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Frequency distribution of a gamma photon emitted without recoil by a nucleus considering multiple scattering processes

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Abstract

The time-dependent frequency distribution of a gamma photon emitted without recoil by an excited source nucleus is calculated; the emitted photon is submitted to nuclear quasi-resonant scattering in an absorber and recorded by a detector. The problem is formulated for an arbitrary number of quasi-resonant nuclei in the absorber, including both radiative decay (decay by means of gamma radiation) and decay by electron conversion (decay where the nuclear de-excitation energy is transmitted directly to an atomic electron). The problem of multiple scattering is treated by means of the so-called coherent path model developed years ago. The properties of the radiation leaving the absorber under these conditions are complex. The calculated probabilities of finding a gamma photon thus emitted as a function of frequency show a rich structure, with among others, maxima and minima that vary with time. The frequency distribution changes continuously as a function of time. For times long compared to the nuclear mean life of the excited state, the envelop of the frequency distribution resembles a Lorentz distribution, the details show some oscillatory behaviour as well as an asymmetry with respect to the nuclear frequency in the case of quasi-resonance between the source and the absorber nuclei. If the source and absorber nuclei are in exact resonance, the frequency distribution is symmetric but the oscillatory behaviour is still present. The frequency distribution is determined by the source nucleus as well as by the quasi-resonant absorber nuclei, hence by the multiple scattering processes. The model presented can be used to calculate analytically the properties of the transmitted gamma radiation for any number of absorber nuclei, although the simulation of the results will obviously necessitate some tedious computer work.

Keywords: *Gamma photon; multiple scattering processes; Mössbauer Effect; Frequency distribution; Lorentz distribution; resonant nuclei*

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Introduction

In an earlier paper (Hoy and Odeurs, 2012) the frequency distribution of a gamma photon was studied when an excited nucleus decays by emitting gamma radiation. It was reported that the frequency distribution changes with time to reach its limit form when the time is much longer than the average lifetime of the excited nuclear state. This limiting form is the well-known Lorentz distribution, concentrated about a central

frequency and having a width at half maximum equal to the natural linewidth.

In this paper the frequency distribution is studied in the case where the photon emitted by a source nucleus undergoes multiple scatterings from quasi-resonant nuclei in an absorber, where the emission and absorption of gamma rays without recoil, called the Mössbauer Effect (Greenwood and Gibb, 1971) was considered.

The problem is formulated for an arbitrary number of quasi-resonant nuclei in the absorber, including both radiative decay i.e. decay by means of gamma radiation and decay by electron conversion where, the nuclear de-excitation energy is transmitted directly without production of a gamma photon to an atomic electron (Greenwood and Gibb, 1971). To make the calculations as well as the interpretation somewhat more transparent, the problem of multiple scattering by two nuclei in the absorber will be considered, although the problem can be solved for the general case, leading to exceedingly long and rather cumbersome expressions for large numbers of scattering nuclei.

In Section 3 the frequency distribution of the scattered gamma radiation is investigated in the (slightly) off-resonance case as well for the exact resonant condition.

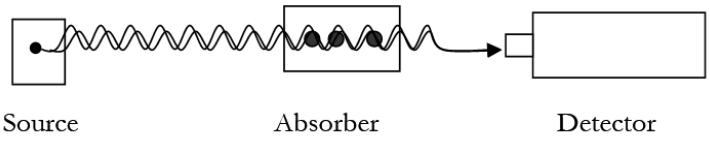
Materials and Methods

The following setup is considered: a polycrystalline absorber containing resonant nuclei, all in the ground state (in this case the stable ground state of ^{57}Fe, having a natural abundance of 2.12%), is placed between a source nucleus, in an excited state. In this case, the first excited state of ^{57}Fe, which is at 14.4 keV above the stable ground state of ^{57}Fe, Figure 2. In an actual experiment a γ-ray detector would be mounted behind the absorber to register and analyse the transmitted gamma radiation.

A forward scattering, i.e., the propagation direction of the gamma radiation is defined by a straight line, chosen to be the x-axis, from the source nucleus to the centre of the detector whose aperture is assumed to be sufficiently small, Figure 1. In the third sub-section of section 2 a more thorough analysis will be shown with respect to this forward propagation direction.

Figure 1

Scheme of multiple scattering



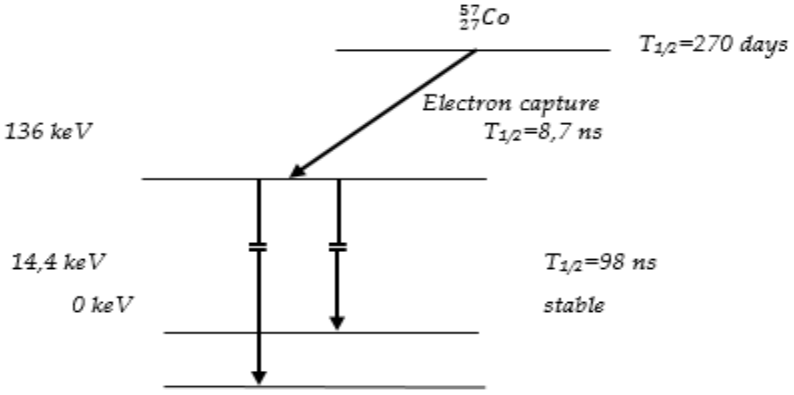
When gamma radiation emitted by the source interacts with the resonant nuclei of the absorber, its intensity versus time can be recorded by the detector. The number N of nuclei encountered depends directly on the thickness of the absorber (Hoy, 1997). At time $t = 0$, only the source nucleus is excited. The problem of determining this initial time is discussed in the fourth sub-Section of section 2.

In Figure 1, multiple scattering processes are represented for the case of three absorber nuclei as per the setup used in this article. When a photon is recorded by the detector, there is no possible way to know which path has been taken by the photon emitted by the source nucleus. According a fundamental principle of Quantum Mechanics, all scattering amplitudes have to be considered and the total detection probability for the scattered photon will be proportional to the absolute value of the square of the total amplitude, which is the sum of all possible scattering amplitudes. Hoy (1997) has shown this first for a radioactive source and a resonant

absorber. Hoy *et al.* (2001) have shown this in detail for synchrotron radiation interacting with quasi-resonant absorber nuclei. This gives rise to interferences, constructive and destructive, between all possible amplitudes. The presence of these interferences is crucial in understanding the properties of the transmitted gamma radiation.

Figure 2

Decay scheme of $^{57}Co/^{57}Fe$, showing the relevant nuclear states



The number of scattering nuclei N has been modelled by means of the following equation (1), (Hoy, 1997):

$$N = \frac{\beta \times \Gamma}{2f \times \Gamma_r} \quad (1)$$

where Γ is the total linewidth, Γ_r the radiative linewidth, β the resonant nuclear thickness parameter (effective thickness) defined by equation (2) (Greenwood and Gibb, 1971):

$$\beta = N_o f \sigma_o d \quad (2)$$

with N_o the number of resonant nuclei per cubic centimetre, f the recoil-free fraction (this is the probability that a nucleus will emit or absorb a gamma photon without recoil, so it is the Mössbauer fraction (Greenwood and Gibb, 1971)), σ_o is the maximum effective cross-section evaluated at resonance, and d is the thickness of the sample (the absorber). In general the value of N, defined by equation (1), will not be an integer. It is implicitly understood that the value for N is the integer closest to the value of the right-hand side of equation (1).

As previously cited (Hoy, 1997; Hoy *et al.*, 2001), the complete quantum system is considered consisting of the source nucleus, the absorber nuclei, the gamma radiation and the conversion electrons. Before proceeding to the equations describing this system, it is worth digressing a little on the process of the production of a conversion electron, also called internal conversion.

The process of producing a conversion electron (internal conversion of a nuclear transition)

An excited nuclear state, spontaneously decaying to a less excited (often stable) state by electromagnetic interaction, has two different possibilities.

The first possibility is the emission of a gamma photon, which can therefore be easily detected using standard nuclear detection techniques. This process is characterised by a constant, called the radiative constant Γ_r, which is directly related to the probability of decay through the production of a gamma photon.

The second possibility, called the internal conversion, is the direct transition from the nuclear excited state to the ground state without the production of an actual gamma photon. So the excited nuclear energy is directly transferred

to an atomic electron (from an inner electronic shell) belonging to the nucleus in question. This electron, called a conversion electron, can therefore be ejected with a certain kinetic energy, which is the difference of the total available nuclear energy and the atomic binding energy of the ejected electron. In the case of ^{57}Fe this energy is of the order of several keV. In principle the properties of this conversion electron can be measured. This process is characterized by a constant Γ_c, characterizing the decay probability of the nucleus through electron conversion.

The total decay constant Γ, with respect to the considered unstable nuclear level, is the sum of the two decay constants: $\Gamma = \Gamma_r + \Gamma_c$. For the first excited state of ^{57}Fe, having a half-life of 98 ns, the internal conversion process is about nine time more probable than the process of radiative decay (Greenwood and Gibb, 1971).

It can be mentioned at this stage that, although the majority of Mössbauer spectroscopy experiments has been based on the detection of transmitted gamma radiation, a non-negligible part is performed on a set-up based on the detection of conversion electrons (Greenwood and Gibb, 1971). This technique, called Conversion Electron Mössbauer Spectroscopy (CEMS), is particularly useful when the electron conversion coefficient is high and for the study of specific properties such as the investigation of nuclear sites situated close the surface of a solid-state material in which they are embedded (Barancira et al., 1996).

Basic equations

The evolution of the complete quantum system defined (Hoy, 1997; Odeurs *et al.*, 2000; Hoy *et al.*, 2001, Odeurs and Hoy, 2006; Havyarimana *et al.*, 2021) is described by the familiar Schrödinger equation in time domain:

$$i\hbar \frac{d|\psi(t)\rangle}{dt} = H|\psi(t)\rangle \qquad (3)$$

The state vector $|\psi(t)\rangle$ of the complete quantum system contains all information. H is the total hamiltonian of the complete system, which will be determined below.

The hamiltonian of the system is divided into two parts. First the unperturbed hamiltonian, H_0, corresponding to the nuclear states, the free radiation field, taken as plane waves, and the conversion electrons. The second part of the total hamiltonian, H_{int}, describes the interactions between the nuclei, the electromagnetic field associated to the gamma photons and the conversion electrons. H_{int} describes the transitions between the eigenstates of H_0 by allowing the nuclei to absorb and emit gamma radiation or conversion electrons. The total hamiltonian H, is of course the sum of H_0 and H_{int}.

The entire system seems to be relatively unmanageable but it will later become clear that a suitable (Fourier) transformation makes the structure more transparent.

In the standard approach the state of the system, $|\psi(t)\rangle$, can be expressed by equation (4).

$$|\psi(t)\rangle = \sum_p a_p(t) e^{-i\left(\frac{E_p t}{\hbar}\right)} |\varphi_p\rangle \qquad (4)$$

where $\{|\varphi_p\rangle\}$ is the ensemble of all eigenstates of H_0. The state $|\varphi_p\rangle$ is associated to the energy E_p.

Solving the Schrodinger equation in the usual way, one arrives at a set of coupled differential equations relating the coefficients $a_p(t)$:

$$i\hbar \frac{da_p(t)}{dt} = \sum_m a_m(t) e^{i(\omega_p - \omega_m)t} |\varphi_p\rangle\langle\varphi_p|H|\varphi_m\rangle + i\hbar\delta_{pl}\delta(t) \qquad (5)$$

The matrix element $\langle\varphi_p|H|\varphi_m\rangle$, which can be written as H_{pm}, describes the transition between the states $|\varphi_m\rangle$ and $|\varphi_p\rangle$. Obviously, the unperturbed part of the hamiltonian, H_0, does not induce transitions between two eigenstates of H_0. Therefore, although the total hamiltonian, H, in equation (5) is maintained, only the perturbation, given by H_{int}, will produce non-zero matrix elements in equation (5).

The Kronecker delta and the delta function on the right-hand side in (5) are needed (Heitler, 1954) to denote that at time $t = 0$ the system is in the state where $p = l$ (i.e. for this case only the source nucleus is excited). In the fourth sub-section of section 2 a short digression is given on the experimental determination of the "initial time" of the system at $t = 0$.

With Heitler (Heitler, 1954), a form of Fourier transformation $A_m(\omega)$ for the amplitude $a_m(t)$ can be introduced:

$$a_m(t) = -\frac{1}{2\pi i}\int_{-\infty}^{\infty} A_m(\omega) e^{i(\omega_m-\omega)t}\, d\omega \qquad (6)$$

The substitution of expression (6) in equation (5) leads (Heitler, 1954) to a system of coupled linear equations for the $A_m(\omega)$. In such a way, the coupled differential equations expressed by (5) are replaced by the set of linear coupled equations (7) – (12), which is an appreciable simplification for the analysis of the whole problem. It should be mentioned at this stage that the variable *t* is now replaced by the new variable ω, having the dimension of s^{-1}, which is, obviously, the complete idea of Fourier analysis.

In Havyarimana *et al*. (2022), the method has been used to study the nuclear radiation field and the nuclear excitation inside a resonant absorber, leading to the concept of nuclear polariton, i.e. the combined system consisting of gamma photons and the resonant nuclei in the absorber that interact with each other. More details of the mathematical technique are also given in (Havyarimana *et al*., 2022).

There are five possible amplitudes for this problem:

- $A_m(\omega)$: the source nucleus, situated at the origin of the coordinates system, is excited and its energy (the excited state energy) is given by $\hbar\omega_o$, where ω_o is the resonant frequency of the source, all absorber nuclei are in the ground state and no photon or conversion electron present;
- $B_k(\omega)$: all nuclei are in the ground state and only one photon is present of wave number k and energy $\hbar\omega_k$ with no conversion electron present (see the discussion in D to explain why the wave numbers *k* are used rather than wave vectors);
- $C_m(\omega)$: the absorber nucleus located at position $x = x_m$ is excited and its energy is given by $\hbar\omega_o'$ where ω_o' is the resonant frequency of the absorber nuclei (here assume that there may be a (slight) difference between the nuclear energy of the source and those of the absorber nuclei), and no photon or conversion electron present;
- $D_p(\omega)$: a conversion electron from the source nucleus is present, its momentum is p while all nuclei are in the ground state (here the problem is not formulated in three dimensions in order to keep the notation as simple as possible without detracting from generality as will be seen that the only effect of the internal conversion processes will be the introduction of a linewidth Γ_c in the equations);
- $E_{mp}(\omega)$: a conversion electron from the absorber nucleus located at position $x = x_m$ is present, while all nuclei are in the ground state and no gamma photon is present.

Before continuing the theoretical analysis, it is important to discuss at this stage the forward direction in some detail. A gamma photon emitted from a source nucleus in the forward direction will arrive at the detector if there has not been any interaction between the gamma photon and the absorber nuclei, initially all in the ground state. If the emission direction is not in the forward direction, the gamma photon will not arrive at the detector if no interaction between the absorber nuclei has occurred. If this process is the only one present, the detector would register gamma photons coming directly from spontaneous gamma emission and the frequency spectrum would be a familiar Lorentzian line (Greenwood and Gibb, 1971; Odeurs *et al.,* 2000) for long times compared to the half-life of the nuclear excited state.

If interactions occur between an emitted gamma photon and the absorber nuclei, one of them will be excited by absorption of the photon, implying that this gamma photon will disappear and the absorber nucleus will pass to the excited state. It will then emit a gamma photon, passing again to the nuclear ground state. This secondary gamma photon will be detected only when it has the right direction, i.e., if it is emitted in the forward direction.

At this stage of the discussion, several remarks, A to G, are made as follows.

A. When a gamma photon is detected, it is impossible to know which physical process has occurred: it could be the photon coming straight from the source nucleus without any interaction with an absorber nucleus or it could be a gamma photon that was emitted by an absorber nucleus after this one absorbed the initial gamma photon coming from the source nucleus. If there are many identical absorber nuclei present in the absorber, each one of them can absorb and re-

emit a gamma photon. Only when this re-emitted gamma photon has the right direction, i.e. again the forward direction, it will reach the detector. There is no possible way to know which absorber nucleus has absorbed and re-emitted this gamma photon (Heeg *et al.,* 2021).

B. Multiple scattering, i.e. multiple absorptions and re-emissions of a gamma photon by two or more absorber nuclei, can occur. Again, when a gamma photon is detected, there is no possible way to know which processes have occurred. Therefore, all possibilities have to be taken into account, each one being described by its quantum mechanical probability amplitude (Heeg *et al.,* 2021).

C. The total quantum mechanical amplitude is the sum of the individual amplitudes. The probability to detect a gamma photon is proportional to the square of the absolute value of the total amplitude. This probability contains a sum of the squares of the absolute values of the individual amplitudes but also "cross terms" containing products of two or more individual amplitudes. These cross terms give rise to the so-called interferences between the various terms (Odeurs and Hoy, 2005). These interference terms appear to be crucial.

D. It is clear that the forward direction plays a crucial role because a detected photon must have the forward direction, otherwise it will not be detected. It could be argued that the secondary gamma photons could be emitted in any direction, as long as the "last" photon produced is in the forward direction. However, the detailed structure of the equations in three dimensions (Odeurs *et al.*, 2000) shows that the processes describing the absorption and re-emission of gamma photons contain the positions of the absorber nuclei leading to "phase factors" such as $exp(\pm i\vec{k}\cdot\vec{r}_m)$ (Schiff, 1968), with $\vec{r}_m$ the position, with respect to the origin of a coordinate system, of absorber nucleus labeled m. In what follows the position of the source nucleus is taken as the origin of the coordinate system. The $\pm$ signs are connected to absorption (+) and emission (−). The detailed analysis presented in Odeurs *et al.,* 2000 shows that for random positions of the nuclei in the absorber, such as those in an amorphous or a polycrystalline material, the presence of the phase factors has as a result that the interference terms, mentioned in C., give a total contribution that is completely negligible for all directions except for the forward direction. It is also shown (Odeurs *et al.,* 2000) that radiation temporarily going "backward", i.e. back in the direction of the source nucleus, will also be associated to a negligible amplitude. Thus, it can be concluded that only the forward direction has to be considered, also for the "intermediate" gamma photons in interacting present in the absorber. It means that the problem can be simplified by restricting it to the forward direction, which will be taken as the x-direction. This has already been anticipated in the notation of the amplitude $B_k(\omega)$, defined above.

E. For an absorber that is a single crystal, there can be constructive interferences for particular directions, called Bragg directions (Ashcroft and Mermin, 1976); for nuclear Bragg scattering, see e.g. Smirnov and Chumakov, (2019). In this case the problem has to be reformulated if these Bragg directions are considered. The forward direction, however, remains here to a privileged direction and the present formulation of the problem in one dimension stands.

F. Emission and re-absorption of conversion electrons can be considered as well. Here too, there will be phase factors, $exp(\pm\frac{\vec{p}}{\hbar}\cdot\vec{r}_m)$, similar to those for photon emission and absorption. $\vec{p}$ is the linear momentum of the conversion electron. However due to the short range of the electrons in a solid-state material, only emission and re-absorption by the same atom/nucleus are considered. It turns out that these processes give only rise to line broadening of the nuclear excited state due to process of conversion electron, leading to the width, Γ_c, of the nuclear excited state due to conversion electron, mentioned already in the second sub-section of Section 2. To maintain the notations as simple as possible, the mathematical problem will be formulated for the conversion electron processes also in one dimension, bearing in

mind that the final result for these processes, the line partial broadening, corresponds to the actual Γ_c.

G. In fact the same discussion could be done for radiative broadening, which is due to emission and sub-sequent self-absorption of gamma photons by a nucleus in the excited state. In the language of quantum electrodynamics, these emission and re-absorption processes are represented by the so-called Feynman diagrams, see e.g. (Schweber, 1994). For the radiative width, the formulation is kept in one dimension although it could be easily extended in three dimensions. The partial line broadening corresponding to the photon emission and self-absorption by a nucleus will always give the radiative width Γ_r.

Using equations (5) and (6) the general expression leads to the following equation (Hoy, 1997):

$$(\omega - \omega_n + i\varepsilon)A_n(\omega) = \sum_q A_q(\omega)\frac{\langle\varphi_n|H|\varphi_q\rangle}{\hbar} + \delta_{nl}. \qquad (7)$$

The presence of the Kronecker delta δ_{nl} in equation (7) accounts for the fact that the system is initially in the $|\Psi_l\rangle$ state (Heitler, 1954), which is the state where the source nucleus is excited, the absorber nuclei in the ground state and no photon or conversion electron are present. The infinitesimal positive real quantity ε is to ensure causality, i.e. the excited nuclear state of the source nucleus cannot be formed before $t = 0$. In the next subsection the precise determination of this initial time, $t = 0$, will be considered further.

Substituting all five possible amplitudes into equation (7), the following system of coupled linear equations are obtained:

$$(\omega - \omega_o + i\varepsilon)A(\omega) = 1 + \sum_k \frac{B_k(\omega)H_k}{\hbar} + \sum_p \frac{D_p(\omega)H_p}{\hbar} \qquad (8)$$

$$(\omega - \omega_k + i\varepsilon)B_k(\omega) = \frac{A(\omega)H_k^*}{\hbar} + \sum_m \frac{C_m(\omega)H_k^*}{\hbar}e^{-ikx_m} \qquad (9)$$

$$(\omega - \omega_o' + i\varepsilon)C_m(\omega) = \sum_k \frac{B_k(\omega)H_k}{\hbar}e^{ikx_m} + \sum_p \frac{E_{mp}(\omega)H_p}{\hbar}e^{i(\frac{p}{\hbar})x_m} \qquad (10)$$

$$(\omega - \omega_p + i\varepsilon)D_p(\omega) = \frac{A(\omega)H_p^*}{\hbar} \qquad (11)$$

$$(\omega - \omega_p + i\varepsilon)E_{mp}(\omega) = \frac{C_m(\omega)H_p^*}{\hbar}e^{-i(\frac{p}{\hbar})x_m} \qquad (12)$$

The factors H_k et H_k^* are matrix elements corresponding to the absorption and emission of the photon respectively while H_p et H_p^* are matrix elements corresponding to the absorption, by the atom to which the nucleus in question belongs, and the emission of a conversion electron, respectively. It is assumed that for $t < 0$, all amplitudes are zero (Heitler, 1954). This important point will be discussed in the third sub-section of Section 2.

The set of equations (8)-(12) can be interpreted in a straightforward manner. To elucidate them, (9) and (10) are considered.

Equation (9) gives the evolution of $B_k(\omega)$, which is the amplitude related to the presence of a photon with wave number k. This photon can be produced by the source nucleus, with amplitude $A(\omega)$, as well as by all absorber nuclei, if they are excited, with amplitudes $C_m(\omega)\ \forall\ m$. The emission by absorber nucleus m is described by the phase factor e^{-ikx_m}, as has been explained in D. above. For the source the phase factor is obviously 1 because it is chosen to be at the origin of the coordinate system.

Equation (10) gives the evolution of $C_m(\omega)$, which is the amplitude related to absorber nucleus m. The sum $\sum_k \frac{B_k(\omega)H_k}{\hbar}e^{ikx_m}$ represents the absorption processes of photons, having wave number k, by nucleus m, hence the presence of the phase factor e^{ikx_m}. $\sum_p \frac{E_{mp}(\omega)H_p}{\hbar}e^{i(\frac{p}{\hbar})x_m}$

represents the self-absorption by nucleus m of the conversion electron produced by it, hence the phase factor $e^{i(\frac{p}{\hbar})x_m}$. As mentioned before, this equation is related to the conversion electron linewidth Γ_c.

All equations can be interpreted in a similar matter.

Determination of $t = 0$

The time $t = 0$ is defined as the time of formation of the source nucleus in its excited state. At this time, all the absorber nuclei are still in the ground state and there is not yet a gamma photon or conversion electron present. The time of formation of the excited nuclear state can be determined experimentally by the precursor method and the detector coincidence method, as will be explained in the following. To illustrate this method, consider the radioactive decay of the Mössbauer isotope of ^{57}Fe (Fig. 2).

By electronic capture the 136 keV excited state above the ground state of ^{57}Fe can be formed from ^{57}Co (which can be produced in a nuclear reactor). From the 136 keV state, ^{57}Fe is de-excited and produces the 14.4 keV state (90% of cases), which is the first excited state of ^{57}Fe. The system evolves over time and the nucleus eventually returns to the stable ground state. The formation time of the 14.4 keV state can be determined using two detectors, D1 and D2. D1 detects the arrival of the 122 keV, "precursor", photon (the difference between 136 keV and 14.4 keV), D2 measures the arrival of the 14.4 keV photon. As long as D1 has not given a signal, the 14.4 keV state has not yet been formed. The time $t = 0$ is then defined by the time when D1 has recorded a 122 keV photon. At this precise moment, D2 is switched on and it is this moment that defines the initial time $t = 0$ of an experiment. This is called the two-detector coincidence method: the second detector, here D2, starts recording only when it has received a signal from the first, D1, meaning here the detection of a 122 keV photon, the "precursor" to the formation of the 14.4 keV nuclear state. The system then evolves from this initial time. By this coincidence method all amplitudes, including the one corresponding to the source nucleus in the 14.4 keV excited state, are zero for $t < 0$, (Zagato et al., 2014) and the system defined above only starts to evolve from $t = 0^+$.

Solution

The system of equations (8)-(12) can be solved exactly for any number of absorber nuclei (Hoy, 1997). In order to highlight the essential elements of the results, the following the case of double scattering, i.e. with two nuclei in the absorber, are considered.

It has been shown (Hoy, 1997) that, when the absorber contains only two quasi-resonant Mössbauer nuclei, the amplitude $b_{\omega_k}(t)$ of finding a photon of frequency ω_k emitted at time t is given by:

$$b_k(t) = \frac{H_k^*}{\hbar}\left[\frac{1 - e^{-i\left(\omega_o - \omega_k - i\frac{\Gamma}{2\hbar}\right)t}}{\omega_k - \omega_o + i\frac{\Gamma}{2\hbar}}\right] + 2\frac{H_k^*}{\hbar}\left(-i\frac{\Gamma_r}{2\hbar}\right)\frac{1}{\left(\omega_k - \omega_o + i\frac{\Gamma}{2\hbar}\right)\left(\omega_k - \omega_o' + i\frac{\Gamma}{2\hbar}\right)} + \frac{H_k^*}{\hbar}\left(-i\frac{\Gamma_r}{2\hbar}\right)^2 \times \frac{1}{\left(\omega_k - \omega_o + i\frac{\Gamma}{2\hbar}\right)\left(\omega_k - \omega_o' + i\frac{\Gamma}{2\hbar}\right)^2} \quad (13)$$

where ω_o is the nuclear Bohr frequency of the source nucleus, ω_o' is the Bohr frequency of the absorber nuclei. At exact resonance, both frequencies are equal.

Results

Non-resonance case ($\omega_o \neq \omega_o'$)

The equation (13) of the amplitude $b_k(t)$ is the starting point to find a photon of frequency ω_k present at time t. The subscript k is changed to ω_k to emphasise the frequency dependence ω_k of $b_k(t)$. The corresponding time-dependent probability density of finding a gamma photon of frequency ω_k is:

$$\rho = Pr\,o\,b_{\omega_k}^{off\_resonance}(t) = \left|b_{\omega_k}(t)\right|^2$$

$$= \left| \frac{H_k^*}{\hbar} \left[\frac{1 - e^{-i\left(\omega_o - \omega_k - i\frac{\Gamma}{2\hbar}\right)t}}{\omega_k - \omega_o + i\frac{\Gamma}{2\hbar}} \right] + 2\frac{H_k^*}{\hbar}\left(-i\frac{\Gamma_r}{2\hbar}\right)\frac{1}{\left(\omega_k - \omega_o + i\frac{\Gamma}{2\hbar}\right)\left(\omega_k - \omega_o' + i\frac{\Gamma}{2\hbar}\right)} + \frac{H_k^*}{\hbar}\left(-i\frac{\Gamma_r}{2\hbar}\right)^2 \times \frac{1}{\left(\omega_k - \omega_o + i\frac{\Gamma}{2\hbar}\right)\left(\omega_k - \omega_o' + i\frac{\Gamma}{2\hbar}\right)^2} \right|^2 \quad (14)$$

After somewhat tedious calculations

$$\rho(t) = Pr\,o\,b_{\omega_k}^{off\_resonance}(t) = \frac{|H_k^*|^2}{\hbar^2} \left\{ \left\{ \left[(\omega_k - \omega_o')^2 - (\omega_k - \omega_o')^2 e^{-\frac{\Gamma}{2\hbar}t} cos(\omega_k - \omega_o)\, t + \left(\frac{\Gamma}{2\hbar}\right)^2 e^{-\frac{\Gamma}{2\hbar}t} cos(\omega_k - \omega_o)\, t - \left(\frac{\Gamma}{2\hbar}\right)^2 - \left(\frac{\Gamma_r}{2\hbar}\right)^2 + \frac{\Gamma_r \Gamma}{2\hbar^2} + \frac{\Gamma}{\hbar}(\omega_k - \omega_o') e^{-\frac{\Gamma}{2\hbar}t} sin(\omega_k - \omega_o)\, t \right]^2 + \left[(\omega_k - \omega_o')^2 e^{-\frac{\Gamma}{2\hbar}t} sin(\omega_k - \omega_o)\, t - \left(\frac{\Gamma}{2\hbar}\right)^2 e^{-\frac{\Gamma}{2\hbar}t} sin(\omega_k - \omega_o)\, t + \frac{\Gamma}{\hbar}(\omega_k - \omega_o') - \frac{\Gamma}{\hbar}(\omega_k - \omega_o') e^{-\frac{\Gamma}{2\hbar}t} cos(\omega_k - \omega_o)\, t - \frac{\Gamma_r}{\hbar}(\omega_k - \omega_o') \right]^2 \right\} \times \left\{ \left[(\omega_k - \omega_o)(\omega_k - \omega_o')^2 - \left(\frac{\Gamma}{2\hbar}\right)^2 (\omega_k - \omega_o) - \frac{\Gamma^2}{2\hbar^2}(\omega_k - \omega_o') \right]^2 + \left[\frac{\Gamma}{\hbar}(\omega_k - \omega_o) \times (\omega_k - \omega_o') + \frac{\Gamma}{2\hbar}(\omega_k - \omega_o')^2 - \left(\frac{\Gamma}{2\hbar}\right)^3 \right]^2 \right\}^{-1} \right\} \quad (15)$$

The equation (15), interpreted in the first subsection of the discussion, gives the time dependence of the frequency composition of the scattered photon during the quasi-resonant nuclear scattering process by two nuclei.

In the next subsection the resonance condition is considered.

Resonance case ($\omega_o = \omega_o'$)

If the source and absorber nuclei are in exact resonance, then $\omega_o = \omega_o'$. Under this condition, the probability of finding a photon of frequency ω_k emitted at time t given by equation (14) simplifies because, by substituting ω_o' by ω_o:

$$\rho(t) = Pr\,o\,b_{\omega_k}^{in_{resonance}}(t)$$

$$= \frac{|H_k^*|^2}{\hbar^2}\left\{\left\{\left[(\omega_k-\omega_0)^2-(\omega_k-\omega_0)^2 e^{-\frac{\Gamma}{2\hbar}t}\cos(\omega_k-\omega_o)\,t+\left(\frac{\Gamma}{2\hbar}\right)^2 e^{-\frac{\Gamma}{2\hbar}t}\cos(\omega_k-\omega_o)\,t\right.\right.\right.$$

$$\left.-\left(\frac{\Gamma}{2\hbar}\right)^2-\left(\frac{\Gamma_r}{2\hbar}\right)^2+\frac{\Gamma_r\Gamma}{2\hbar^2}+\frac{\Gamma}{\hbar}(\omega_k-\omega_0)e^{-\frac{\Gamma}{2\hbar}t}\sin(\omega_k-\omega_o)\,t\right]^2$$

$$+\left[(\omega_k-\omega_0)^2 e^{-\frac{\Gamma}{2\hbar}t}\sin(\omega_k-\omega_o)\,t-\left(\frac{\Gamma}{2\hbar}\right)^2 e^{-\frac{\Gamma}{2\hbar}t}\sin(\omega_k-\omega_o)\,t+\frac{\Gamma}{\hbar}(\omega_k-\omega_0)\right.$$

$$\left.\left.-\frac{\Gamma}{\hbar}(\omega_k-\omega_0)e^{-\frac{\Gamma}{2\hbar}t}\cos(\omega_k-\omega_o)\,t-\frac{\Gamma_r}{\hbar}(\omega_k-\omega_0)\right]^2\right\}$$

$$\times\left\{\left[(\omega_k-\omega_o)^3-\left(\frac{\Gamma}{2\hbar}\right)^2(\omega_k-\omega_o)-\frac{\Gamma^2}{2\hbar^2}(\omega_k-\omega_0)\right]^2\right.$$

$$\left.\left.+\left[\frac{\Gamma}{\hbar}(\omega_k-\omega_o)^2+\frac{\Gamma}{2\hbar}(\omega_k-\omega_0)^2-\left(\frac{\Gamma}{2\hbar}\right)^3\right]^2\right\}^{-1}\right\} \qquad (16)$$

Equation (16) again shows the time dependence of the frequency composition of the scattered photon, the time being expressed again in terms of the natural lifetime τ. The interpretation of this equation (16) is given in the second subsection of the discussion.

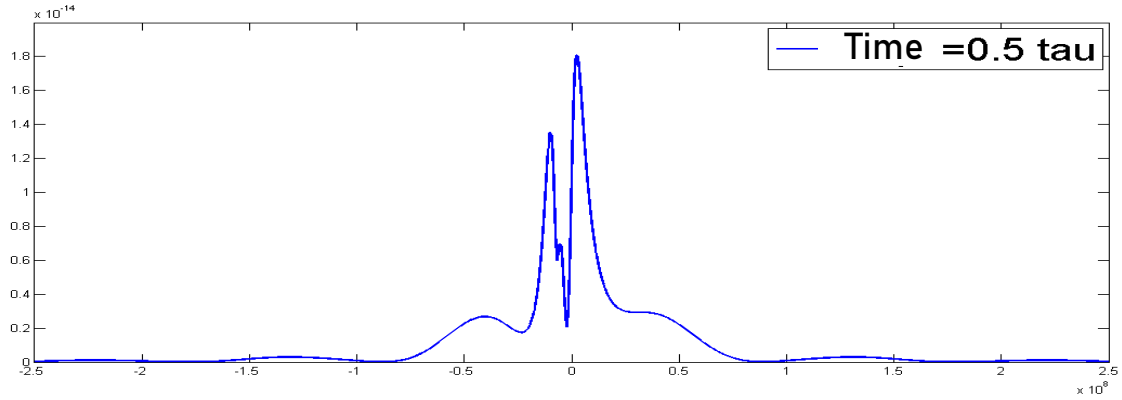
Discussion

Non-resonance case $(\boldsymbol{\omega_o \neq \omega_o'})$

The contribution to the equation (15) contains several terms: emission from the source and scattering from the absorbing nuclei. There are contributions from simple scattering, where the gamma photon is scattered by one nucleus and goes directly to the detector. Another phenomenon is also possible: the photon emitted by the source can be absorbed by the first absorber nucleus, which in turn will emit a photon. This photon can go straight to the detector, or it can be absorbed by the other absorber nucleus before reaching the detector. It has been shown (Odeurs *et al.*, 2000) that backscattering processes, where a photon "returns backward", do not have to be considered because of destructive interference, as has been mentioned before.

Figure 3

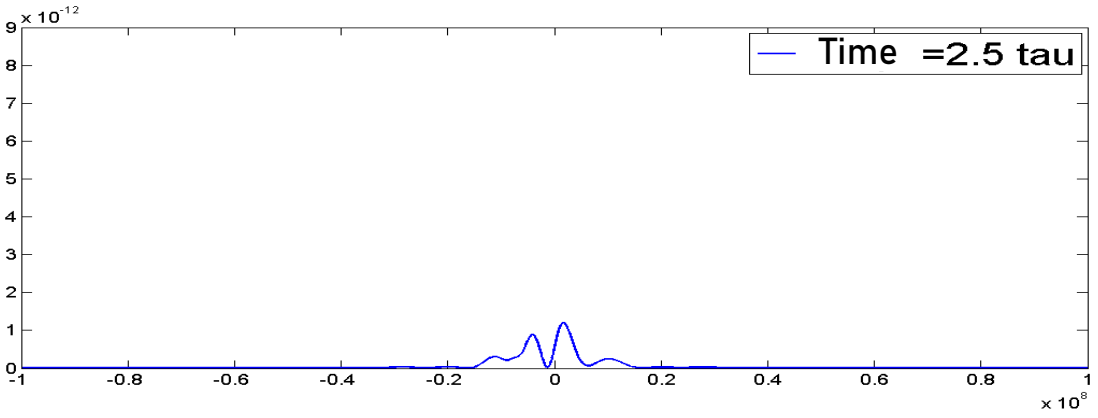
Probability density for $t = 0.5\tau$ *case of* ^{57}Fe *off-resonance where* $\omega_o' - \omega_o = \frac{\Gamma}{\hbar}$.



$\omega_k - \omega_o$

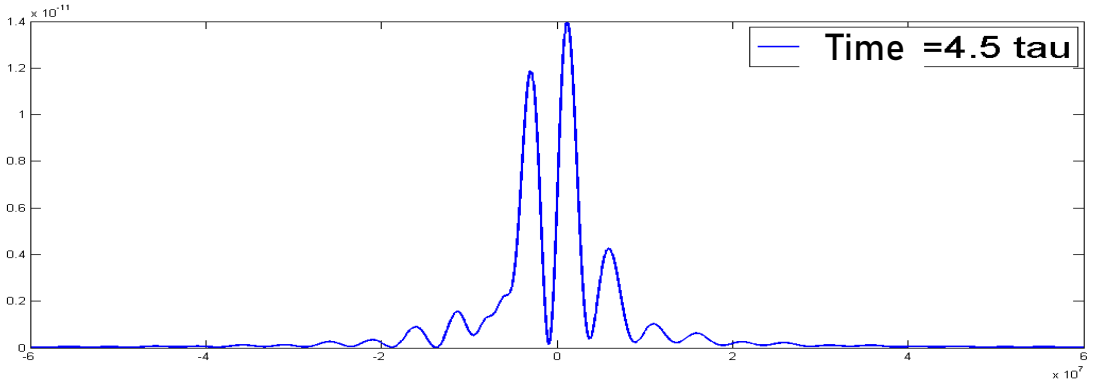
Figure 4

Probability density for $t = 2.5\tau$ *case of* ^{57}Fe *off-resonance where* $\omega_o' - \omega_o = \frac{\Gamma}{\hbar}$.



$\Omega_k - \omega_o$

Figure 5. *Probability density for* $t = 4.5\tau$ *case of* ^{57}Fe *off-resonance where* $\omega_o' - \omega_o = \frac{\Gamma}{\hbar}$



$\omega_k - \omega_o$

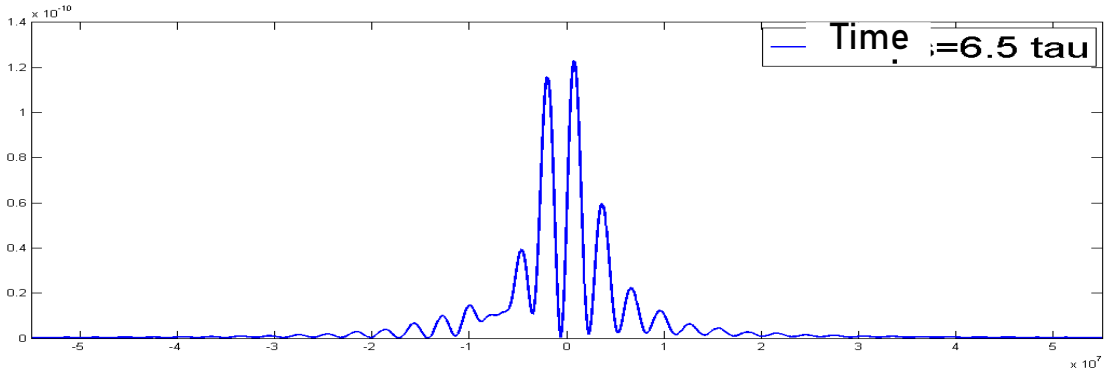
Figures (3) to (7) illustrate the photon probability density as a function of the difference of the photon frequency and the nuclear frequency, $\omega_k - \omega_o$, for various times that are expressed in terms of the lifetime τ of the nuclear excited state, in the case of the off-resonance condition, where $\omega_o' - \omega_o = \frac{\Gamma}{\hbar}$ is taken, i.e. the difference of the two nuclear energies is equal to one total linewidth.

Overall, as the time increases, it is noticed that the frequencies and the overall appearance of the scattered photon change. The envelope of the frequency distribution resembles the Lorentz distribution for a large time interval as shown on figure 7 ($t = 20.5\ \tau$). Moreover, the probability of finding certain frequencies increases considerably with time, figure 6 confirms this assertion. With increasing time, the area defined by the curve and the horizontal axis continues to increase until it reaches its maximum value for large t. This statement can also be confirmed by these figures if they are put on the same scale. It

can be seen that the behaviour of the gamma photon is quite complex, which is due to the scattering processes as well as to the off-resonance condition.

Figure 6.

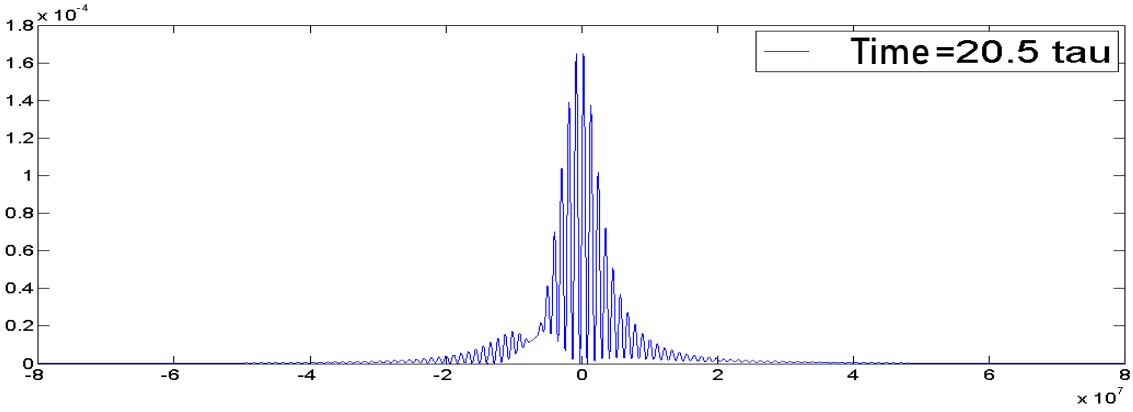
Probability density for $t = 6.5\tau$ *case of* ^{57}Fe *off-resonance where* $\omega_o' - \omega_o = \frac{\Gamma}{\hbar}$.



$\omega_k - \omega_o$

Figure 7

Probability density for $t = 20.5\tau$ *case of* ^{57}Fe *off-resonance resonance where* $\omega_o' - \omega_o = \frac{\Gamma}{\hbar}$.



$\omega_k - \omega_o$

Resonance case $(\omega_o = \omega_o')$

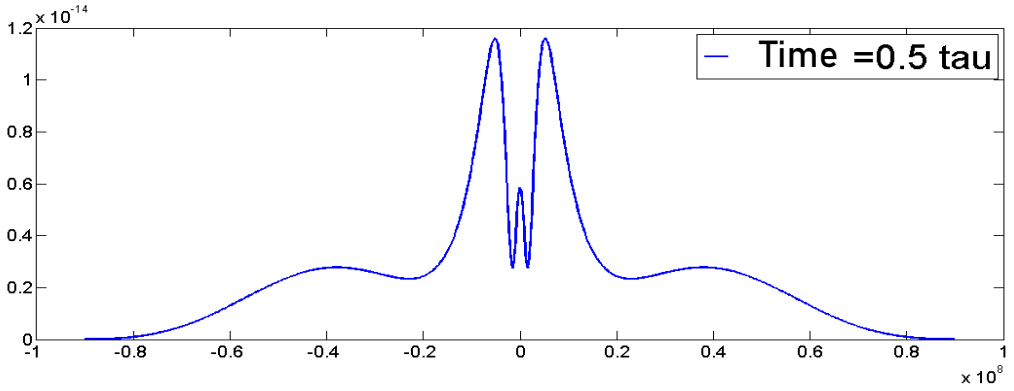
The simulations of the probability density, expressed by equation (16) and Figures 8 to 13, are analysed for several times t, again expressed using the mean lifetime τ.

Although each figure has its own scale, a comparison can be made, as will be explained below.

As time increases, the frequencies and the overall appearance of the scattered photon change drastically. Note that the probability that the radiation has a frequency $\omega_k = \omega_o$ is almost zero for a time close to $t = 0$. For this time, the scale in Figure 8 shows that the absolute value of the frequency distribution function is very small, which of course is due to the fact that the source nucleus is still with a high probability in the excited state so that the probability of finding a gamma photon is still very low. The observed extremum for $\omega_k = \omega_o$ can be understood as the absorption of the present gamma photon (albeit with low probability) by the absorbing nuclei. Thus, frequencies ω_k close to ω_o will be absorbed with a higher probability than others. As time increases, the probability of a gamma photon absorption increases. Indeed, it is observed that, between times $t = 0$ and $t = 3.5\tau$, an increase in the probability density for the radiation to have a frequency $\omega_k = \omega_o$, until it reaches its maximum value.

Figure 8.

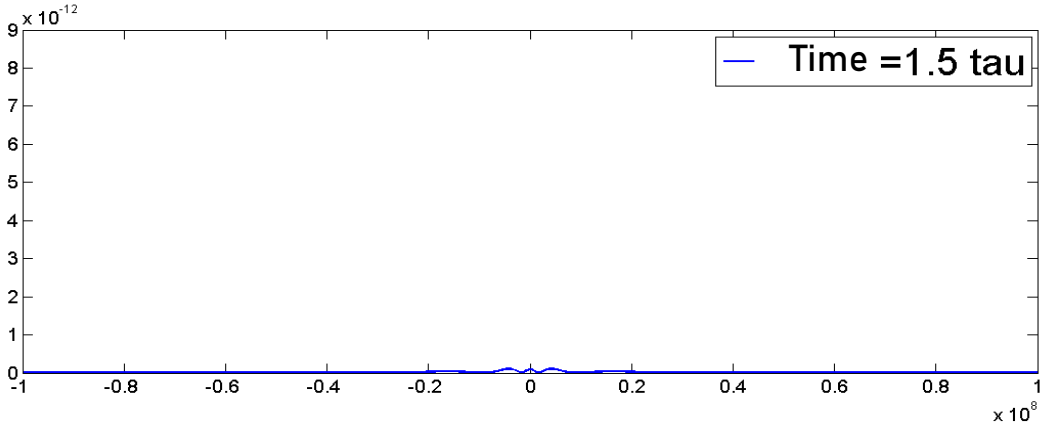
Probability density for $t = 0.5\,\tau$ case of ^{57}Fe in the resonance case



$\omega_k - \omega_o$

Figure 9.

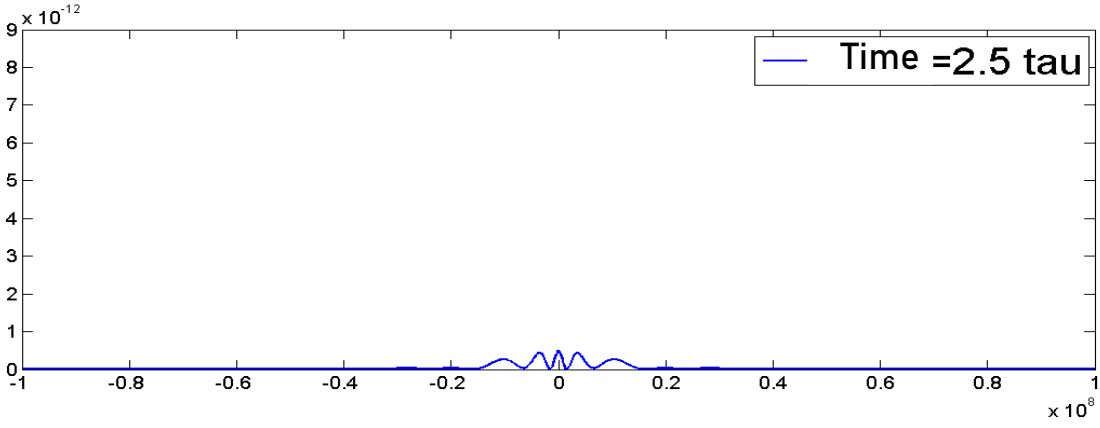
Probability density for $t = 1.5\tau$ case of ^{57}Fe in the resonance case



$\omega_k - \omega_o$

Figure 10.

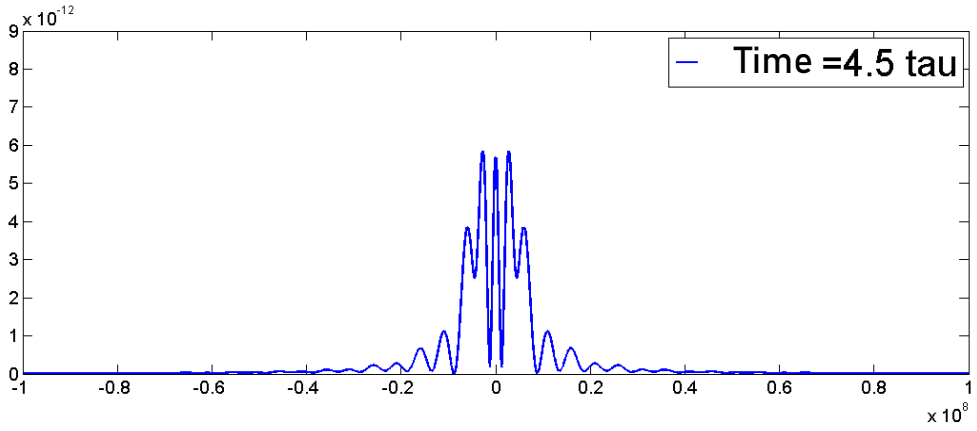
Probability density for $t = 2.5\tau$ case of ^{57}Fe in the resonance case



$\omega_k - \omega_o$

Figure 11.

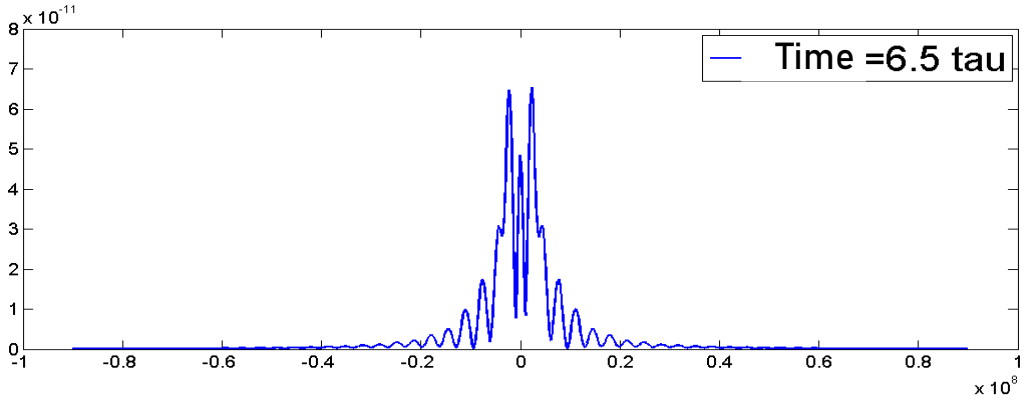
Probability density for $t = 4.5\ \tau$ *case of* ^{57}Fe *in the resonance case*



$\omega_k - \omega_o$

Figure 12.

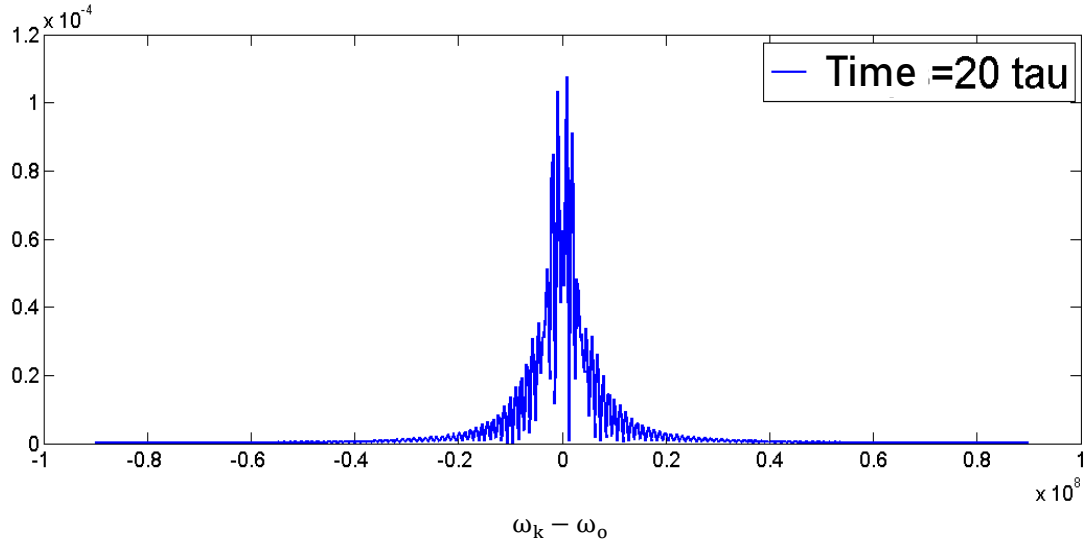
Probability density for $t = 6.5\ \tau$ *case of* ^{57}Fe *in the resonance case*



$\omega_k - \omega_o$

Figure 13.

Probability density for $t = 20\,\tau$ case of ^{57}Fe in the resonance case



For times greater than $t = 3.5\tau$, the probability density of having certain frequencies increases with time to such an extent that the probability of the radiation having a frequency $\omega_k = \omega_o$ is no longer maximum. For very large times, the frequency distribution tends towards the distribution shown in Figure 13; it is not really a Lorentz distribution because of oscillations. Finally, with increasing time, the area bounded by the curve and the horizontal axis continues to increase until it reaches its maximum value for very large t. Figures 10 and 11 confirm this assertion by showing how, with time, this area increases, as they are on the same scale. Furthermore, Figures 8 to 13 allow a better appreciation of the increase of the area bounded by the curve and the horizontal axis. It is worth noting, however, that the details of the extremely rapid oscillations are probably difficult to demonstrate experimentally.

Conclusion

The study included the interaction of gamma radiation, emitted by an excited Mössbauer nucleus, with a polycrystalline absorber containing Mössbauer nuclei in the ground state. The study of the radiation emitted in the resonant nuclear scattering process of two nuclei also reveals that, when the absorber nuclei are not at exact resonance with the source nucleus, the probability of finding a photon with frequency equal to the nuclear frequency, $\omega_k = \omega_o$, is no longer always maximal. Moreover, the area defined by the curve and the horizontal axis increases with time and some frequencies become more probable than others. For long times, the envelope of the distribution looks like a Lorentz curve.

In the case of exact resonance between the source and absorber nuclei, the frequency distribution is symmetric with respect to the value $\omega_k = \omega_o$. From this study, it appears that the sheer presence of the absorber nuclei between the source and the detector means that the photons emitted no longer have the same properties as those emitted when the source is isolated (Hoy & Odeurs, 2012). Note that the frequency distribution is given by a complicated expression, which is impossible to "guess" intuitively: cumbersome calculations are necessary. The presence of the absorber nuclei therefore fundamentally changes the properties of the gamma radiation emitted by the source. These conclusions remain valid for an arbitrary number

of absorbers, *N*, although the expressions become more and more complicated. The model presented here can be used to calculate analytically the properties of the transmitted gamma radiation for any number of absorber nuclei (for any value of the thickness of the absorber), although the simulation of the results will obviously necessitate some tedious computer work.

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