Quantum mechanical theory of the interference pattern of reference and object waves using scattering processes without and with recoil: Nuclear emission holography

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(Received 22 November 2004; revised manuscript received 7 July 2005; published 24 August 2005)

The interference pattern eventually leading to nuclear emission holography results from the interference of radiation, produced by an excited source nucleus, that goes directly to a detector and radiation that is first resonantly scattered by neighboring identical nuclei in the ground state before going to the detector. The interference between these two processes gives rise to fluctuations in the radiated intensity as a function of the emission angle, giving information about the surrounding of the emitting nucleus. The quantum mechanical theory of the interference pattern using γ-radiation is developed in frequency domain. It is shown that if the wavelengths of the relevant vibrational modes, which are excited due to the recoil of nuclei, are large compared with the relative distances of the absorber nuclei, with respect to the source nucleus, processes without as well as with recoil can give a contribution to the interference pattern. For nuclei that are situated at large distances from the source nucleus, only processes without recoil have to be considered.

DOI: 10.1103/PhysRevB.72.064119 PACS number(s): 76.80.+y, 61.18.−j

I. INTRODUCTION

Probably the earliest articles mentioning the possibility of the determination of chemical structures using gamma radiation go back to the early 1970's. Hannon and coworkers used already the term holography in their papers,[1] dealing with the description of the emission of γ rays from crystals based on covariant quantum electrodynamics.[2]

Internal source holography[3] was proposed as a tool for three-dimensional imaging of atomic structures. It was performed with electrons,[4] and later with x rays,[5] γ rays,[6] and neutrons.[7]

In the implementation of γ-ray holography, the Mössbauer effect and the internal detector principle of holography have been utilized.[6,8,9] Simulations[10,11] and subsequently experiments[12] were performed to study three-dimensional imaging of local iron arrangements in sublattices of magnetite.

Recent reviews of x-ray and γ-ray holography can be found in Refs. 13 and 14. Internal source holography using γ radiation is based on the interference between two processes. The first process corresponds to the emission of a photon by a radioactive nucleus, embedded in a solid-state lattice, going directly to a far-field detector. This is the reference wave. The second process corresponds to radiation produced by resonant scattering of the emitted photon by neighboring nuclei, before it goes to the detector. This is the object wave. This version of holography has not been realized so far. In Ref. 15, an expression for the interference pattern of the reference and object waves was calculated using a fully quantum mechanical approach. In the analysis presented in Ref. 15, it was assumed that emission and scattering occur without recoil, i.e., the Mössbauer effect. If the state of the lattice would be taken into account, the Mössbauer-Lamb factor would appear naturally in the theory.

In the present paper, the influence of the lattice on the formation of the interference pattern will be investigated. The expressions of Ref. 15 will be generalized by including the lattice terms. Furthermore, it will be shown that if the wavelengths associated to the relevant vibrational modes, excited because of the recoiling nucleus, are large compared to the relative distance of the absorber nuclei with respect to the source nucleus, there will be a contribution to the interference pattern due to processes with recoil. The interpretation of this would be that the vibrational effects because of the recoil by the source nucleus are the same for all absorber nuclei situated in the vicinity of the source nucleus. This situation would only be realized for nuclei in the immediate surrounding (at distances of the order of a few ångstroms) of the source nucleus, which are precisely the nuclei that are considered for the construction of the hologram.

With our scheme, an interference pattern could be obtained using a Mössbauer isotope incorporated in a sample having a recoilless fraction that is small because of, e.g., the sample being at high temperature. Also γ radiation with higher energies than those utilized in Mössbauer spectroscopy could be used, although it should be mentioned that the nuclear scattering cross section decreases with increasing energy.

As has been mentioned before, a hologram results from the interference between a well-defined reference wave and various components of this wave that are scattered by the object to be imaged. The main problem arising in the reconstruction of the image is the presence of twin images,[16] which cause the disappearance of images in real-space reconstruction. Recently,[17] a solution to twin images problem in γ-ray holography has been given. If interference patterns using multiple γ-ray energies would become possible, they could be used to suppress twin image effects as well as other aberrations and artifacts in reconstructed images.[18]

II. ESTABLISHMENT OF THE EQUATIONS

A. Mathematical formalism

The method discussed in this paper makes use of the Schrödinger equation in frequency domain.[19–22] This has the advantage of transforming coupled differential equations for the coefficients into a set of linear algebraic equations.

If a quantum mechanical system is described by a Hamiltonian H, which is the sum of an unperturbed part H_0 and a perturbation V, the actual state of the system can be developed into eigenstates $|\varphi_l(0)\rangle$, of H_0

$$|\psi(t)\rangle = \sum_l a_l(t)e^{-iE_lt/\hbar}|\varphi_l(0)\rangle \quad (1)$$

with

$$H_0|\varphi_l(0)\rangle = E_l|\varphi_l(0)\rangle. \quad (2)$$

Substituting Eq. (1) into the Schrödinger equation leads to the well-known coupled differential equations

$$i\hbar\frac{da_l}{dt} = \sum_q a_q(t)e^{i(\omega_l-\omega_q)t}\langle\varphi_l(0)|V|\varphi_q(0)\rangle, \quad (3)$$

where $\omega_l-\omega_q=(E_l-E_q)/\hbar$.

Generally, the system is initially in a well-defined eigenstate of H_0, say $|\phi_n\rangle$. This means that $a_l(0)=0$ and $a_n(+0)=1$, where $t=+0$ means that t approaches zero from the positive side.

Following Heitler,[19] the solution will be extended to the negative time axis. All a_l's will be chosen such that $a_l(t)=a_n(t)=0$ for $t<0$. Heitler showed that adding an inhomogeneous term to the right-hand side of expression (3) takes into account the initial condition and the discontinuity of $a_n(t)$ at $t=0$. One has

$$i\hbar\frac{da_l}{dt} = \sum_q a_q(t)e^{i(\omega_l-\omega_q)t}V_{lq} + i\hbar\delta_{\ln}\delta(t), \quad (4)$$

where

$$V_{lq} = \langle\varphi_l(0)|V|\varphi_q(0)\rangle \quad (5)$$

$\delta_{\ln}$ is the Kronecker delta, which takes into account the initial condition, and $\delta(t)$ the Dirac delta function, which correctly describes the discontinuity at $t=0$.

Again following Heitler,[19] a particular type of Fourier transform is introduced

$$a_l(t) = -\frac{1}{2\pi i}\int_{-\infty}^{\infty} d\omega A_l(\omega)e^{i(\omega_l-\omega)t}. \quad (6)$$

$A_l(\omega)$ is the amplitude in frequency domain. It can be shown that one has now a set of coupled linear equations relating these amplitudes.

$$(\omega-\omega_l+i\varepsilon)A_l(\omega) = \sum_q A_q(\omega)\frac{V_{lq}}{\hbar} + \delta_{\ln}. \quad (7)$$

The infinitesimal positive quantity ε has been introduced to guarantee causality when going back to time again. This general formalism will be applied to the study of the interactions of electromagnetic radiation with nuclei embedded in a solid-state lattice, taking into account the state of the lattice.

B. Base states

As the initial state we consider an excited nucleus, the "source nucleus," surrounded by identical ground state nuclei m, the "scattering" nuclei, with no photons or conversion electrons present. Initially, the lattice is described by a state $\{\alpha_s\}$, which gives the phonon occupations of the different phonon modes s. The source nucleus is at the origin of a coordinate system, the scattering nucleus m is at $\vec{r}_m$. To simplify the notations and the equations a bit, we will not take into account the conversion electrons, although the complete analysis could be done including them. We will show later how the expressions are modified if the conversion electron processes are taken into account. The initial state of the global system nuclei-radiation field-solid-state lattice can be written as

$$|\Phi_{\{\alpha_s\}}\rangle = |e_0,\{g_m\},\{0\},\{\alpha_s\}\rangle. \quad (8)$$

Although the notation looks cumbersome, it should be straightforward: The first symbol inside the ket of the right-hand side of Eq. (8) represents the source nucleus excited, the second symbol represents all scattering nuclei in the ground state, the third symbol represents the vacuum of the electromagnetic radiation field (no photons in any of the field modes), and, finally, the fourth symbol represents the lattice expressed in terms of phonon occupations of the vibrational modes s. $|\Phi_{\{\alpha_s\}}\rangle$ is the base state from which the system will evolve. At finite temperatures, the state of the lattice is not unique and, rather than having a unique lattice state $|\{\alpha_s\}\rangle$, one has a statistical mixture of lattice states, described by a Boltzmann distribution $g(\{\alpha_s\})$, which gives the occupation of the phonon modes s. It will be shown further how this will affect the calculation. Other base states can be considered. There are "intermediate" states where nucleus m is excited, all other nuclei in the ground state, no photons present and the lattice in the same state as before.

$$|\Phi_{m,\{\alpha_s\}}\rangle = |g_0,e_m,\{g_{m'\neq m}\},\{0\},\{\alpha_s\}\rangle, \quad (9)$$

where $g_0,e_m,\{g_{m'\neq m}\}$ means that the source nucleus is in the ground state, nucleus m excited and all other scattering nuclei in the ground state. One need not consider states where there is a different lattice state, say, $\{n_s\}\neq\{\alpha_s\}$, because we do not consider a transition without resonant character. The nonresonant character would imply a negligible amplitude for this transition. This can be shown after a straightforward albeit lengthy calculation.

States corresponding to the presence of a photon having wave vector $\vec{k}$, all nuclei in the ground state and the lattice in a state $\{\beta_s\}$ should also be considered. There are a great number of these states, because of the many possibilities for $\{\beta_s\}$. They are denoted

$$|\Phi_{\vec{k},\{\beta_s\}}\rangle = |g_0,\{g_m\},\vec{k},\{\beta_s\}\rangle. \tag{10}$$

The interpretation of expression (10) should be obvious. The first symbol inside the ket of the right-hand side of expression (10) represents the source nucleus in the ground state, the second symbol represents all absorber nuclei in the ground state, the third symbol represents the photon with wave vector $\vec{k}$, and the fourth symbol the lattice state. By allowing these base states, it is implicitly allowed, among other things, that creation of phonons is possible due to emission by the source nucleus and that these phonons can be absorbed again by nucleus m, so that it can be excited. In other words, we allow for nuclear resonant scattering with phonon creation and annihilation. Strictly speaking, we should have characterized the photon state by a second subscript, describing the photon polarization. This would be necessary if the nuclear system is submitted to hyperfine interactions. In this paper we will not consider this for simplicity.

The state vector of the global system can be developed as a function of the base states, which are eigenstates of the unperturbed Hamiltonian H_0. We have not given the explicit expression of H_0. It is a lengthy expression, which is the sum of the Hamiltonians of the "free" nucleus, the "free" radiation field, and the "free" solid-state lattice.

The coefficients corresponding to each base state are time dependent functions. Applying the formalism sketched in Sec. II A, amplitudes in frequency domain can be assigned to these coefficients. In the next section the fundamental equations in frequency domain will be established.

C. Fundamental equations

Specializing the formalism of Sec. II A to the global system nucleus-radiation field-solid-state lattice, one has the following set of coupled equations relating the amplitudes in frequency domain:

$$(\omega - \omega_{\vec{k}} + i\varepsilon)C_{\vec{k},\{\beta_s\}}(\omega) = V^*_{\vec{k},\{\beta_s\}}A_{\{\alpha_s\}}(\omega) + \sum_m V^*_{m,\vec{k},\{\beta_s\}}B_{m,\{\alpha_s\}}(\omega), \tag{11}$$

$$(\omega - \omega_0 + i\varepsilon)A_{\{\alpha_s\}}(\omega) = \sum_{\vec{k}}\sum_{\{\beta_s\}} V_{\vec{k},\{\beta_s\}}C_{\vec{k},\{\beta_s\}}(\omega) + 1, \tag{12}$$

$$(\omega - \omega_0 + i\varepsilon)B_{m,\{\alpha_s\}}(\omega) = \sum_{\vec{k}}\sum_{\{\beta_s\}} V_{m,\vec{k},\{\beta_s\}}C_{\vec{k},\{\beta_s\}}(\omega). \tag{13}$$

We have incorporated $\hbar$ [see expression (7)] into the coupling constants $V_{\vec{k},\{\beta_s\}}$ and $V_{m,\vec{k},\{\beta_s\}}$ and their complex conjugate. In expression (11), we have assumed that the source nucleus and the scattering nuclei experience the same phonon ensemble, which is of course defined by the state of the lattice in which the nuclei are incorporated, hence the same subscript $\{\alpha_s\}$ in the right-hand side.

It has to be noticed here that the same value for ω_0 occurs in Eqs. (12) and (13). The coupling constants appearing in expressions (11)–(13) can be written explicitly as

$$V_{\vec{k},\{\beta_s\}} = \langle e_0,\{\alpha_s\}|H_{\text{int}}|g_0,\{\beta_s\}\rangle. \tag{14}$$

In the ket and the bra of expression (14), we do not have explicitly written the photon part and the part associated to the scattering nuclei.

The interaction H_{int} can be written in general terms as usual as

$$H_{\text{int}} = \left(\vec{p} - \frac{q}{m_0}\vec{A}\right)^2 \tag{15}$$

with $\vec{p}$ the momentum operator of the radiating system, q its electric charge, m_o its mass, and $\vec{A}$ the vector potential. Following a standard procedure (see, e.g., Ref. 23), which is basically the development of the vector potential in terms of plane waves, also used in the theoretical description of the Mössbauer effect, it can be shown that the matrix element (14) can be written in terms of a nuclear part, $K_{\vec{k},nucl}$, and a lattice part, $\langle\{\alpha_s\}|e^{i\vec{k}\cdot\vec{u}_0}|\{\beta_s\}\rangle$, where $\vec{u}_0$ is the displacement from equilibrium of the source nucleus. So expression (14) becomes then

$$V_{\vec{k},\{\beta_s\}} = K_{\vec{k},nucl}\langle\{\alpha_s\}|e^{i\vec{k}\cdot\vec{u}_0}|\{\beta_s\}\rangle. \tag{16}$$

Analogously

$$V_{m,\vec{k},\{\beta_s\}} = K_{\vec{k},nucl}e^{i\vec{k}\cdot\vec{r}_m}\langle\{\alpha_s\}|e^{i\vec{k}\cdot\vec{u}_m}|\{\beta_s\}\rangle, \tag{17}$$

where $e^{i\vec{k}\cdot\vec{r}_m}$ is a factor describing the equilibrium position $\vec{r}_m$ where absorption took place and $\vec{u}_m$ is the displacement from equilibrium of nucleus m.

The frequencies involved in expressions (11)–(13) are explicitly

$$\omega_0 = \omega_{0,nucl} + \Omega(\alpha_s), \tag{18}$$

$$\omega_{\vec{k}} = \omega_{\vec{k},phot} + \Omega(\beta_s), \tag{19}$$

where $\hbar\omega_{0,nucl}$ is the energy of the excited nuclear state of the "free" nucleus, $\hbar\omega_{\vec{k},phot}$ is the photon energy, and $\hbar\Omega(\alpha_s)$ and $\hbar\Omega(\beta_s)$ are the phonon energies corresponding to the initial and final phonon distribution, respectively.

Explicitly, the Eqs. (11)–(13) become now

$$(\omega - \omega_{\vec{k}} + i\varepsilon)C_{\vec{k},\{\beta_s\}}(\omega) = K^*_{\vec{k},nucl}\langle\{\beta_s\}|e^{-i\vec{k}\cdot\vec{u}_0}|\{\alpha_s\}\rangle A_{\{\alpha_s\}}(\omega) + \sum_m K^*_{\vec{k},nucl}e^{-i\vec{k}\cdot\vec{r}_m}\langle\{\beta_s\}|e^{-i\vec{k}\cdot\vec{u}_m}|\{\alpha_s\}\rangle B_{m,\{\alpha_s\}}(\omega), \tag{20}$$

$$(\omega-\omega_0+i\varepsilon)A(\omega) = \sum_{\vec{k}}\sum_{\{\beta_s\}} K_{\vec{k},nucl}\langle\{\alpha_s\}|e^{i\vec{k}\cdot\vec{u}_0}|\{\beta_s\}\rangle C_{\vec{k},\{\beta_s\}}(\omega)+1, \quad (21)$$

$$(\omega-\omega_0+i\varepsilon)B_{m,\{\alpha_s\}}(\omega) = \sum_{\vec{k}}\sum_{\{\beta_s\}} K_{\vec{k},nucl}e^{i\vec{k}\cdot\vec{r}_m}\langle\{\alpha_s\}|e^{i\vec{k}\cdot\vec{u}_m}|\{\beta_s\}\rangle C_{\vec{k},\{\beta_s\}}(\omega). \quad (22)$$

Equations (20)–(22) are the fundamental equations describing the complete system. In the next section, these equations will be solved under simplifying assumptions.

III. SOLUTION OF THE EQUATIONS

A. Solution for $A_{\{\alpha_s\}}(\omega)$

Solving Eq. (20) for $C_{\vec{k},\{\beta_s\}}(\omega)$ gives

$$C_{\vec{k},\{\beta_s\}}(\omega) = \frac{1}{\omega-\omega_{\vec{k}}+i\varepsilon}K^*_{\vec{k},nucl}\langle\{\beta_s\}|e^{-i\vec{k}\cdot\vec{u}_0}|\{\alpha_s\}\rangle A_{\{\alpha_s\}}(\omega) + \frac{1}{\omega-\omega_{\vec{k}}+i\varepsilon}\sum_m K^*_{\vec{k},nucl}e^{-i\vec{k}\cdot\vec{r}_m}\langle\{\beta_s\}|e^{-i\vec{k}\cdot\vec{u}_m}|\{\alpha_s\}\rangle \times B_{m,\{\alpha_s\}}(\omega). \quad (23)$$

Substituting expression (23) into Eq. (21) gives

$$(\omega-\omega_0+i\varepsilon)A_{\{\alpha_s\}}(\omega) = \sum_{\{\beta_s\}}\sum_{\vec{k}}\frac{1}{\omega-\omega_{\vec{k}}+i\varepsilon}|K_{\vec{k},nucl}|^2\langle\{\beta_s\}|e^{-i\vec{k}\cdot\vec{u}_0}|\{\alpha_s\}\rangle\langle\{\alpha_s\}|e^{i\vec{k}\cdot\vec{u}_0}|\{\beta_s\}\rangle A_{\{\alpha_s\}}(\omega) + \sum_m\sum_{\{\beta_s\}}\sum_{\vec{k}}\frac{1}{\omega-\omega_{\vec{k}}+i\varepsilon}|K_{\vec{k},nucl}|^2\langle\{\beta_s\}|e^{-i\vec{k}\cdot\vec{u}_m}|\{\alpha_s\}\rangle\langle\{\alpha_s\}|e^{i\vec{k}\cdot\vec{u}_0}|\{\beta_s\}\rangle e^{-i\vec{k}\cdot\vec{r}_m}B_{m,\{\alpha_s\}}(\omega)+1. \quad (24)$$

If we were allowed to neglect the second series of the right-hand side of Eq. (24), then we would have

$$(\omega-\omega_0+i\varepsilon)A_{\{\alpha_s\}}(\omega) = \sum_{\{\beta_s\}}\sum_{\vec{k}}\frac{1}{\omega-\omega_{\vec{k}}+i\varepsilon}|K_{\vec{k},nucl}|^2\langle\{\beta_s\}|e^{-i\vec{k}\cdot\vec{u}_0}|\{\alpha_s\}\rangle \times\langle\{\alpha_s\}|e^{i\vec{k}\cdot\vec{u}_0}|\{\beta_s\}\rangle A_{\{\alpha_s\}}(\omega)+1. \quad (25)$$

The magnitude of $\vec{k}$ varies within a small range that is determined by the linewidth of the nuclear excited state in the case of recoilless processes or, in the case of processes with recoil, by an energy of the order of the energy of the phonons, which is still an order of 10^6 times smaller than the nuclear energy. Furthermore, we suppose that the lattice in which the nuclei are embedded is isotropic. Then the lattice part $\langle\{\alpha_s\}|e^{i\vec{k}\cdot\vec{u}_0}|\{\beta_s\}\rangle$ and its complex conjugate can be evaluated for some value $\vec{k}_0$, whose magnitude is defined by the center of the nuclear linewidth and whose direction is arbitrary. The lattice part can then be put in front of the sum over $\vec{k}$. The reason why $\langle\{\alpha_s\}|e^{i\vec{k}\cdot\vec{u}_0}|\{\beta_s\}\rangle$ does not depend on direction has to do with the fact that the phonon states are related to the properties of the entire solid-state lattice. Because of its isotropy, nothing should depend on the direction of $\vec{k}$.

Expression (25) can then be written as

$$(\omega-\omega_0+i\varepsilon)A_{\{\alpha_s\}}(\omega) = \sum_{\{\beta_s\}}\langle\{\beta_s\}|e^{-i\vec{k}_0\cdot\vec{u}_0}|\{\alpha_s\}\rangle\langle\{\alpha_s\}|e^{i\vec{k}_0\cdot\vec{u}_0}|\{\beta_s\}\rangle \times\sum_{\vec{k}}\frac{1}{\omega-\omega_{\vec{k}}+i\varepsilon}|K_{\vec{k},nucl}|^2A_{\{\alpha_s\}}(\omega)+1. \quad (26)$$

It is shown in Appendix A that

$$\sum_{\vec{k}}\frac{1}{\omega-\omega_{\vec{k}}+i\varepsilon}|K_{\vec{k},nucl}|^2 = \Delta\omega_0 - i\frac{\gamma_R}{2}, \quad (27)$$

where $\Delta\omega_0$ and γ_R are real quantities. The explicit expression for γ_R can be shown to be

$$\gamma_R = \frac{V}{\pi c^3}|K_{\vec{k},nucl}(\omega_{0,nucl})|^2\omega_{0,nucl}{}^2. \quad (28)$$

This definition of γ_R coincides with the linewidth (divided by $\hbar$) defined in Ref. 15. It is the usual radiative linewidth.[24] It can be mentioned here that the presence of V, a volume, in the expressions above and below is only apparent because the matrix element $K_{\vec{k},nucl}(\omega_{0,nucl})$ contains $1/\sqrt{V}$.[24]

With expression (27) and the closure relation $\Sigma_{\{\beta_s\}}|\{\beta_s\}\rangle\langle\{\beta_s\}|=1$, expression (26) can then be written as

$$(\omega-\omega_0+i\varepsilon)A_{\{\alpha_s\}}(\omega) = \left(\Delta\omega_0 - i\frac{\gamma_R}{2}\right)A_{\{\alpha_s\}}(\omega)+1. \quad (29)$$

When moving the term with $A_{\{\alpha_s\}}(\omega)$ to the left-hand side of expression (29), one has

$$A_{\{\alpha_s\}}(\omega)=\frac{1}{\omega-\omega_0+i\frac{\gamma_R}{2}}, \tag{30}$$

where the energy shift $\Delta\omega_0$ has been incorporated into ω_0. When going back to time domain, the exponential radiative decay results.

If the second term of the right-hand side of expression (24) would be retained, the expression for $A_{\{\alpha_s\}}(\omega)$ would contain all $B_{m,\{\alpha_s\}}(\omega)$. This term describes the absorption by the source nucleus of radiation scattered by the absorber nuclei. An iteration procedure, based on expression (31) of the next section, would show that the effective linewidth of the source nucleus would be altered because of the presence of the resonant absorber nuclei. It can be shown that this effect is only appreciable if the distance of the absorber nuclei to the source nucleus is less than 1 Å, which does not occur in a solid. Therefore, this reabsorption term will be neglected, as was the case in the model developed in Ref. 15. It means that we neglect the processes where a photon emitted by the source nucleus is reabsorbed by it after being scattered by an absorber nucleus.

Expression (30) is the usual expression of the source amplitude. It gives a Lorentzian frequency distribution, centered on the energy ω_0. If the conversion electron processes would have been included, γ_R would have been replaced by the total linewidth Γ, which is the sum of the radiative width and the width due to the processes with electron conversion.

B. Solution for $B_{m,\{\alpha_s\}}(\omega)$

Substituting expression (23) into expression (22) gives

$$(\omega-\omega_0+i\varepsilon)B_{m,\{\alpha_s\}}(\omega)=\sum_{\vec{k}}\sum_{\{\beta_s\}}|K_{\vec{k},nucl}|^2e^{i\vec{k}\cdot\vec{r}_m}\frac{1}{\omega-\omega_{\vec{k}}+i\varepsilon}\langle\{\alpha_s\}|e^{i\vec{k}\cdot\vec{u}_m}|\{\beta_s\}\rangle\langle\{\beta_s\}|e^{-i\vec{k}\cdot\vec{u}_0}|\{\alpha_s\}\rangle A_{\{\alpha_s\}}(\omega)$$
$$+\sum_{\vec{k}}\sum_{\{\beta_s\}}\sum_{m'}|K_{\vec{k},nucl}|^2\frac{1}{\omega-\omega_{\vec{k}}+i\varepsilon}e^{i\vec{k}\cdot(\vec{r}_m-\vec{r}_{m'})}\langle\{\beta_s\}|e^{-i\vec{k}\cdot\vec{u}_{m'}}|\{\alpha_s\}\rangle\langle\{\alpha_s\}|e^{i\vec{k}\cdot\vec{u}_m}|\{\beta_s\}\rangle B_{m',\{\alpha_s\}}(\omega). \tag{31}$$

We will calculate successively the first sum, denoted I_m, and the second sum over $\vec{k}$, $\{\beta_s\}$ in the right-hand side of expression (31). The second sum can be split in terms with $m=m'$, denoted $I_{m,m}$, and terms with $m\neq m'$, denoted $I_{m,m'}$. We have then

$$(\omega-\omega_0+i\varepsilon)B_{m,\{\alpha_s\}}(\omega)=I_mA_{\{\alpha_s\}}(\omega)+I_{m,m}B_{m,\{\alpha_s\}}(\omega)+\sum_{m'\neq m}I_{m,m'}B_{m',\{\alpha_s\}}(\omega). \tag{32}$$

As before, the lattice contributions can be evaluated at some central value $\vec{k}_0$. $I_{m,m}$ has been calculated before [see expression (26) and the discussion following it]. It will again give rise to a shift, to be incorporated into ω_0, and a radiative width $\gamma_R/2$ when brought to the left-hand side of expression (32), which can be written then as

$$\left(\omega-\omega_0+i\frac{\gamma_R}{2}\right)B_{m,\{\alpha_s\}}(\omega)=I_mA_{\{\alpha_s\}}(\omega)+\sum_{m'\neq m}I_{m,m'}B_{m',\{\alpha_s\}}(\omega). \tag{33}$$

I_m is explicitly given by

$$I_m=\langle\{\alpha_s\}|e^{i\vec{k}_0\cdot(\vec{u}_m-\vec{u}_0)}|\{\alpha_s\}\rangle\sum_{\vec{k}}\frac{1}{\omega-\omega_{\vec{k}}+i\varepsilon}|K_{\vec{k},nucl}|^2e^{i\vec{k}\cdot\vec{r}_m}, \tag{34}$$

where the closure relation $\Sigma_{\{\beta_s\}}|\{\beta_s\}\rangle\langle\{\beta_s\}|=1$ has again been applied.

The remaining sum over $\vec{k}$ can again be transformed into an integral, where we can replace $\omega_{\vec{k}}$ by $\omega_{\vec{k},phot}$.

$$\sum_{\vec{k}}|K_{\vec{k},nucl}|^2e^{i\vec{k}\cdot\vec{r}_m}\frac{1}{\omega-\omega_{\vec{k},phot}+i\varepsilon}=\frac{V}{(2\pi)^3}\int\int\int|K_{\vec{k},nucl}|^2\frac{1}{\omega-\omega_{\vec{k},phot}+i\varepsilon}e^{i\vec{k}\cdot\vec{r}_m}d^3\vec{k}. \tag{35}$$

The integral of the right-hand side of expression (35) has been calculated in Ref. 15. One has

$$\frac{V}{(2\pi)^3}\int\int\int|K_{\vec{k},nucl}|^2\frac{1}{\omega-\omega_{\vec{k},phot}+i\varepsilon}e^{i\vec{k}\cdot\vec{r}_m}d^3\vec{k}=-\frac{\gamma_R}{2}\frac{e^{i\omega r_m/c}}{\omega r_m/c}. \tag{36}$$

Substituting expression (36) into expression (34) gives

$$I_m = -\frac{\gamma_R}{2}\frac{e^{i\omega r_m/c}}{\omega r_m/c}\langle\{\alpha_s\}|e^{i\vec{k}_0\cdot(\vec{u}_m-\vec{u}_0)}|\{\alpha_s\}\rangle. \quad (37)$$

We still have to calculate $I_{m,m'}$. Using again the closure relation $\Sigma_{\{\beta_s\}}|\{\beta_s\}\rangle\langle\{\beta_s\}|=1$, $I_{m,m'}$ is explicitly given by

$$I_{m,m'} = \langle\{\alpha_s\}|e^{i\vec{k}_0\cdot(\vec{u}_m-\vec{u}_{m'})}|\{\alpha_s\}\rangle \times\sum_{\vec{k}}|K_{\vec{k},nucl}|^2\frac{1}{\omega-\omega_{\vec{k}}+i\varepsilon}e^{i\vec{k}\cdot(\vec{r}_m-\vec{r}_{m'})}. \quad (38)$$

The remaining sum over $\vec{k}$ can again be transformed into an integral, which has the same structure as the one in expression (35). Therefore, we have

$$I_{m,m'} = \frac{-\gamma_R}{2}\frac{e^{i\omega|\vec{r}_m-\vec{r}_{m'}|/c}}{\omega|\vec{r}_m-\vec{r}_{m'}|/c}\langle\{\alpha_s\}|e^{i\vec{k}_0\cdot(\vec{u}_m-\vec{u}_{m'})}|\{\alpha_s\}\rangle. \quad (39)$$

When expressions (37) and (39) are substituted into expression (33), we have

$$\left(\omega-\omega_0+i\frac{\gamma_R}{2}\right)B_{m,\{\alpha_s\}}(\omega) = -\frac{\gamma_R}{2}\frac{e^{i\omega r_m/c}}{\omega r_m/c}\langle\{\alpha_s\}|e^{i\vec{k}_0\cdot(\vec{u}_m-\vec{u}_0)}|\{\alpha_s\}\rangle A_{\{\alpha_s\}}(\omega) - \frac{\gamma_R}{2}\sum_{m'\neq m}\frac{e^{i\omega|\vec{r}_m-\vec{r}_{m'}|/c}}{\omega|\vec{r}_m-\vec{r}_{m'}|/c}\langle\{\alpha_s\}|e^{i\vec{k}_0\cdot(\vec{u}_m-\vec{u}_{m'})}|\{\alpha_s\}\rangle B_{m',\{\alpha_s\}}(\omega). \quad (40)$$

The interpretation of expression (40) is straightforward. The first term of its right-hand side gives the excitation amplitude of absorber m, situated at $\vec{r}_m$, due to the field produced by the source nucleus. The sum over m' gives the excitation amplitude of nucleus m due to the field that is produced by scattering of the radiation by all other absorber nuclei m', situated at $\vec{r}_{m'}$.

Later we will consider the condition $\vec{u}_m=\vec{u}_{m'}$ (see Sec. IV). If this condition is realized and if it is assumed that the surroundings of each absorber nucleus are identical, the second term of the right-hand side of expression (40) will be equivalent to an average field, produced by all absorber nuclei. This average field will be the same for all absorber nuclei. In the holographic image (see Sec. IV), this will produce only an attenuation of the holographic contrast, without changing the holographic information. In the following discussion, this average part, which corresponds in fact to multiple scattering, will not be considered anymore. In Ref. 15, the multiple scattering processes were neglected altogether, corresponding to the single scattering approximation, which is certainly valid for small samples.

C. Solution for $C_{\vec{k},\{\beta_s\}}(\omega)$ and $c_{\vec{k},\{\beta_s\}}(t)$

If we substitute expressions (30) and (40), not considering the multiple scattering processes, into expression (20), we obtain

$$(\omega-\omega_{\vec{k}}+i\varepsilon)C_{\vec{k},\{\beta_s\}}(\omega) = K^*_{\vec{k},nucl}\langle\{\beta_s\}|e^{-i\vec{k}\cdot\vec{u}_0}|\{\alpha_s\}\rangle\frac{1}{\omega-\omega_0+i\frac{\gamma_R}{2}} - \frac{\gamma_R}{2}\frac{1}{\left(\omega-\omega_0+i\frac{\gamma_R}{2}\right)^2}\sum_m K^*_{\vec{k},nucl}e^{-i\vec{k}\cdot\vec{r}_m}\frac{e^{i\omega r_m/c}}{\omega r_m/c} \times\langle\{\beta_s\}|e^{-i\vec{k}\cdot\vec{u}_m}|\{\alpha_s\}\rangle\langle\{\alpha_s\}|e^{i\vec{k}_0\cdot(\vec{u}_m-\vec{u}_0)}|\{\alpha_s\}\rangle. \quad (41)$$

When going back to time domain, it is shown in Appendix B that $c_{\vec{k},\{\beta_s\}}(t)$ is given by the sum of two terms

$$c_{\vec{k},\{\beta_s\}}(t) = c^{(0)}_{\vec{k},\{\beta_s\}}(t) + c^{(1)}_{\vec{k},\{\beta_s\}}(t) \quad (42)$$

with

$$c^{(0)}_{\vec{k},\{\beta_s\}}(t) = K^*_{\vec{k},nucl}\langle\{\beta_s\}|e^{-i\vec{k}\cdot\vec{u}_0}|\{\alpha_s\}\rangle \times\frac{1-e^{i(\omega_{\vec{k},phot}-\omega_{0,nucl}+\Omega(\beta_s)-\Omega(\alpha_s)+i\gamma_R/2)t}}{\omega_{\vec{k},phot}-\omega_{0,nucl}+\Omega(\beta_s)-\Omega(\alpha_s)+i\frac{\gamma_R}{2}} \quad (43)$$

and

$$c^{(1)}_{\vec{k},\{\beta_s\}}(t) = -\frac{\gamma_R}{2}\sum_m K^*_{\vec{k},nucl}e^{-i\vec{k}\cdot\vec{r}_m}\langle\{\beta_s\}|e^{-i\vec{k}\cdot\vec{u}_m}|\{\alpha_s\}\rangle\langle\{\alpha_s\}|e^{i\vec{k}_0\cdot(\vec{u}_m-\vec{u}_0)}|\{\alpha_s\}\rangle\frac{1}{\omega_{0,nucl}r_m/c}\left\{\frac{e^{i\omega_{\vec{k}}r_m/c}}{\left(\omega_{\vec{k},phot}-\omega_0+i\frac{\gamma_R}{2}\right)^2} + \left[i\left(\omega_{\vec{k}}-\omega_0+i\frac{\gamma_R}{2}\right)\left(\frac{r_m}{c}-t\right)-1\right]\frac{e^{i(r_m/c-t)(\omega_0-i\gamma_R/2)}}{\omega_{\vec{k}}-\omega_0+i\frac{\gamma_R}{2}}\right\}. \quad (44)$$

In the next section, these expressions will be used to calculate the probability to find a gamma ray with wave vector $\vec{k}$ present due to processes without and with recoil.

IV. DISCUSSION

As has been mentioned already, $c^{(0)}_{\vec{k},\{\beta_s\}}(t)$, given by expression (43), is the probability amplitude of finding a photon with wave vector $\vec{k}$, with the lattice in state $\{\beta_s\}$, taking into account only the source nucleus. For long times, the probability distribution is given by $P^{(0)}_{\vec{k},\{\beta_s\}}(+\infty)=|c^{(0)}_{\vec{k},\{\beta_s\}}(+\infty)|^2$. In fact, the initial lattice state $\{\alpha_s\}$ is not unique. We have to take into account the probability $g(\{\alpha_s\})$ to find the lattice in the state $\{\alpha_s\}$.[25] with the obvious normalization condition $\Sigma_{\{\alpha_s\}}g(\{\alpha_s\})=1$. So each $P^{(0)}_{\vec{k},\{\beta_s\}}(+\infty)$ has to be multiplied by $g(\{\alpha_s\})$. We also have to sum over all final states $\{\beta_s\}$. The probability to find a photon with wave vector $\vec{k}$ present for long times is then given by

$$W^{(0)}_{\vec{k}}(+\infty)=|K^*_{\vec{k},nucl}|^2\sum_{\{\beta_s\}}\sum_{\{\alpha_s\}}g(\{\alpha_s\})|\langle\{\beta_s\}|e^{-i\vec{k}\cdot\vec{u}_0}|\{\alpha_s\}\rangle|^2 \times\frac{1}{[\omega_{\vec{k},phot}-\omega_{0,nucl}+\Omega(\beta_s)-\Omega(\alpha_s)]^2+\frac{\gamma_R^2}{4}}. \tag{45}$$

It can be noted at this stage that we do not have to consider lattice interference terms because they vanish due to the presence of random phases since the $\{\beta_s\}$ states are distinguishable.

Expression (45) represents a sum of Lorentzian distributions with width $\gamma_R/2$, concentrated around the value $\omega_{0,nucl}+\Omega(\alpha_s)-\Omega(\beta_s)$. Each Lorentzian has a coefficient $g(\{\alpha_s\})|K^*_{\vec{k},nucl}|^2|\langle\{\beta_s\}|e^{-i\vec{k}\cdot\vec{u}_0}|\{\alpha_s\}\rangle|^2$. For each lattice combination $\{\alpha_s\}$, $\{\beta_s\}$, there is one Lorentzian.

For processes without recoil, we have $|\{\alpha_s\}\rangle=|\{\beta_s\}\rangle$, so that expression (45) can be simplified

$$W^{(0)}_{\vec{k},nrec}(+\infty)=|K^*_{\vec{k},nucl}|^2\sum_{\{\alpha_s\}}g(\{\alpha_s\})|\langle\{\alpha_s\}|e^{-i\vec{k}\cdot\vec{u}_0}|\{\alpha_s\}\rangle|^2 \times\frac{1}{(\omega_{\vec{k},phot}-\omega_{0,nucl})^2+\frac{\gamma_R^2}{4}} =f|K^*_{\vec{k},nucl}|^2\frac{1}{(\omega_{\vec{k},phot}-\omega_{0,nucl})^2+\frac{\gamma_R^2}{4}}, \tag{46}$$

where we have used the definition of the recoilless fraction[25] $f=\Sigma_{\{\alpha_s\}}g(\{\alpha_s\})|\langle\{\alpha_s\}|e^{-i\vec{k}\cdot\vec{u}_0}|\{\alpha_s\}\rangle|^2$.

This is nothing but the familiar Lorentzian distribution of photons emitted by a source nucleus, centered at the nuclear energy $\omega_{0,nucl}$ and having a width $\gamma_R/2$. The present analysis automatically takes into account the recoilless fraction.

$c^{(1)}_{\vec{k},\{\beta_s\}}(t)$, given by expression (44), is the probability amplitude of having a photon of wave vector $\vec{k}$, with the lattice in state $\{\beta_s\}$ due to the scattering by all resonant nuclei m at positions $\vec{r}_m$. For long times, the explicit expression is given by

$$c^{(1)}_{\vec{k},\{\beta_s\}}(+\infty)=-\frac{\gamma_R}{2}\sum_m K^*_{\vec{k},nucl}e^{-i\vec{k}\cdot\vec{r}_m}\langle\{\beta_s\}|e^{-i\vec{k}\cdot\vec{u}_m}|\{\alpha_s\}\rangle \times\langle\{\alpha_s\}|e^{i\vec{k}_0\cdot(\vec{u}_m-\vec{u}_0)}|\{\alpha_s\}\rangle\frac{e^{ir_m/c[\omega_{\vec{k},phot}+\Omega(\beta_s)]}}{\omega_{0,nucl}r_m/c} \times\frac{1}{\left(\omega_{\vec{k},phot}-\omega_{0,nucl}+\Omega(\beta_s)-\Omega(\alpha_s)+i\frac{\gamma_R}{2}\right)^2}, \tag{47}$$

where expressions (18) and (19) have been used.

It is well-known (see, e.g., Ref. 13) that translational symmetry of the lattice severely distorts the hologram so that only samples where the absorber nuclei in the immediate environment (of the order of a few ångstroms) of the source nucleus are suitable for emission holography. In Ref. 13 ways are discussed to get rid of possible effects of translational symmetry to some extent. As will be discussed later in this section, correlated vibrations could make the source and near-neighbor absorber nuclei move together, so that processes with recoil could give a contribution to the holographic image. If we consider absorber nuclei at a few ångstroms from the source nucleus, and for phonon energies[26] of the order of 0.01 eV, then $e^{ir_m/c\Omega(\beta_s)}\cong 1$. Then it can be shown that the expression for $c^{(1)}_{\vec{k},\{\beta_s\}}(+\infty)$ can be written as

$$c^{(1)}_{\vec{k},\{\beta_s\}}(+\infty)=-\frac{\gamma_R}{2}K^*_{\vec{k},nucl}\sum_m e^{-i\vec{k}\cdot\vec{r}_m}\frac{e^{ir_m/c\omega_{\vec{k},phot}}}{\omega_{0,nucl}r_m/c} \times\frac{1}{\left(\omega_{\vec{k},phot}-\omega_{0,nucl}+\Omega(\beta_s)-\Omega(\alpha_s)+i\frac{\gamma_R}{2}\right)^2} \times\langle\{\beta_s\}|e^{-i\vec{k}\cdot\vec{u}_m}|\{\alpha_s\}\rangle\langle\{\alpha_s\}|e^{i\vec{k}_0\cdot(\vec{u}_m-\vec{u}_0)}|\{\alpha_s\}\rangle. \tag{48}$$

If only recoilless processes are considered, then this expression can be simplified

$$c^{(1)}_{\vec{k},\{\alpha_s\},nrec}(+\infty)=-\frac{\gamma_R}{2}K^*_{\vec{k},nucl}\sum_m e^{-i\vec{k}\cdot\vec{r}_m}\frac{e^{ir_m/c\omega_{\vec{k},phot}}}{\omega_{0,nucl}r_m/c} \times\frac{1}{\left(\omega_{\vec{k},phot}-\omega_{0,nucl}+i\frac{\gamma_R}{2}\right)^2} \cdot\langle\{\alpha_s\}|e^{-i\vec{k}\cdot\vec{u}_m}|\{\alpha_s\}\rangle\langle\{\alpha_s\}|e^{i\vec{k}_0\cdot(\vec{u}_m-\vec{u}_0)}|\{\alpha_s\}\rangle. \tag{49}$$

If we take again into account the fact that there is a distribution of initial lattice states, the probability of finding a photon with wave vector $\vec{k}$ present for long times if only recoilless processes are considered is then given by

$$P_{\vec{k},nrec}(+\infty) = W^{(0)}_{\vec{k},nrec}(+\infty) + \sum_{\{\alpha_s\}} g(\{\alpha_s\})|c^{(1)}_{\vec{k},\{\alpha_s\},nrec}(+\infty)|^2 + 2\,\mathrm{Re}\left[\sum_{\{\alpha_s\}} g(\{\alpha_s\})c^{(0)}_{\vec{k},\{\alpha_s\},nrec}(+\infty) \times c^{(1)}_{\vec{k},\{\alpha_s\},nrec}(+\infty)^*\right] \tag{50}$$

with $W^{(0)}_{\vec{k},nrec}(+\infty)$ given by expression (46) and $c^{(0)}_{\vec{k},\{\alpha_s\},nrec}(+\infty)$ defined by [see expression (44) specialized for $|\{\alpha_s\}\rangle = |\{\beta_s\}\rangle$ and for $t \to +\infty$]

$$c^{(0)}_{\vec{k},\{\alpha_s\},nrec}(+\infty) = K^*_{\vec{k},nucl}\langle\{\alpha_s\}|e^{-i\vec{k}\cdot\vec{u}_0}|\{\alpha_s\}\rangle \times \frac{1}{\omega_{\vec{k},phot} - \omega_{0,nucl} + i\frac{\gamma_R}{2}}. \tag{51}$$

The last term of expression (50) gives the holographic information, as has been explained in Ref. 15. It gives the interference of the path where the photon travels directly from the source nucleus to the detector and the path where the photon emitted by the source nucleus is first scattered by the absorber nuclei before going to the detector.

If we consider also the conversion electron processes, it can be shown that the interference term is given explicitly by

$$2\,\mathrm{Re}\left[\sum_{\{\alpha_s\}} g(\{\alpha_s\})c^{(0)}_{\vec{k},nrec}(+\infty)c^{(1)}_{\vec{k},nrec}(+\infty)^*\right] = -\mathrm{Re}\left[\frac{|K^*_{\vec{k},nucl}|^2\gamma_R}{\omega_{\vec{k},phot} - \omega_{0,nucl} + i\frac{\Gamma}{2}} \frac{1}{\left(\omega_{\vec{k},phot} - \omega_{0,nucl} - i\frac{\Gamma}{2}\right)^2} \sum_m e^{i\vec{k}\cdot\vec{r}_m} \frac{e^{-ir_m/c\,\omega_{\vec{k},phot}}}{\omega_{0,nucl}r_m/c} \times \sum_{\{\alpha_s\}} g(\{\alpha_s\})\langle\{\alpha_s\}|e^{-i\vec{k}\cdot\vec{u}_0}|\{\alpha_s\}\rangle\langle\{\alpha_s\}|e^{i\vec{k}\cdot\vec{u}_m}|\{\alpha_s\}\rangle\langle\{\alpha_s\}|e^{-i\vec{k}_0\cdot\vec{u}_m}|\{\alpha_s\}\rangle \times \langle\{\alpha_s\}|e^{i\vec{k}_0\cdot(\vec{u}_m-\vec{u}_0)}|\{\alpha_s\}\rangle\right]. \tag{52}$$

This term is analogous to expression (23) of Ref. 15 with the only difference the sum over $\{\alpha_s\}$, which describes explicitly the influence of the lattice part. In Ref. 15 it was assumed that all processes occur without recoil. Expression (52) is a generalization of the interference term eventually leading to nuclear emission holography.

The lattice part in expression (52),

$$\sum_{\{\alpha_s\}} g(\{\alpha_s\})\langle\{\alpha_s\}|e^{-i\vec{k}\cdot\vec{u}_0}|\{\alpha_s\}\rangle\langle\{\alpha_s\}|e^{i\vec{k}\cdot\vec{u}_m}|\{\alpha_s\}\rangle \times \langle\{\alpha_s\}|e^{i\vec{k}_0\cdot(\vec{u}_m-\vec{u}_0)}|\{\alpha_s\}\rangle,$$

is a complicated expression that can be calculated using methods from quantum field theory. For low temperatures, the lattice is in its lowest state and all α_s will be zero. In this case, the sum over $\{\alpha_s\}$ is reduced to the square f^2 of the recoilless fraction f.

Let us go back to the more general case, described by Eq. (48) for $c^{(1)}_{\vec{k},\{\beta_s\}}(+\infty)$ and by $c^{(0)}_{\vec{k},\{\beta_s\}}(+\infty)$ given by [see expression (43), specialized for $t \to +\infty$]

$$c^{(0)}_{k,\{\beta_s\}}(+\infty) = K^*_{k,nucl}\langle\{\beta_s\}|e^{-i\vec{k}\cdot\vec{u}_0}|\{\alpha_s\}\rangle \times \frac{1}{\omega_{\vec{k},phot} - \omega_{0,nucl} + \Omega(\beta_s) - \Omega(\alpha_s) + i\frac{\gamma_R}{2}} \tag{53}$$

and

$$c^{(1)}_{k,\{\beta_S\}}(+\infty) = -\frac{\gamma_R}{2}K^*_{\vec{k},\mathrm{nucl}}\sum_m e^{-i\vec{k}\cdot\vec{r}_m}\frac{e^{ir_m/c\,\omega_{\vec{k},\mathrm{phot}}}}{\omega_{0,\mathrm{nucl}}r_m/c} \times \langle\{\beta_s\}|e^{-i\vec{k}\cdot\vec{u}_m}|\{\alpha_s\}\rangle \times \frac{1}{\left(\omega_{\vec{k},\mathrm{phot}} - \omega_{0,\mathrm{nucl}} + \Omega(\beta_s) - \Omega(\alpha_s) + i\frac{\gamma_R}{2}\right)^2} \times \langle\{\alpha_s\}|e^{i\vec{k}_0\cdot(\vec{u}_m-\vec{u}_0)}|\{\alpha_s\}\rangle. \tag{54}$$

The interference term, $I_{\vec{k},\{\beta_s\}} = 2\mathrm{Re}[c^{(0)}_{\vec{k},\{\beta_s\}}(+\infty)c^{(1)}_{\vec{k},\{\beta_s\}}(+\infty)^*]$ leading to the holographic information is now, using expressions (53) and (54) and taking into account also the conversion electron processes

$$I_{\vec{k},\{\beta_s\}} = -\gamma_R |K_{\vec{k},\mathrm{nucl}}|^2 \,\mathrm{Re}\left[\langle\{\beta_s\}|e^{-i\vec{k}\cdot\vec{u}_0}|\{\alpha_s\}\rangle \frac{1}{\omega_{\vec{k},\mathrm{phot}} - \omega_{0,\mathrm{nucl}} + \Omega(\beta_s) - \Omega(\alpha_s) + i\frac{\Gamma}{2}} \times \sum_m e^{i\vec{k}\cdot\vec{r}_m} \frac{e^{-ir_m/c\,\omega_{\vec{k},\mathrm{phot}}}}{\omega_{0,\mathrm{nucl}} r_m/c} \langle\{\alpha_s\}|e^{i\vec{k}\cdot\vec{u}_m}|\{\beta_s\}\rangle \times \frac{1}{\left(\omega_{\vec{k},\mathrm{phot}} - \omega_{0,\mathrm{nucl}} + \Omega(\beta_s) - \Omega(\alpha_s) - i\frac{\Gamma}{2}\right)^2} \langle\{\alpha_s\}|e^{-i\vec{k}_0\cdot(\vec{u}_m - \vec{u}_0)}|\{\alpha_s\}\rangle \right]. \tag{55}$$

If we were allowed to write $\vec{u}_m = \vec{u}_0$, for all m, expression (55) would be simplified and one would have

$$I_{\vec{k},\{\beta_s\}} = -\gamma_R |K_{\vec{k},\mathrm{nucl}}|^2 \,\mathrm{Re}\left[\frac{1}{\left(\omega_{\vec{k},\mathrm{phot}} - \omega_{0,\mathrm{nucl}} + \Omega(\beta_s) - \Omega(\alpha_s) + i\frac{\Gamma}{2}\right)\left(\omega_{\vec{k},\mathrm{phot}} - \omega_{0,\mathrm{nucl}} + \Omega(\beta_s) - \Omega(\alpha_s) - i\frac{\Gamma}{2}\right)^2} \times |\langle\{\beta_s\}|e^{-i\vec{k}\cdot\vec{u}_0}|\{\alpha_s\}\rangle|^2 \sum_m e^{i\vec{k}\cdot\vec{r}_m} \frac{e^{-ir_m/c\,\omega_{\vec{k},\mathrm{phot}}}}{\omega_{0,\mathrm{nucl}} r_m/c} \right]. \tag{56}$$

As has been explained in detail in Ref. 15, the holographic information, from which the reconstruction to obtain the real image will have to be made, is contained in the sum over m in expression (56).

If we compare this with the expression for the "direct" term [using expression (53)]

$$|c^{(0)}_{\vec{k},\{\beta_S\}}(+\infty)|^2 = |K_{\vec{k},\mathrm{nucl}}|^2 \times \left| \frac{1}{\omega_{\vec{k},\mathrm{phot}} - \omega_{0,\mathrm{nucl}} + \Omega(\beta_s) - \Omega(\alpha_s) + i\frac{\Gamma}{2}} \right|^2 \times |\langle\{\beta_s\}|e^{-i\vec{k}\cdot\vec{u}_0}|\{\alpha_s\}\rangle|^2, \tag{57}$$

we see that the lattice part is exactly the same in both expressions. The ratio of the direct term and the interference term is not zero for processes with recoil. This means that we could have interference contributing to the holographic image from processes with as well as without recoil.

It has to be pointed out that the structural part,

$$\sum_m e^{i\vec{k}\cdot\vec{r}_m} \frac{e^{-ir_m/c\,\omega_{\vec{k}\cdot\mathrm{phot}}}}{\omega_{0,\mathrm{nucl}} r_m/c},$$

is the same for all $\vec{k}$ values, this due to the fact that the phonon energies are of the order of 0.01 eV and for r_m not larger than a few ångstroms. This means that the interference patterns are the same for all relevant $\vec{k}$ values.

The complete holographic image will be formed by an incoherent superposition of images for each combination $\{\alpha_s\},\{\beta_s\}$, all images having the same structural factor. The lattice part $|\langle\{\beta_s\}|e^{-i\vec{k}\cdot\vec{u}_0}|\{\alpha_s\}\rangle|^2$ will be the same for each combination.

Let us examine the condition $\vec{u}_m = \vec{u}_0$. Let us recall the definition of $\vec{u}_0$ and $\vec{u}_m$. They are the displacement vectors of the source nucleus and nucleus m, respectively, with respect to their equilibrium positions. The equilibrium position of the source nucleus has been chosen as the origin of the coordinate system and the equilibrium position of nucleus m is at $\vec{r}_m$. The displacement vectors can be developed in normal modes[27] as follows:

$$\vec{u}_0 = \sum_{j=1}^{3N} \sqrt{\frac{\hbar}{2M\omega_j N}} \vec{e}_j (a_j + a_j^+), \tag{58}$$

$$\vec{u}_m = \sum_{j=1}^{3N} \sqrt{\frac{\hbar}{2M\omega_j N}} \vec{e}_j (a_j e^{i\vec{q}_j\cdot\vec{r}_m} + a_j^+ e^{-i\vec{q}_j\cdot\vec{r}_m}). \tag{59}$$

M is the mass of the nucleus, N the number of atoms in the crystal, ω_j is the frequency associated to mode j, $\vec{q}_j$ is the wave vector of the vibrational mode j. a_j and a_j^+ are, respectively, the annihilation and creation operators of phonons associated to mode j. $\vec{e}_j$ is the polarization vector (unit vector) associated to mode j. Because the equilibrium position of the source nucleus is at the origin of the coordinate system, there are no terms like $e^{\pm i\vec{q}_j\cdot\vec{r}_m}$ in expression (58).

The physical interpretation of the assumption $\vec{u}_m = \vec{u}_0$ is that the source nucleus, situated at the origin, and absorber nucleus m, situated at $\vec{r}_m$, have the same displacement with respect to their equilibrium positions. Using expressions (58) and (59), the function $e^{-i\vec{k}_0(\vec{u}_m - \vec{u}_0)}$ present in expression (55) can be written as

$$e^{-i\vec{k}_0(\vec{u}_m-\vec{u}_0)} = e^{-i\vec{k}_0\cdot\sum_{j=1}^{3N}\sqrt{\frac{\hbar}{2M\omega_j N}}\vec{e}_j[a_j(e^{i\vec{q}_j\cdot\vec{r}_m}-1)+a_j^+(e^{-i\vec{q}_j\cdot\vec{r}_m}-1)]}. \tag{60}$$

In terms of wave vectors associated to the vibrational modes, the condition $\vec{u}_m=\vec{u}_0$ means that the magnitude of the relevant wave vectors $|\vec{q}_j|$ is small compared to $|\vec{r}_m|^{-1}$, and this for all m. This means that the wavelengths associated to the relevant vibrational modes are large compared to the distance of the absorber nuclei with respect to the source nucleus. This could be interpreted by saying that the vibrational effects produced in the lattice by the recoil due to photon emission by the source nucleus is the same for all absorber nuclei situated in the vicinity of the source nucleus. Heuristically, one could say that the wave produced by the source nucleus due to its recoil is such that all absorber nuclei experience the same perturbation. If this correlated vibrational movement of source and absorber nuclei could occur, it would only be for an absorber nuclei close to the source nucleus,[28] thus for those nuclei that are involved in the formation of the holographic image. At the end of this section, more discussions will be devoted to the influence of these correlated effects.

For nuclei far from the source nucleus, the condition $\vec{u}_m=\vec{u}_0$ is certainly not valid, and one has to consider expression (55) rather than expression (56). If expression (60) is inserted into expression (55), we have the factor

$$\langle\{\alpha_s\}|e^{-i\vec{k}_0\cdot\sum_{j=1}^{3N}\sqrt{\frac{\hbar}{2M\omega_j N}}\vec{e}_j[a_j(e^{i\vec{q}_j\cdot\vec{r}_m}-1)+a_j^+(e^{-i\vec{q}_j\cdot\vec{r}_m}-1)]}|\{\alpha_s\}\rangle.$$

If the magnitude of some of the relevant wave vectors $|\vec{q}_j|$ is comparable or larger than $|\vec{r}_m|^{-1}$, then the nuclei m do not move coherently with respect to the source nucleus and the exponential terms will produce destructive interference. This will have the effect that the processes with recoil will produce a negligible contribution in the interference term for nuclei far from the source nucleus, i.e., we would have only a contribution coming from the processes without recoil. If the recoilless fraction goes to zero, in this case there would not be any holographic image.

In order to be a bit more specific, we will give an estimate of the influence of the correlated motion of the source nucleus and the near-neighbor absorber nuclei for a single crystal of ^{57}Fe, where an excited ^{57}Fe nucleus, at the center of a cubic unit cell, is surrounded by eight nearest-neighbor nuclei in the ground state. This will give us a quantitative estimate of the holographic analogue of the Mössbauer-Lamb factor for this case. We will use a model explained in Ref. 28 (see also Refs. 29 and 30 for equivalent approaches). What needs to be calculated is in fact $e^{-1/2k^2\sigma_m^2}$, where σ_m^2 is the mean square relative displacement of nucleus m, situated at a distance r_m from the source nucleus, which is 2.4855 Å for the case we considered above. In the case of uncorrelated motion, this would result in the ordinary Mössbauer-Lamb factor. For nearest-neighbor nuclei, the vibrational factors $e^{-1/2k^2\sigma_m^2}$ are much closer to unity[28] than would be the case for uncorrelated motion. What is required now is a method for calculating σ_m^2 using a correlated vibrational model. Using a simple correlated Debye model, it can be shown[28] that

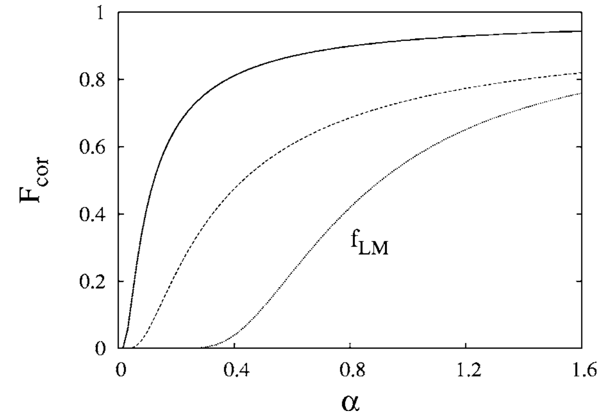

FIG. 1. Holographic analog, $F_{cor}=e^{-1/2k^2\sigma_m}$, of the Lamb-Mössbauer factor, as a function of $\alpha=\theta_D T$ for r_m=2.4855 Å (solid line) for a source nucleus at the center of a unit cell of ^{57}Fe, surrounded by eight nearest-neighbor absorber nuclei in the ground state. The ordinary Lamb-Mössbauer factor f_{LM} is also shown for comparison (dotted line). The dashed line shows F_{cor} for the (hypothetical) case of r_m=40 Å.

$$\sigma_m^2 = \frac{3\hbar^2}{2Mk_B\theta_D q_D^2}\Bigg\{q_D^2 + 4\frac{q_D^2}{\alpha^2}\left[\frac{\pi^2}{6} - \sum_{n=1}^{\infty}\left(\frac{1}{n^2}+\frac{\alpha}{n}\right)e^{-\alpha n}\right] - \frac{2}{r_m^2}(1-\cos\beta_m) + 4\frac{q_D}{r_m\alpha}\sum_{n=1}^{\infty}\frac{1}{n^2+\frac{\beta_m^2}{\alpha^2}} \times\left[e^{-\alpha n}\left(n\sin\beta_m + \frac{\beta_m}{\alpha}\cos\beta_m\right) - \frac{\beta_m}{\alpha}\right]\Bigg\}. \tag{61}$$

M is the mass of ^{57}Fe, k_B is the Boltzmann constant, T the temperature of the crystal, θ_D is the Debye temperature, and q_D is the Debye wave number of the crystal, $\alpha=\theta_D/T$ and $\beta_m=q_D r_m$. For an iron crystal, we take a Debye temperature of 440 K. The other constants[31] are $M=9.5\times10^{-26}$ kg, $k_B=1.38\times10^{-23}$ JK^{-1}, and $q_D=0.96\times10^{10}$ m^{-1}.

Figure 1 shows $F_{cor}=e^{-1/2k^2\sigma_m^2}$ as a function of $\alpha=\theta_D/T$ for r_m=2.4855 Å (solid line) for the ^{57}Fe configuration we considered above. The ordinary Lamb-Mössbauer factor f_{LM} is also shown for comparison (dotted line). F_{cor} is also shown for the (hypothetical) case of r_m=40 Å (dashed line). It is clearly seen that the effect of the correlated movement of the absorber nuclei in the immediate surroundings of the source nucleus drastically increases the value of the holographic analogue of the Lamb-Mössbauer factor F_{cor}. As is well-known, the ordinary Lamb-Mössbauer factor goes rather fast to zero for temperatures above the Debye temperature, while in the correlated model F_{cor} decreases much more slowly in a fairly wide temperature range.

More information on correlated models as well as refinements for the calculations can be found in, e.g., Ref. 28.

A final word should be said about the nonresonant processes. A photon emitted by a nucleus inside a solid can also be scattered by atomic electrons, Thomson scattering, which is a non-resonant process. This can also produce interference patterns, which are of course different from the ones we have considered in this article. These nonresonant interference patterns have been entirely neglected in the present analysis. The scattering cross section[32] from atomic electrons in Fe is of the order of 40 b/sr in the forward direction and an order of magnitude less at 60°. The effective coherent cross section per crystal site for resonant nuclear scattering is about $3\chi^2 10^3$ b/sr for ^{57}Fe, where χ is the isotopic enrichment of resonant nuclei in the sample. For enriched samples, the nuclear cross section is orders of magnitude larger than that of the Thomson scattering, so that the contribution of electronic scattering can be neglected in a first approximation. It is well-known that the nuclear scattering cross section decreases with increasing energy. If the electronic interference pattern would be dominant in non-enriched samples, it would be required to use enriched samples if the nuclear cross section is too small.

V. CONCLUSION

The quantum mechanical theory, based on perturbation theory in frequency domain, of the formation of the interference pattern in the concept of nuclear emission holography using gamma radiation has been developed. Radiation emitted by a radioactive nucleus incorporated in a solid state lattice can go directly to a detector or it can scatter resonantly by neighboring nuclei in the ground state, before going to the detector. The two types of radiation interfere, which gives rise to oscillations, as a function of the emission direction, in the intensity reaching the detector. These oscillations contain information about the relative position of the nuclei involved in the scattering processes. For nuclei at a certain distance from the source nucleus, processes without recoil, the so-called Mössbauer effect, have to be considered. However, if the wavelengths associated to the relevant vibrational modes, which are excited due to processes with recoil, are large compared to the distance of the absorber nuclei with respect to the source nucleus, then processes with recoil can also give a contribution to the interference pattern. The physical origin of this effect would be the correlated movement of the source nucleus and the near-neighbor nuclei. Preliminary calculations show that these processes can be important for the holographic image. This would not only open the possibility to use nuclear isomers having higher transition energies then the ones used in Mössbauer spectroscopy but also it would open the possibility to study interference patterns using multiple energies within the same isotope, which could be used to suppress twin image effects as well as other aberrations and artifacts in reconstructed images. However, a final remark should be made here. The nuclear resonant cross section for gamma excitation decreases with the inverse of the wavelength of the gamma rays, which means that for small wavelengths, hence high energies, the probability for resonant scattering will be very small so that the probability for the formation of the holographic image will also be very small.

APPENDIX A

Let us calculate $\Sigma_{\vec{k}} 1/\omega - \omega_{\vec{k}} + i\varepsilon |K_{\vec{k},nucl}|^2$.

The sum over $\vec{k}$ can be transformed into an integral[24]

$$\sum_{\vec{k}} \bullet \to \frac{V}{(2\pi)^3} \int\int\int d^3\vec{k} \bullet \tag{A1}$$

It can be anticipated that the photon frequency $\omega_{\vec{k},phot}$ will be of the order of the nuclear transition energy, i.e., of the order of 10 keV or more. The phonon energies are of the order of meV, so that they can be safely neglected for the calculation of the integral. Then it can be shown[19] that $1/(\omega - \omega_{\vec{k}} + i\varepsilon)$ can be approximated by

$$\frac{1}{\omega - \omega_{\vec{k},phot} + i\varepsilon} = P \frac{1}{\omega - \omega_{\vec{k},phot}} - i\pi\delta(\omega - \omega_{\vec{k},phot}), \tag{A2}$$

where P represents the principal part of the integral over $\vec{k}$. Using Eqs. (A1) and (A2), we can write

$$\begin{aligned} &\sum_{\vec{k}} \frac{1}{\omega - \omega_{\vec{k}} + i\varepsilon} |K_{\vec{k},nucl}|^2 \\ &= P \frac{V}{(2\pi)^3} \int\int\int d^3\vec{k} \frac{1}{\omega - \omega_{\vec{k},phot}} |K_{\vec{k},nucl}|^2 \\ &\quad - i\pi \frac{V}{(2\pi)^3} \int\int\int d^3\vec{k} |K_{\vec{k},nucl}|^2 \delta(\omega - \omega_{\vec{k},phot}). \end{aligned} \tag{A3}$$

The principal part, which is a real number, is denoted $\Delta\omega_0$. The imaginary part is denoted $-i\gamma_R/2$, where the real quantity $\gamma_R/2$ is given by

$$\frac{\gamma_R}{2} = \pi \frac{V}{(2\pi)^3} \int\int\int d^3\vec{k} |K_{\vec{k},nucl}|^2 \delta(\omega - \omega_{\vec{k},phot}). \tag{A4}$$

The integral in the right-hand side of expression (A4) can be calculated and yields for γ_R

$$\gamma_R = \frac{V}{\pi c^3} |K_{\vec{k},nucl}(\omega)|^2 \omega^2. \tag{A5}$$

A comprehensive analysis performed by Heitler[19] shows that in order to evaluate the width and the shift ω can be replaced by $\omega_{0,nucl}$. This shows up only after going back to time domain for times such that $t \gg 1/\omega_{0,nucl}$. In the nuclear realm, $\omega_{0,nucl} \cong 10^{18}$ s^{-1} or larger, so that the condition would be $t \gg 10^{-18}$ s. In all practical applications, the time scales involved are much larger than this value, therefore the condition of constant width and shift is entirely justified. We then can write

$$\gamma_R = \frac{V}{\pi c^3} |K_{\vec{k},nucl}(\omega_{0,nucl})|^2 \omega_{0,nucl}{}^2, \tag{A6}$$

which is expression (28).

APPENDIX B

If we consider expression (41) and go back to time domain, there will be two integrals. The first integral is given by

$$c_{\vec{k},\{\beta_S\}}^{(0)}(t) = -\frac{1}{2\pi i}K_{\vec{k},nucl}^{*}\langle\{\beta_s\}|e^{-i\vec{k}\cdot\vec{u}_0}|\{\alpha_s\}\rangle \times\int_{-\infty}^{+\infty}\frac{e^{i(\omega_{\vec{k}}-\omega)t}}{\left(\omega-\omega_0+i\frac{\gamma_R}{2}\right)(\omega-\omega_0+i\varepsilon)}d\omega. \quad \text{(B1)}$$

This integral will be calculated using contour integration.

For $t<0$, the contour has to be closed by a semicircle in the upper half plane, where there are no poles, giving $c_{\vec{k},\{\beta_s\}}^{(0)}(t)=0$.

For $t>0$, the contour has to be closed in the lower half plane, where there are two poles. One has

$$\int_{-\infty}^{+\infty}\frac{e^{i(\omega_{\vec{k}}-\omega)t}}{\left(\omega-\omega_0+i\frac{\gamma_R}{2}\right)(\omega-\omega_0+i\varepsilon)}d\omega = -2\pi i\left[\frac{1-e^{i(\omega_{\vec{k}}-\omega_0+i\gamma_R/2)t}}{\omega_{\vec{k}}-\omega_0+i\frac{\gamma_R}{2}}\right]. \quad \text{(B2)}$$

Substituting expression (B2) into expression (B1) and using expressions (18) and (19), we have

$$c_{\vec{k},\{\beta_S\}}^{(0)}(t) = K_{\vec{k},nucl}^{*}\langle\{\beta_s\}|e^{-i\vec{k}\cdot\vec{u}_0}|\{\alpha_s\}\rangle \times\frac{1-e^{i(\omega_{\vec{k},phot}-\omega_{0,nucl}+\Omega(\beta_s)-\Omega(\alpha_s)+i\gamma_R/2)t}}{\omega_{\vec{k},phot}-\omega_{0,nucl}+\Omega(\beta_s)-\Omega(\alpha_s)+i\frac{\gamma_R}{2}}, \quad \text{(B3)}$$

which is expression (43).

The second integral, if we go back time domain, is explicitly given by

$$c_{\vec{k},\{\beta_s\}}^{(1)}(t) = \frac{1}{4\pi i}\sum_m K_{\vec{k},nucl}^{*}e^{-i\vec{k}\cdot\vec{r}_m}\langle\{\beta_s\}|e^{-i\vec{k}\cdot\vec{u}_m}|\{\alpha_s\}\rangle \times\langle\{\alpha_s\}|e^{i\vec{k}_0\cdot(\vec{u}_m-\vec{u}_0)}|\{\alpha_s\}\rangle \times\int_{-\infty}^{+\infty}\frac{\gamma_R}{\left(\omega-\omega_0+i\frac{\gamma_R}{2}\right)^2}\frac{e^{i(\omega_{\vec{k}}-\omega)t}}{(\omega-\omega_{\vec{k}}+i\varepsilon)} \times\frac{e^{i\omega r_m/c}}{\omega r_m/c}d\omega. \quad \text{(B4)}$$

The relevant integral is

$$I=\int_{-\infty}^{+\infty}\frac{\gamma_R}{\left(\omega-\omega_0+i\frac{\gamma_R}{2}\right)^2}\frac{e^{i(\omega_{\vec{k}}-\omega)t}}{(\omega-\omega_{\vec{k}}+i\varepsilon)}\frac{e^{i\omega r_m/c}}{\omega r_m/c}d\omega. \quad \text{(B5)}$$

It has to be mentioned that $\omega=0$ is not a pole because γ_R contains ω^2 in the numerator [see expression (A5)].

The real and imaginary parts of $1/[(\omega-\omega_0+i\gamma_R/2)^2]$ are strongly peaked functions of ω concentrated around $\omega=\omega_0$. So we can put $\gamma_R/\omega r_m/c$ in front of the integral and evaluate it for $\omega=\omega_{0,nucl}$, since $\Omega(\alpha_s)\ll\omega_{0,nucl}$. We then have

$$I=\frac{\gamma_R e^{i\omega_{\vec{k}}t}}{\omega_{0,nucl}r_m/c}\int_{-\infty}^{+\infty}\frac{e^{i(r_m/c-t)\omega}}{\left(\omega-\omega_0+i\frac{\gamma_R}{2}\right)^2(\omega-\omega_{\vec{k}}+i\varepsilon)}d\omega. \quad \text{(B6)}$$

This integral will be calculated again using contour integration.

For $r_m/c-t>0$, the contour has to be closed by a semicircle in the upper half plane, where there are no poles, so that the integral is zero in this case. This is again the principle of causality that states here that it takes a certain time before nucleus m, at distance r_m from the source nucleus, can be excited by radiation coming from the source nucleus.

For $r_m/c-t<0$, the contour has to be closed by a semicircle in the lower half plane, where there are two poles, one of first order and one of second order. Using the residue formula, one obtains after some algebra

$$I=-2\pi i\frac{\gamma_R}{\omega_{0,nucl}r_m/c}\left\{\frac{e^{i\omega_{\vec{k}}r_m/c}}{\left(\omega_{\vec{k}}-\omega_0+i\frac{\gamma_R}{2}\right)^2} + \left[i\left(\omega_{\vec{k}}-\omega_0+i\frac{\gamma_R}{2}\right)\left(\frac{r_m}{c}-t\right)-1\right]\frac{e^{i(r_m/c-t)(\omega_0-i\gamma_R/2)}}{\omega_{\vec{k}}-\omega_0+i\frac{\gamma_R}{2}}\right\}. \quad \text{(B7)}$$

Substituting Eq. (B7) into expression (B4) finally gives

$$c_{\vec{k},\{\beta_s\}}^{(1)}(t) = -\frac{\gamma_R}{2}\sum_m K_{\vec{k},nucl}^{*}e^{-i\vec{k}\cdot\vec{r}_m}\langle\{\beta_s\}|e^{-\vec{k}\cdot\vec{u}_m}|\{\alpha_s\}\rangle \times\langle\{\alpha_s\}|e^{i\vec{k}_0\cdot(\vec{u}_m-\vec{u}_0)}|\{\alpha_s\}\rangle \times\frac{1}{\omega_{0,nucl}r_m/c}\left\{\frac{e^{i\omega_{\vec{k}}r_m/c}}{\left(\omega_{\vec{k}}-\omega_0+i\frac{\gamma_R}{2}\right)^2} + \left[i\left(\omega_{\vec{k}}-\omega_0+i\frac{\gamma_R}{2}\right)\left(\frac{r_m}{c}-t\right)-1\right] \times\frac{e^{i(r_m/c-t)(\omega_0-i\gamma_R/2)}}{\omega_{\vec{k}}-\omega_0+i\frac{\gamma_R}{2}}\right\}. \quad \text{(B8)}$$

This is expression (44).

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