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Nuclear polaritons: Mössbauer source and resonant absorber investigated using the coherent paths model

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Abstract

Resonant nuclei of a Mössbauer absorber, interacting with the recoil free emitted radiation from a Mössbauer source, can re-emit it without recoil, leading to nuclear-resonant scattering. During the nuclear resonance scattering in the Mössbauer absorber, intermediate states which are combinations of nuclear excited states and electromagnetic radiation (gamma radiation) states are produced. These states are called nuclear polaritons. In this paper, a description of the nuclear polariton inside a Mössbauer absorber is presented by adapting the quantum model previously developed by Heitler, Harris and Hoy, called "the coherent paths model". This model allows the calculation of all spatial and temporal properties of the nuclear excited states as well as the electromagnetic radiation present inside a resonant absorber. The thickness of the absorber is modeled using a parameter N. The nuclear polariton results then from the magnetic dipole interaction between the quantified electromagnetic radiation and the resonant nuclei of the absorber. It is an entangled state, composed by the excitation of the nuclei (exciton) and the electromagnetic field. The evolution of the nuclear exciton, both as a function of time and as a function of the position of the resonant nuclei in the absorber, is studied. This constitutes the nuclear part of the polariton. The field associated with the gamma radiation inside the absorber is studied also as a function of position in the absorber and as a function of time, which is the field part of the polariton. Using the purely quantum coherent paths model, we find out that the energy of the polariton inside the absorber oscillates between the nuclear excitation and the field of electromagnetic radiation. Nuclear polariton study is then a potential method for probing matter at the subatomic scale.

Keywords: *Coherent path model; exciton; nuclear polariton; Mössbauer spectroscopy; photon*

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Introduction

Since the discovery of the Mössbauer effect in 1958 (Mössbauer, 1958) , the so-called Mössbauer spectroscopy has been (Greenwood and Gibb, 1971). In this spectroscopy, resonant nuclei absorb and re-emit the radiation without recoil leading to nuclear-resonant scattering (NRS). During the NRS in a Mössbauer absorber, complex intermediate states are formed, containing the nuclear excitation with all resonant nuclei involved and the electromagnetic

radiation. This state is called the nuclear polariton. The collective nuclear state containing all nuclei is called the nuclear exciton.

Nuclear polaritons in Mössbauer spectroscopy have aroused the interest of researchers since it was highlighted by M. Haas (Haas, 2007; Haas *et al.*, 1988) and, following him, by other researchers (Smirnov *et al.*, 2005, 2007) .

In this paper, we propose a description based on the quantum model of the so called coherent paths, which was developed by Hoy (Harris, 1961; Heitler, 1984; Hoy, 1997). The model has been successfully used to study NRS in the forward direction for various cases (Hoy, 1997, 2001; Hoy *et al.*, 2001; Odeurs and Hoy, 2007).

The model is based on a detailed microscopic description to study the interaction of recoil-free gamma radiation with resonant nuclei embedded in a solid state material. Other theoretical approaches, such as the semi-classical optical model (Kagan *et al.*, 1979), have been used to study the same physical system. It has been shown (Hoy and Odeurs, 2009) that the coherent path model leads to the same numerical results as the semi-classical optical model for a synchrotron radiation excited absorber. For the radioactive case, Harris (Harris, 1961) showed that the results of the coherent path model agree with those of the semi-classical optical model (Hoy *et al.*, 2001) Although the two models are completely different, they lead to equivalent results, meaning that either one could be used to analyse experimental data.

Materials and methods

Nuclear resonant forward scattering

Schrödinger equation for the complete system

The evolution of the global system consisting of a source nucleus, resonant absorber nuclei and gamma radiation will be investigated. The hamiltonian of this system is:

$$H = H_n + H_c + H_{DM} \qquad (1)$$

where H_n corresponds to the nuclear states, H_c to the gamma radiation field and H_{DM} represents the interaction between the nuclei and the field.

The hamiltonian is divided into: H_0 corresponding to the nuclear states and the free radiation field taken as plane waves and H_{DM} which is responsible for the transitions between eigenstates $|\phi\rangle$ of H_0 allowing nuclei to absorb or emit a radiation. So, we have: $H = H_0 + H_{DM}$.

The state $|\psi(t)\rangle$ of the system can be expressed as follows:

$$|\psi(t)\rangle = \sum_m a_m\,(t) e^{-\frac{iE_m t}{\hbar}} |\phi_m\rangle \qquad (2)$$

where $|\phi_m\rangle$ is an eigenstate of the undisturbed system.

We have to solve the time-dependent Schrödinger equation:

$$(H_0 + H_{DM})|\psi(t)\rangle = i\hbar \frac{d}{dt}|\psi(t)\rangle \qquad (3)$$

Substituting $|\psi(t)\rangle$ by its expression given by (2) in (3) and considering that

$H_0|\phi_m\rangle = E_m|\phi_m\rangle$, we have:

$$i\hbar\left(\sum_m \frac{da_m(t)}{dt} e^{-i\left(E_m\frac{t}{\hbar}\right)} |\phi_m\rangle\right) = \sum_m a_m(t)\, e^{-i\left(E_m\frac{t}{\hbar}\right)} H_{DM}|\phi_m\rangle \qquad (4)$$

Projecting this equation on any eigenstate, say $|\phi_p\rangle$, of H_0 we have:

$$i\hbar \frac{da_p(t)}{dt} = \sum_m a_m(t)\, e^{i(\omega_p - \omega_m)t} \langle\phi_p|H_{DM}|\phi_m\rangle \qquad (5)$$

where $\omega_p = \frac{E_p}{\hbar}$ and $\omega_m = \frac{E_m}{\hbar}$.

We will specify now the initial condition of the problem. By experimental approach, the solution $a_p(t)$ of this equation has a physical meaning only for $t \geq 0$. For analytical reasons, we extend the solution of the system of coupled equation (5) to the negative time axis by adding an inhomogeneous term that takes into account the jump of $a_\ell(t)$ (we associate the index ℓ to the eigenvalue of the system at t = 0). The solution of the equation for the amplitude associated with the initial state jumps abruptly from 0 to 1. So, in the vicinity of $t = 0$, $\frac{da_\ell(t)}{dt}$ behaves like a delta function, $\delta(t)$. This jump is added to the expression (5) to give the equation below:

$$i\hbar\frac{da_p(t)}{dt} = \sum_m a_m(t)\, e^{i(\omega_p-\omega_m)t}\langle\phi_p|H_{DM}|\phi_m\rangle + i\hbar\delta_{p\ell}\delta(t) \quad (6)$$

From (6) follows that a_p ($p \neq \ell$) is continuous at t = 0, considering the fact that only a_ℓ makes a jump.

To solve (6) we introduce a Fourier transformation as defined in references (Heitler, 1984; Hoy, 1997).

After some calculations we have:

$$(\omega-\omega_p)A_p(\omega) = \sum_m A_m(\omega)\frac{\langle\phi_p(0)|H_{DM}|\phi_m(0)\rangle}{\hbar} + \delta_{pl} \quad (7)$$

Dividing expression (7) by $(\omega-\omega_p)$, we have a discontinuity in the right-hand side for $\omega = \omega_p$. So, we have a pole in the complex plane of ω. However, in order to have $a_p(t) = 0$ for all negative values of t, $A_p(\omega)$ must have a pole only in the lower half of the complex plane, this to guarantee the causality (Heitler, 1984). To ensure this, (7) is rewritten as follows:

$$(\omega-\omega_p+i\varepsilon)A_p(\omega) = \sum_m A_m(\omega)\frac{\langle\phi_p(0)|H_{DM}|\phi_m(0)\rangle}{\hbar} + \delta_{pl} \quad (9)$$

This system of equations will be specified in section 2.3 to study the nuclear scattering problem. The number of effective nuclei, N, is related to the thickness parameter $\beta = N_0 f \sigma_0 d$ (Hoy, 1997) where N_0 is the number of resonant nuclei per volume unit, f is the recoilless fraction, σ_0 is the maximum cross section evaluated at resonance and d the thickness of the sample. N and β are related by : $N = \frac{1}{2}\frac{\beta}{f}\frac{\Gamma}{\Gamma_r}$ (Hoy, 1997) where Γ and Γ_r are respectively the total line width and radiative width of the excited state.

Fundamental coupled equations

Consider ^{57}Fe in the first excited state at 14.4 keV above the ground state. The time t = 0 is defined by the formation of the state of 14.4 keV, which can be determined precisely by the detection of the 122 keV precursor. The 14.4 keV level of ^{57}Fe can decay according to two different ways: gamma photon production or electron conversion, where the nuclear energy of the excited state is transferred directly to an atomic electron. For the 14.4 keV transition in the ^{57}Fe, the electron conversion process is more or less nine times more likely than the gamma transition (Greenwood and Gibb, 1971).

At t = 0, there is an excited source nucleus, all the resonant absorber nuclei are in the ground state and there is no gamma radiation present. We study the scattering problem for these nuclei by restricting the calculations to the forward scattering. Five amplitudes as defined in (Hoy, 1997): $A(\omega)$, $B_k(\omega)$, $C_m(\omega)$, $D_p(\omega$, $E_{mp}(\omega)$ will be considered.

Assuming that at time t = 0, the source nucleus is excited, and substituting these amplitudes into (9) gives the following set of coupled linear equations (Hoy, 1997):

$$(\omega-\omega_0+i\varepsilon)A(\omega) = 1 + \sum_k \frac{B_k(\omega)H_k}{\hbar} + \sum_p \frac{D_p(\omega)H_p}{\hbar} \quad (10)$$

$$(\omega-\omega_k+i\varepsilon)B_k(\omega) = \frac{A(\omega)H_k^*}{\hbar} + \sum_m \frac{C_m(\omega)H_k^*}{\hbar}e^{-ikx_m} \quad (11)$$

$$(\omega-\omega_0'+i\varepsilon)C_m(\omega) = \sum_m \frac{B_k(\omega)H_k}{\hbar}e^{ikx_m} + \sum_p \frac{E_{mp}(\omega)H_p}{\hbar}e^{i(\frac{p}{\hbar})x_m} \quad (12)$$

$$(\omega-\omega_p+i\varepsilon)D_p(\omega) = \frac{A(\omega)H_p^*}{\hbar} \quad (13)$$

$$(\omega-\omega_p+i\varepsilon)E_{mp}(\omega) = \frac{C_m(\omega)H_p^*}{\hbar}e^{-i(\frac{p}{\hbar})x_m} \quad (14)$$

where H_k and H_k^* are the matrix elements corresponding to absorption and emission of a photon, respectively. Analogously, H_p and H_p^* are the matrix elements corresponding to absorption and emission of a conversion electron, respectively.

The solution of the scattering problem is obtained by solving this set of coupled linear equations (Hoy, 1997). For $A(\omega)$, we have:

$$A(\omega) = \frac{1}{\omega - \omega_0 + i\frac{\Gamma}{2\hbar}} \quad (15)$$

where $\frac{\Gamma}{2\hbar}$ is the total width characterising the lorentzian distribution of the excited-level energy of 14.4 keV. All other amplitudes can be found in (Hoy, 1997). These coefficients will be utilized to study the excitation of the nuclei which constitutes the nuclear part of the polariton. For $C_m(\omega)$, we have:

$$C_m(\omega) = -i\frac{\Gamma_r}{2\hbar}\frac{e^{i\left(\frac{\omega}{c}\right)x_m}A(\omega)}{\left(\omega - \omega_0' + i\frac{\Gamma}{2\hbar}\right)} \times \left[1 - i\frac{\Gamma_r}{2\hbar}\frac{1}{\omega - \omega_0' + i\frac{\Gamma}{2\hbar}}\right]^{m-1} \quad (16)$$

where Γ_r is the radiative width. For $B_k(\omega)$, we have:

$$B_k(\omega) = \frac{H_k^*}{\hbar}\frac{1}{\left(\omega - \omega_0 + i\frac{\Gamma}{2\hbar}\right)(\omega - \omega_k + i\varepsilon)} \times \left[1 + \sum_{m=0}^{N-1} e^{i\left(\frac{\omega}{c} - k\right)x_{m+1}}\alpha(\alpha+1)^m\right] \quad 17)$$

where $\alpha = -i\frac{\Gamma_r}{2\hbar}\frac{1}{\left(\omega - \omega_0' + i\frac{\Gamma}{2\hbar}\right)}$

Before using the Fourier transform to go back to the time domain, we can express $B_k(\omega)$ in a symbolic form by examining the equation (17). We have:

$$B_k(\omega) = B_k^{source}(\omega) + \sum_{m=0}^{N-1} B_{k,m}^{absorber}(\omega) \quad (18)$$

The first term is due to the source alone while the summation term describes the effect due to each absorber nucleus according to the value of *m* which does not only indicate the position of the nucleus considered in the absorber but which also takes into account the effect of the nuclei *in front* of the nucleus situated at $x = x_m$. By Fourier transform, we have:

$$b_k(t) = -\frac{1}{2\pi i}\int_{-\infty}^{+\infty} B_k(\omega)e^{i(\omega_k - \omega)t}d\omega \quad (19)$$

which formally gives, using expression (18):

$$b_k(t) = b_k^{source}(t) + \sum_{m=0}^{N-1} b_{k,m}^{absorber}(t) \quad (20)$$

This amplitude allows us to find the expression of the field of gamma radiation inside the absorber.

Amplitude of the radiation field for recoil-free processes

Using lengthy calculations, Hoy (Hoy, 1997) managed to find the amplitude of the transmitted radiation. To do so, Hoy considered the radiation as a superposition of plane waves, each one with a coefficient $b_k(t)$, which can be calculated according to the lines explained above.

At resonance, $\omega_0 = \omega_0'$, the total amplitude of the radiation leaving the absorber is given by the expression:

$$\psi_r(t') = \psi_r^{source}(t')\left[1 + \sum_{n=1}^{N}\binom{N}{n}\left(-\frac{\Gamma_r t'}{2\hbar}\right)^n\frac{1}{n!}\right] \quad (21)$$

where $\psi_r^{source}(t') = e^{-i\left(\omega_0 - i\frac{\Gamma}{2\hbar}\right)t'}$ and t' is the time measured from the moment of the formation of the first excited level in the source ($t' = t - \frac{x}{c}$). x is a position behind the absorber.

If only a part of the absorptions and reemissions of the photons occurs without recoil, expression (21) will have to be modified. Only for the processes without recoil, it is impossible to distinguish the path followed by each photon towards the detector. Thus all the paths must be added in a coherent way: all the amplitudes must be added to have the amplitude of the radiation which reaches the detector. Therefore, we multiply the right-hand side of the equation (21) by $\frac{\Gamma_r}{2}$ and by the recoil-free fraction f.

So the amplitude of the radiation field reaching the detector is:

$$\psi_r(t') = \sqrt{\frac{f\Gamma_r}{2\hbar}}e^{-i(\omega_0 - i\frac{\Gamma}{2\hbar})t'}\left[1 + \sum_{n=1}^{N}\binom{N}{n}\left(-\frac{f\Gamma_r t'}{2\hbar}\right)^n\frac{1}{n!}\right] \quad (22)$$

Results

Nuclear polaritons

Temporal and spatial behaviour of the nuclear part of the polariton

To study the excitation of nuclei, we start from the expression (16) of $C_m(\omega)$. We use the Fourier transformation to have this amplitude of excitation of the nuclei as a function of time. At resonance, $\omega_0 = \omega_0'$ and considering the recoil free fraction f as in equation (22), it can be shown that $C_m(\omega)$ can be written as:

$$C_m(\omega) = \sum_{n=0}^{m-1} \left(-i\frac{f\Gamma_r}{2\hbar}\right)^{n+1} \binom{m-1}{n} \frac{e^{i(\frac{\omega}{c})x_m}}{\left(\omega - \omega_0 + i\frac{\Gamma}{2\hbar}\right)^{n+2}} \quad (23)$$

It can be shown that:

$$C_m(x_m,t) = \sum_{n=0}^{m-1} \binom{m-1}{n} (-1)^{n+1} \left(\frac{f\Gamma_r}{2\hbar}\right)^{n+1} \frac{1}{(n+1)!} \left[i\left(\frac{x_m}{c} - t\right)\right]^{n+1} e^{i\left[\left(\frac{x_m}{c}-t\right)\left(\omega_0 - i\frac{\Gamma}{2\hbar}\right)+\omega_m t\right]} \quad (24)$$

where we have added the apparently explicit dependence on x_m, the position of the nucleus m.

By taking its module, neglecting x_m/c and by considering $e^{x_m\Gamma/2c\hbar} = 1$ we have:

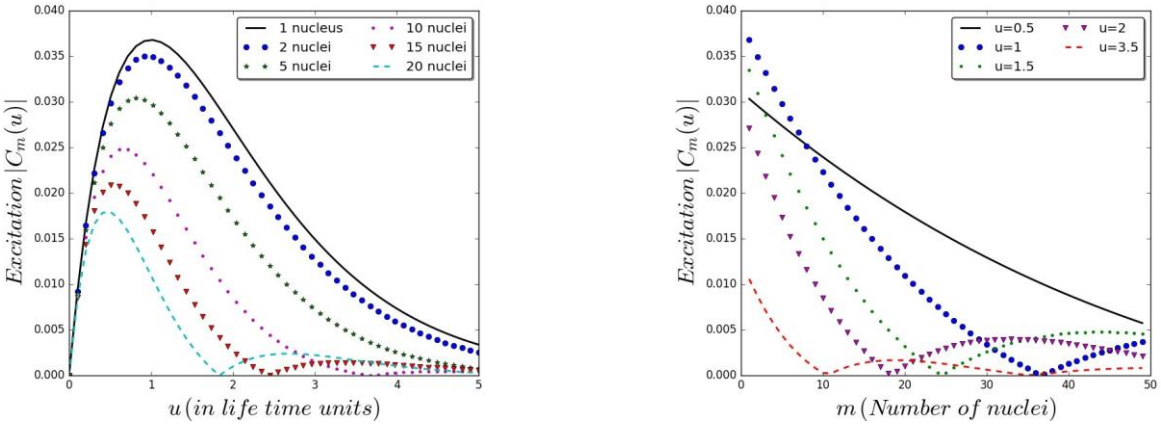
$$|C_m(t)| = \left|\sum_{n=0}^{m-1} \binom{m-1}{n} (-1)^{2(n+1)} \left(\frac{f\Gamma_r}{2\hbar}\right)^{n+1} \times \frac{1}{(n+1)!} t^{n+1}\right| e^{-t\frac{\Gamma}{2\hbar}} \quad (25)$$

so that there is no explicit dependence of x_m anymore.

Introducing $\frac{2\hbar}{\Gamma} = \tau$, $\frac{t}{\tau} = u$, $\Gamma_r = \frac{\Gamma}{10}$ equation (26) becomes:

$$|C_m(u)| = \left|\sum_{n=0}^{m-1} \binom{m-1}{n} (-1)^{n+1} \frac{1}{(n+1)!} \left(\frac{fu}{10}\right)^{n+1}\right| e^{-u} \quad (26)$$

This expression informs us about the excitation of the nuclei as a function of time and implicitly the position of the nucleus m. In figure 2, we have chosen $f = 1$.



(a) (b)

Figure 1. (a) Amplitude of probability of excitation of 1 (solid line), 2 (filled circles),5 (stars symbols), 10 (dotted), 15 (filled triangles) and 20 (discontinuous lines)) nucleias a function of time. (b) Amplitude of excitation of the nuclei as a function of the position in the absorber for u equal to 0.5 (solid line), 1 (filled circles), 1.5 (dotted line), 2 (filled rectangles) and 3.5 (discontinuous line).

Temporal and spatial behaviour of the field part of the polariton

Let us examine the amplitude of the radiation field as a function of time and position at resonance. From the expression

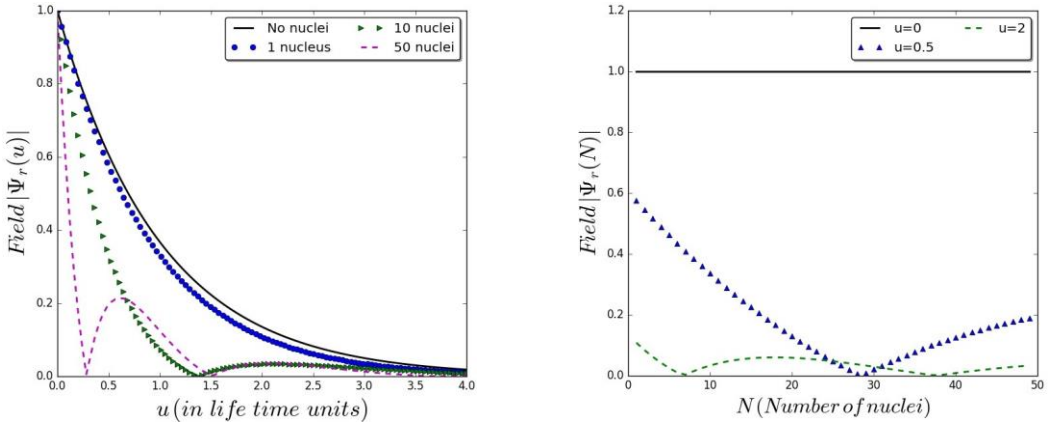
(22), neglecting the normalization terms, the amplitude of the field is:

$$\psi_r(t') = e^{-i(\omega_0 - i\frac{\Gamma}{2\hbar})t'}\left[1 + \sum_{n=1}^{N}\binom{N}{n}\left(-f\frac{\Gamma_r t'}{2\hbar}\right)^n \frac{1}{n!}\right] \quad (27)$$

In terms of u, we end up with the expression (28), which gives the amplitude of thefield at a position between the absorber and the detector:

$$|\psi_r(u)| = \left|\left[1 + \sum_{n=1}^{N}\binom{N}{n}\left(-\frac{fu}{10}\right)^n \frac{1}{n!}\right]\right| e^{-u} \quad (28)$$

This equation will be used to study the field inside the absorber by varying N which is related to the thickness of the absorber. If we take $N = n$, we would have the field at the position of the n^{th} nucleus in the absorber.We utilize then the expression to calculate the field amplitude everywhere inside the absorber.



(a) (b)

Figure 2. (a) Comparison of the amplitudes of the field as a function of time (u)for no nuclei (solid line), 1 nucleus (filled circles), ten nuclei (filled triangles) and fiftynuclei (discontinuous line), (b) Amplitude of the field as a function of the position inthe absorber for u = 0 (solid line), u = 0.5 (filled triangles), u = 2 (discontinuous line).

Temporal and spatial behaviour of the polariton

In the figures 3 and 4, we show the excitation amplitude of the nuclei as a function of time as well as the amplitude of the field as a function of time (figure 3) and as a function of the position (figure 4).

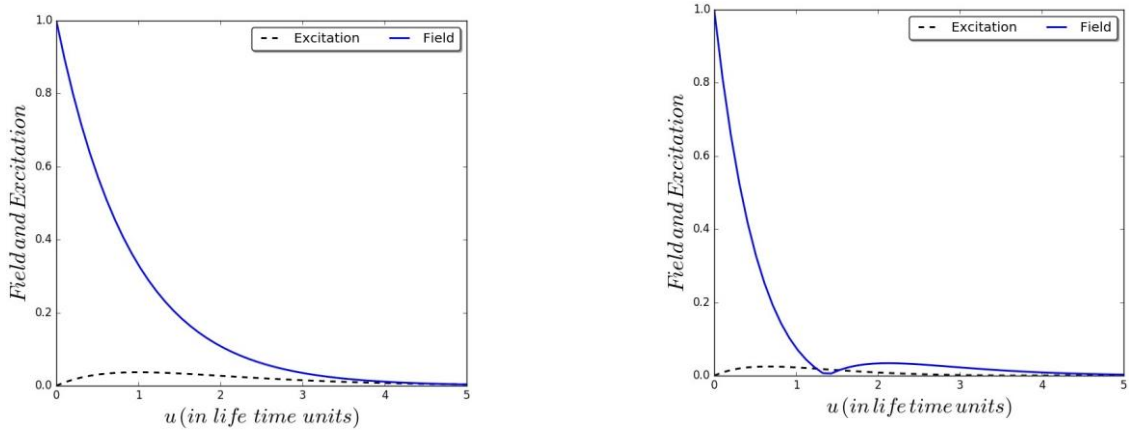
Discussion

Figure 1(a) illustrates the excitation amplitude of the nuclei as a function of time, the time being varied by fixing m (the position) for 1, 2, 5, 10, 15 and 20 nuclei. We notice that for 10, 15 and 20 nuclei we have a discontinuity of the derivative because of the absolute value. This discontinuity has no physical meaning but the function is continuous. The figure shows that the excitation is maximum between u = 0.5 and u = 1.5 for all the numbers of nuclei considered. Figure 1(b) represents the excitation amplitude of the nuclei as a function of the position in the absorber. The time (u = 0.5, 1, 1.5, 2 and 3 units) is fixed while the position varies. For an increasing number of nuclei, the excitation decreases for small values of u.

Figure 2(a) shows the amplitude of the field versus the time for no nucleus, 1, 10, 15 and 50 nuclei; whereas figure 2(b) represents the amplitude of the field versus the position for u = 0, u = 0.5 and u = 2. The field strength at a particular nucleus depends on the number of nuclei in front of it.

For u = 0, all absorber nuclei have a probability amplitude equal to 1 of being excited. In reality it is $1/N$ but it has been normalized to 1. For a time u < 1, there is a decrease as shown in the figure 2(b) when u = 0.5. For a long time (u > 1), the amplitude decreases rapidly and increases again

if we progress in the absorber because of the photons coming from the decaying of nuclei in the absorber. This is clearer in the same figure 2(b) for $u = 2$.



(a) (b)

Figure 3. (a) Polariton as a function of time for a single absorber nucleus (b) Polariton as a function of time for N = 10.

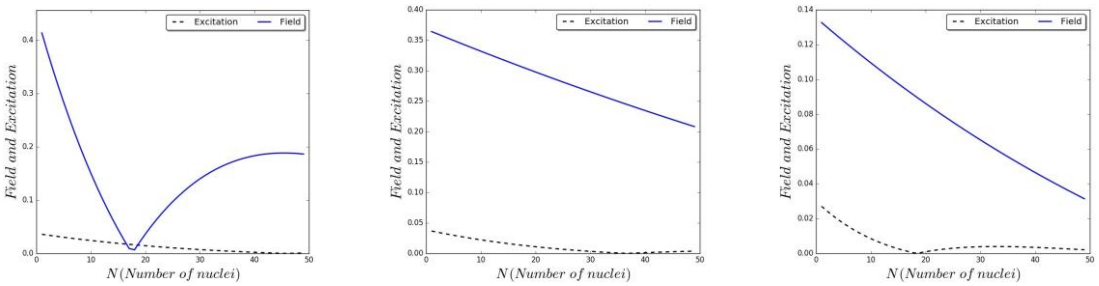
Figures 1 and 2 show two effects: the so-called *speed-up* and the *dynamical beats*. For a high number of resonant nuclei there is a decay of the gamma radiation wave function that is faster than simple exponential decay. This phenomenon is called speed-up (Hoy, 2001). For a high number of resonant nuclei, the gamma radiation wave function shows also an oscillatory time behaviour. This effect is called dynamical beats (Hoy, 2001). The coherent-path model shows that the *speed-up* and *dynamical beating* effects are a direct consequence of the interference of all quantum mechanical amplitudes involved in the multiple scattering processes. In the expression (28) there is a factor e^{-u}, which represents simple exponential decay. In the square brackets there is a polynomial of t', with a succession of + and – signs. It is shown in (Hoy, 2001) that this succession of terms with positive and negative signs can explain all details of the speed-up and the dynamical beats effects.

Dependence on time

Figure 3(a) represents polaritons as a function of time for one absorber nucleus. The amplitude of the field decreases (solid line) while the amplitude of excitation (dashed line) increases for $u < 1$, reaches its maximum at $u \approx 1$ and decreases then slowly. Figure 3(b) represents the polariton as a function of time for 10 nuclei. If we compare the two cases (1 nucleus and 10 nuclei) of the polariton as a function of time, we see that its nuclear excitation part is less pronounced for 10 nuclei compared with that of the case of a nucleus.

Dependence on the position

The dashed lines of figure 4 represent the amplitude of the nuclear excitation, the solid line represents the amplitude of the field. Figure 4(a) represents the polariton behaviour depending on the position for $u = 0.8$; Figure 4(b) represents the polariton behaviour depending on the position for $u = 1$ while Figures 4(c) represents the polariton behaviour depending on the position for $u = 2$.



(a) (b) (c)

Figure 4. (a) Polariton depending on the position for $u = 0.8$ (b) Polaritondepending on the position for $u = 1$ (c) Polariton depending on the position for $u = 2$.

Conclusion

The radiation emitted by a Mössbauer source interacts with N effective Mössbauer nuclei in a resonant absorber before leaving it. Solving the forward scattering problem by the coherent path model allowed us to describe the properties of the resulting nuclear polariton. The time evolution of the excitation amplitude of the nuclei in the absorber as well as the time and space evolution of the radiation field inside the absorber have been investigated. The polariton *nucleus* + *radiation field* inside the absorber oscillates between the nuclear excitation and the field of electromagnetic radiation. The details of these behaviours can be understood as interference between all quantum mechanical amplitudes relative to all possible scattering events in the forward direction.

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