that the perturbation on the nuclei due to the lattice is described by a Dirac delta-like correlation function, such as the one introduced in [1–3]. This implies that the average of the product is equal to the product of averages in (10)

$$\mathrm{d}f(\tau) = f(\tau)\langle \mathrm{e}^{\mathrm{i}\alpha} - 1\rangle. \tag{11}$$

Again making use of the ergodic theorem, the time average can be replaced by an ensemble average (which is over all possible interactions). The value of $\langle \mathrm{e}^{\mathrm{i}\alpha} - 1\rangle$ depends on the nature of the interactions between the nucleus and the lattice. It depends also on the geometry of the surroundings of the radiating nuclei. The ensemble average (over all possible interactions during $\mathrm{d}\tau$) is proportional to $\mathrm{d}\tau$. If the centre of the nucleus is taken as the origin O of a coordinate system, the 'number' of nucleus–lattice interactions during $\mathrm{d}\tau$, due to what happens in the infinitesimal volume $\mathrm{d}^3\boldsymbol{r}$ around the point given by $\boldsymbol{r}$, can be written as $\mathrm{d}\tau\, h(\boldsymbol{r})\,\mathrm{d}^3\boldsymbol{r}$ with $h(\boldsymbol{r})$ a function depending on the nature of the interactions and on the geometry. Because of the fluctuations corresponding to the volume $\mathrm{d}^3\boldsymbol{r}$, there will be a phase shift, which will now be a function of $\boldsymbol{r}$, $\phi(\boldsymbol{r})$. In order to understand better the significance of $\phi(\boldsymbol{r})$, one could assume a potential V describing the interactions of the nucleus with the lattice. Let us suppose first that the radiating nucleus interacts with only one neighbouring atom situated at a distance $\boldsymbol{r}'$ from it. $\boldsymbol{r}'$ will thus depend on time, according to the hypothesis of fluctuating interactions. The potential $V(\boldsymbol{r}')$ will thus also depend on time. The frequency shift $\Delta\omega$ (with respect to the unperturbed frequency ω_n) in the nuclear transition corresponding to the potential $V(\boldsymbol{r}')$ at point $\boldsymbol{r}'$ is then

$$\hbar\Delta\omega(\boldsymbol{r}') = V(\boldsymbol{r}'). \tag{12}$$

Let us define another coordinate system with origin O′, situated at a fixed point $\boldsymbol{r}$ with respect to O (figure 1). Since $\boldsymbol{r}'(t)$ depends on time one has

$$\boldsymbol{r}'(t) = \boldsymbol{r} + \boldsymbol{a}(t) \tag{13}$$

with $\boldsymbol{a}(t)$ a vector depending on time.

The phase shift corresponding to all interactions during the time interval $(-\infty, +\infty)$ is

$$\phi(\boldsymbol{r}) = \int_{-\infty}^{+\infty} \Delta\omega\,\mathrm{d}t = \frac{1}{\hbar}\int V(\boldsymbol{r}+\boldsymbol{a}(t))\,\mathrm{d}t \tag{14}$$

where (12) and (13) have been used. After integration (14) is of course a function of $\boldsymbol{r}$, $\phi(\boldsymbol{r})$. If the interaction between the radiating nucleus and the lattice is due to many atoms acting simultaneously, the same reasoning as before can be applied.

Even if it is not possible to describe the nucleus–lattice interactions in terms of a potential, it is still possible to define a phase shift corresponding to $\boldsymbol{r}$. So the ensemble average of $\langle \mathrm{e}^{\mathrm{i}\alpha} - 1\rangle$ becomes then

$$\langle \mathrm{e}^{\mathrm{i}\alpha} - 1\rangle = \mathrm{d}\tau \int\!\!\int\!\!\int h(\boldsymbol{r})[\cos\phi(\boldsymbol{r}) - 1]\,\mathrm{d}^3\boldsymbol{r} + \mathrm{i}\,\mathrm{d}\tau \int\!\!\int\!\!\int h(\boldsymbol{r})\sin\phi(\boldsymbol{r})\,\mathrm{d}^3\boldsymbol{r}. \tag{15}$$

Calling

$$\frac{\gamma_b}{2} = \int\!\!\int\!\!\int h(\boldsymbol{r})[1 - \cos\phi(\boldsymbol{r})]\,\mathrm{d}^3\boldsymbol{r} \tag{16}$$

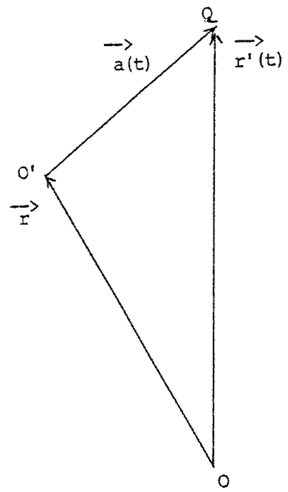

Figure 1. O is the centre of the radiating nucleus, Q is the position of an atom interacting via the potential $V(\boldsymbol{r}')$ with the nucleus.

and

$$\omega_s = \int\int\int h(\boldsymbol{r}) \sin\phi(\boldsymbol{r})\, \mathrm{d}^3\boldsymbol{r} \tag{17}$$

one can write

$$\langle \mathrm{e}^{\mathrm{i}\alpha} - 1 \rangle = -\mathrm{d}\tau \left(\frac{\gamma_b}{2} - \mathrm{i}\omega_s \right). \tag{18}$$

Putting (18) into (11) gives

$$\mathrm{d}f(\tau) = -\left(\frac{\gamma_b}{2} - \mathrm{i}\omega_s \right) f(\tau)\, \mathrm{d}\tau \tag{19}$$

which can be integrated immediately

$$f(\tau) = \exp\left(-\frac{\gamma_b}{2}\tau + \mathrm{i}\omega_s\tau \right). \tag{20}$$

Putting (20) into (6) gives the first-order correlation function

$$g^{(1)}(\tau) = \exp\left[-\left(\frac{\gamma_b + \gamma}{2} \right)\tau \right] \exp\left[-\mathrm{i}\left(\omega_n - \omega_s \right)\tau \right]. \tag{21}$$

A simple integration gives, according to (1), the radiated frequency spectrum

$$F(\omega) = \frac{1}{2\pi} \frac{\gamma + \gamma_b}{(\omega - \omega_n + \omega_s)^2 + (\gamma + \gamma_b)^2/4}. \tag{22}$$

In the next section a discussion of this result will be given.

3.3. Discussion

Equation (22) shows that the frequency spectrum is a Lorentzian with width $\gamma + \gamma_b$.

If $\gamma_b \gg \gamma$ then the line broadening is essentially due to the interactions of the nuclei with the lattice, a conclusion analogous to the one reached earlier [1–3, 7]. It should be obvious from our analysis that the line broadening is a homogeneous one, since all long-lived nuclei go through the same processes during their complete lifetime. For the usual short-lived nuclear states, the interactions of the nuclei with the lattice in which they are embedded produce a non-homogeneous broadening, since during their (short) lifetime one group of nuclei experiences one kind of interaction while another group sees another kind. In particular for these short-lived nuclei the ergodic theorem does not apply. In the next section a rough estimate of the line broadening will be given.

Simultaneously a frequency shift ω_s is found. Such a shift is absent in all previous analyses. The fundamental origin of the shift is the following. Each time a quantum system (be it a nucleus, an atom or whatever) interacts with another quantum system, where the result is a continuum of final states, such as is the case considered in this article, there is, apart from a line broadening, also a line shift [13]. In the next section an order of magnitude of this shift will be given.

3.4. Numerical estimations of the induced line broadening and shift

If the analysis presented in the previous section is to be applied to particular cases of long-lived isomeric states such as is the case for ^{109}Ag, a specific interaction model will have to be introduced. The validity of the ergodic theorem will also be determined by the nature of the interactions between the radioactive nucleus and its surrounding. This needs a thorough discussion in order to determine whether shorter-lived states, such as the 93 keV state in ^{67}Zn with a nuclear lifetime of 1.3×10^{-5} s and a corresponding natural linewidth of $\sim 5 \times 10^{-11}$ eV, can still be treated with the ergodic theory and, consequently, can give rise also to homogeneous line broadening.

At this stage only a rough estimate of the magnitude of the induced width and shift will be given. Detailed calculations will depend evidently on the actual perturbation mechanisms, which in turn will depend on the specific nucleus–lattice combination. This would constitute a new research field.

A possible simple mechanism [14] for the perturbation that a radioactive nucleus in a solid experiences arises from the spread in precession rates produced by the magnetic field that the neighbouring nuclei produce at the position of the radioactive nucleus, roughly giving a correlation time of about 100 μs. This value could thus be considered as the order of magnitude to determine whether one has a long-lived or a short-lived nuclear state. With this value, the isomeric state of ^{109}Ag (lifetime of about 40 s) would be a long-lived state. However, the 93 keV state in ^{67}Zn would be a short-lived state, for which the present analysis would not be applicable, because the ergodic theorem is not valid.

Finally, an estimation of the energy broadening and line shift will be given below. For a spontaneously emitted wave track, due to e.g. a radioactive nucleus, the coherence time τ can be considered as the inverse of the decay constant γ of the unstable state [13]. The homogeneous line broadening corresponding to this 'relaxation' process is given, as is well known, by $\hbar\gamma$. The radiative decay can be treated as a phase diffusion process, due to which the first-order coherence function [13] decays with a time constant $\tau = 1/\gamma$. Quantum mechanically, this phase diffusion can be considered as produced by the vacuum fluctuations [9]. Analogously to this image, the neighbouring nuclei, perturbing the radioactive nucleus, produce also a phase diffusion of the emitted wave track. If the correlation time of these perturbations is of the order of 100 μs, as has been mentioned above, then the corresponding homogeneous line broadening is of the order of 10^{-12} eV. The estimation of the line shift is impossible without detailed calculations. Assuming a van der Waals type potential for the dipole–dipole interaction [15], which is a crude but for our purpose sufficient approximation, the ratio of the shift to the width, assuming a simple model known as the impact approximation [16], is of the order of 0.18. As has been mentioned above, detailed calculations assuming specific perturbation mechanisms should provide more precise estimations of the line broadening and shift. However, it is often impossible to obtain realistic interaction models (to have e.g. realistic potentials describing the perturbations experienced by a radioactive nucleus from the action of its surroundings). The experimental measurements of the line broadening and line shift could provide information on these interactions.

4. Conclusions

The continuous interactions between a nucleus and its surroundings, in a solid state lattice in which they are embedded, modify the nuclear Bohr frequency. This modification can be considered as a stochastic process. The stochastic variable X associated with this process is the difference between the unperturbed nuclear frequency ω_n and the instantaneous frequency $\omega(t)$. The first-order correlation function, whose Fourier transform gives the frequency spectrum of the photons emitted by the nuclei, depends amongst other things on $\langle \exp[\mathrm{i} \int_t^{t+\tau} x(t')\,\mathrm{d}t'] \rangle$ with $\langle\rangle$ the ensemble average, $x(t')$ being the value of X at time t'. The calculation of this average has been based on the application of the ergodic theorem. The frequency spectrum deduced from it is Lorentzian with total homogeneous width equal to the sum of the natural line width and a lattice induced width. Simultaneously a frequency shift has been found. The detailed calculations of the magnitude of the line broadening and shift depend on particular interaction models. Rough estimates give for the line broadening 10^{-12} eV and for the shift an order of magnitude lower. This homogeneous line broadening could be invoked to explain why a Mössbauer effect could be observed using long-lived nuclear states such as the isomeric state in ^{109}Ag.

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