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Quantum mechanical theory of the formation of a nuclear emission hologram

J. Odeurs, R. Coussement, and C. L'abbé
Instituut voor Kern- en Stralingsfysica, Katholieke Universiteit Leuven, Celestijnenlaan 200 D, B-3001 Leuven, Belgium

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The quantum mechanical theory of the formation of an emission hologram with γ radiation is developed. Radiation, produced by a radioactive source nucleus, can go directly to a detector or can be resonantly scattered by neighboring resonant nuclei 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. These fluctuations contain information about the surrounding of the emitting nucleus. The coupled system of equations describing the scattering amplitudes has been solved in the single scattering approximation. [S0163-1829(99)02733-2]

I. INTRODUCTION

The idea to use γ radiation for holography with atomic resolution has been suggested recently.[1,2] The scheme is simple, in principle: a photon emitted without recoil (Mössbauer effect) can reach a (far-field) detector directly (this is the holographic reference wave) or after it has been scattered from closely situated nuclei (the object wave). The two waves interfere and form the holographic image. Measuring the intensity as a function of the angle of the emitted radiation gives information on the relative position of the neighboring nuclei. In a recent experiment[3] nuclear holography has been presented in a slightly different way: a photon from an external radioactive source can be resonantly absorbed directly by a nucleus in a crystal or, indirectly, after the photon has been resonantly scattered by the neighboring nuclei. The interference of these processes gives oscillations of the total number of deexcitation events measured as a function of the incidence angle of the photon. These holographic oscillations give information on the local environment of the nucleus. In this paper we present the theoretical analysis of ''internal'' or nuclear emission holography, as originally proposed in Refs. 1 and 2.

In Sec. II the mathematical background will be defined. In Sec. III the details of the analysis will be given. In Sec. IV the discussion of the results will be presented.

II. MATHEMATICAL BACKGROUND

The general method used in this paper is discussed in Heitler,[4] Harris,[5] and in a recent article.[6] The method applies quantum-mechanical perturbation theory in the frequency domain to obtain a set of coupled equations. The Hamiltonian of the system is divided into two parts. H_0 is the unperturbed part which describes the evolution of the nuclear states and the free radiation field in the absence of coupling between the nuclear states and the radiation field. The eigenstates of H_0 correspond in this case to nuclear states of an ensemble of nuclei and the states of the free radiation field taken here as plane waves. Any nucleus in an excited state can be located at any one of the nuclear positions in the medium. The perturbing part of the Hamiltonian is denoted by V and is responsible for making transitions between the nuclear levels.

The actual state of the system is then expressed as

$$|\Psi(t)\rangle=\sum_l a_l(t)e^{-i(E_l t/\hbar)}|\varphi_l(0)\rangle, \tag{1}$$

where $|\varphi_l(0)\rangle$ is an eigenstate of H_0 and E_l the corresponding energy. Solving the Schrödinger equation leads to a set of coupled differential equations relating the expansion coefficients $a_l(t)$

$$i\hbar\frac{da_l}{dt}=\sum_q a_q(t)e^{i(\omega_l-\omega_q)t}\langle\varphi_l(0)|V|\varphi_q(0)\rangle, \tag{2}$$

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

A solution of this system of coupled differential equations is wanted that satisfies an initial condition such that at $t=0$ the system is in a well-defined state, say n, and all other probability amplitudes are zero: $a_l(0)=0$ and $a_n(+0)=1$, where $t=+0$ means that t approaches zero from the positive side. Although a physically meaningful solution only involves positive times ($t\geqslant 0$), for analytical reasons however, following Heitler,[4] the solution will be extended to the negative time axis.

We choose the a_l's such that $a_l(t)=a_n(t)=0$ for $t<0$. It follows then that a_n has a discontinuity that can be dealt with.[4] Heitler has shown that adding an inhomogeneous term to the right hand side of expression (2) takes care of the initial condition and the discontinuity completely

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

where δ_{ln} is the Kronecker delta and $\delta(t)$ the Dirac delta function. Next introducing the Fourier transform[4]

$$a_l(t)=-\frac{1}{2\pi i}\int_{-\infty}^{\infty}d\omega A_l(\omega)e^{i(\omega_l-\omega)t}, \tag{4}$$

Eq. (2) can be rewritten in the frequency domain

$$(\omega-\omega_l)A_l(\omega)=\sum_q A_q(\omega)\frac{V_{lq}}{\hbar}+\delta_{ln}, \tag{5}$$

where V_{lq} is the matrix element inducing a transition from the qth unperturbed state to the lth unperturbed state, $V_{lq} = \langle \varphi_l(0) | V | \varphi_q(0) \rangle$. The integral representation of the Dirac delta function has been used.[4] To obtain an equation for $A_l(\omega)$, we would have to divide by $(\omega - \omega_l)$. This division will not be unique[4] and it can be shown that if the a_l's are to fulfill the initial conditions the result of the division by $(\omega - \omega_l)$ must be a factor $\lim_{\varepsilon \to 0^+} 1/(\omega - \omega_l + i\varepsilon)$. This has only a mathematical meaning when integrals are involved, which eventually is always the case (see later). In fact the replacement of $(\omega - \omega_l)$ by $(\omega - \omega_l + i\varepsilon)$ (ε an infinitesimal, positive number) defines the path of integration, guaranteeing causality.[4] This is completely analogous to the definition of integration paths in the propagator concept in quantum electrodynamics. Of course the device $i\varepsilon$ will always disappear from the physical answers, as will become clear later. So Eq. (5) will be rewritten as

$$(\omega - \omega_l + i\varepsilon) A_l(\omega) = \sum_q A_q(\omega) \frac{V_{lq}}{\hbar} + \delta_{ln}. \tag{6}$$

The advantage of the set of equations, Eq. (6), is that it is a linear (coupled) system. In the next section this general formalism will be applied to the study of the interaction of radiation with nuclei embedded in a lattice.

III. ANALYSIS

A. Fundamental equations

Let us consider, at $t=0$, an excited nucleus, the "source" nucleus, surrounded by identical ground state nuclei, the "scattering" nuclei, and no photons or conversion electrons present. The source nucleus is taken at the origin of a coordinate system. The coordinate of the scattering nucleus m is denoted $\mathbf{r}_m$. The different quantum mechanical amplitudes are defined below.

$A(\omega)$: the amplitude of the source nucleus excited, all scattering nuclei in their ground states, no photons or conversion electrons present.

$B_m(\omega)$: the amplitude corresponding to excitation of the mth scattering nucleus to one of its excited states $\hbar\omega_0$ and no photons or conversion electrons present. To keep the analysis as simple as possible, just one excited state will be considered. This can be generalized.

$C_{\mathbf{k}}(\omega)$: the amplitude where all nuclei are in the ground state, there are no conversion electrons, and a photon of wave vector $\mathbf{k}$ is present.

$D_{\mathbf{p}}(\omega)$: the amplitude corresponding to the presence of a conversion electron with momentum $\mathbf{p}$, produced by the source nucleus, all nuclei in the ground state and no photons present.

$D_{m,\mathbf{p}}(\omega)$: the amplitude corresponding to the presence of a conversion electron, with momentum $\mathbf{p}$, from the mth nucleus, all nuclei in their ground states and no photons present.

The coupled equations relating these amplitudes can then be shown to be

$$(\omega - \omega_0 + i\varepsilon) A(\omega) = \sum_{\mathbf{k}} \frac{V_{\mathbf{k}}}{\hbar} C_{\mathbf{k}}(\omega) + \sum_{\mathbf{p}} \frac{V_{\mathbf{p}}}{\hbar} D_{\mathbf{p}}(\omega) + 1, \tag{7}$$

$$(\omega - \omega_{\mathbf{p}} + i\varepsilon) D_{\mathbf{p}}(\omega) = \frac{V_{\mathbf{p}}^*}{\hbar} A(\omega), \tag{8}$$

$$(\omega - \omega_0 + i\varepsilon) B_m(\omega) = \sum_{\mathbf{k}} \frac{V_{\mathbf{k}}}{\hbar} e^{i\mathbf{k}\cdot\mathbf{r}_m} C_{\mathbf{k}}(\omega) + \sum_{\mathbf{p}} \frac{V_{\mathbf{p}}}{\hbar} e^{i\mathbf{p}\cdot\mathbf{r}_m/\hbar} D_{m,\mathbf{p}}(\omega), \tag{9}$$

$$(\omega - \omega_{\mathbf{p}} + i\varepsilon) D_{m,\mathbf{p}}(\omega) = \frac{V_{\mathbf{p}}^*}{\hbar} e^{-i\mathbf{p}\cdot\mathbf{r}_m/\hbar} B_m(\omega), \tag{10}$$

$$(\omega - \omega_{\mathbf{k}} + i\varepsilon) C_{\mathbf{k}}(\omega) = \frac{V_{\mathbf{k}}^*}{\hbar} A(\omega) + \sum_m \frac{V_{\mathbf{k}}^*}{\hbar} e^{-i\mathbf{k}\cdot\mathbf{r}_m} B_m(\omega), \tag{11}$$

where $V_{\mathbf{k}}$ and $V_{\mathbf{k}}^*$ are the matrix elements describing, respectively, absorption and emission of a photon with wave vector $\mathbf{k}$. Analogous definitions and notations hold for the matrix elements, $V_{\mathbf{p}}$ and $V_{\mathbf{p}}^*$, describing electron conversion processes, with $\mathbf{p}$ the momentum of the conversion electron.

An understanding of the structure of these equations can be obtained by considering Eq. (11). Equation (11) expresses the amplitude for finding a photon present, $C_{\mathbf{k}}(\omega)$. The first term on the right-hand side of Eq. (11) corresponds to having the source nucleus emit such a photon. The sum over m on the right-hand side of Eq. (11) corresponds to the emission of a photon by the scattering nuclei. Since each nucleus can do this we must sum over all resonant scattering nuclei. One must keep track of where that emission took place by introducing the appropriate phase factor. An analogous interpretation can be done for all other processes. It should be emphasized at this stage that the treatment of the electron conversion processes is only necessary to produce a width (and also a shift that will be incorporated into ω_0) due to electron conversion which, combined with the radiative width (see later), will give the total width of the nuclear excited state. This has been explained in detail in Appendix A. Combining Eqs. (7)–(11) it has been shown in Appendix A that

$$\left(\omega - \omega_0 + i\frac{\Gamma}{2\hbar}\right) A(\omega) = 1 + \sum_m \sum_{\mathbf{k}} \frac{1}{\omega - \omega_{\mathbf{k}} + i\varepsilon} \frac{|V_{\mathbf{k}}|^2}{\hbar^2} e^{-i\mathbf{k}\cdot\mathbf{r}_m} B_m(\omega), \tag{12}$$

where Γ is the total width of the nuclear excited state, given by the sum of the electron conversion width γ_c, and the radiative width γ_R, defined by Eqs. (A5) and (A9), respectively. ω_0 incorporates a level shift coming from the summation over $\mathbf{k}$ in Eq. (7). This is explained in Appendix A.

It has been shown also in Appendix A that

$$\left(\omega-\omega_0+i\frac{\Gamma}{2\hbar}\right)B_m(\omega)=A(\omega)\sum_{\mathbf{k}}\frac{1}{\omega-\omega_{\mathbf{k}}+i\varepsilon}\frac{|V_{\mathbf{k}}|^2}{\hbar^2}e^{i\mathbf{k}\cdot\mathbf{r}_m}+\sum_{\mathbf{k}}\sum_{m'\neq m}B_{m'}(\omega)\frac{1}{\omega-\omega_{\mathbf{k}}+i\varepsilon}\frac{|V_{\mathbf{k}}|^2}{\hbar^2}e^{i\mathbf{k}\cdot(\mathbf{r}_m-\mathbf{r}_{m'})}. \tag{13}$$

Equations (11), (12), and (13) are the fundamental equations describing multiple scattering in the frequency domain. The positions of all resonant nuclei occur in the expressions.

The system of equations has been solved exactly in the case of radiation coming from a radioactive source,[6] scattered in the forward direction. In the following we will use another approach, where an approximate solution will be presented. This solution will lead to the idea of holography with γ radiation.

B. Approximate solution

Hypotheses. (i) The probability of the source nucleus reexcited by radiation coming from the neighboring nuclei is negligible. This can be justified for internuclear distances >1 Å, which is always the case. (ii) Single scattering by the surrounding nuclei. This will be an excellent approximation for thin samples. In fact, both hypotheses constitute the single scattering approximation.

The sums over m and $\mathbf{k}$ in the right-hand side of Eq. (12) describe absorption by the source nucleus of radiation coming from the scattering nuclei. According to hypothesis (i), this process will be neglected. Therefore,

$$A(\omega)=\frac{1}{\omega-\omega_0+i(\Gamma/2\hbar)}. \tag{14}$$

This is nothing but the familiar (Lorentzian) frequency spectrum centered around ω_0 with width $\Gamma/2\hbar$.

The last series on the right-hand side of Eq. (13) describes the excitation of scattering nucleus at position $\mathbf{r}_m$ due to radiation coming from the other scattering nuclei. These processes are neglected according to hypothesis (ii). Therefore,

$$\left(\omega-\omega_0+i\frac{\Gamma}{2\hbar}\right)B_m(\omega)=A(\omega)\sum_{\mathbf{k}}\frac{1}{\omega-\omega_{\mathbf{k}}+i\varepsilon}\frac{|V_{\mathbf{k}}|^2}{\hbar^2}e^{i\mathbf{k}\cdot\mathbf{r}_m} \tag{15}$$

or with Eq. (14)

$$\left(\omega-\omega_0+i\frac{\Gamma}{2\hbar}\right)B_m(\omega)=\frac{1}{\omega-\omega_0+i(\Gamma/2\hbar)}\sum_{\mathbf{k}}\frac{1}{\omega-\omega_{\mathbf{k}}+i\varepsilon}\frac{|V_{\mathbf{k}}|^2}{\hbar^2}e^{i\mathbf{k}\cdot\mathbf{r}_m}. \tag{16}$$

Making use of Eqs. (11) and (16), $C_{\mathbf{k}}(\omega)$ and, after going back to time domain, $c_{\mathbf{k}}(t)$ can be calculated explicitly. This has been done in Appendix B. One finds

$$\begin{aligned}c_{\mathbf{k}}(t)=&\frac{V_{\mathbf{k}}^*}{\hbar}\frac{1}{\omega_{\mathbf{k}}-\omega_0+i(\Gamma/2\hbar)}(1-e^{i(\omega_{\mathbf{k}}-\omega_0+i\Gamma/2\hbar)t})\\&-\frac{V_{\mathbf{k}}^*}{2\hbar^2}\frac{\gamma_R}{[\omega_{\mathbf{k}}-\omega_0+i(\Gamma/2\hbar)]^2}\sum_m e^{-i\mathbf{k}\cdot\mathbf{r}_m}\\&\times\frac{e^{i\omega_{\mathbf{k}}r_m/c}}{\omega_0 r_m/c}\{1+[i(\omega_{\mathbf{k}}-\omega_0+i\Gamma/2\hbar)\\&\times(t-r_m/c)e^{i(\omega_{\mathbf{k}}-\omega_0+i\Gamma/2\hbar)(t-r_m/c)}\\&-e^{i(\omega_{\mathbf{k}}-\omega_0+i\Gamma/2\hbar)(t-r_m/c)}]\}.\end{aligned} \tag{17}$$

The total probability amplitude for having a photon with wave vector $\mathbf{k}$ is the sum of two terms

$$c_{\mathbf{k}}(t)=c_{\mathbf{k}}^{(0)}(t)+c_{\mathbf{k}}^{(1)}(t) \tag{18}$$

with

$$c_{\mathbf{k}}^{(0)}(t)=\frac{V_{\mathbf{k}}^*}{\hbar}\frac{1}{\omega_{\mathbf{k}}-\omega_0+i\frac{\Gamma}{2\hbar}}(1-e^{i(\omega_{\mathbf{k}}-\omega_0+i\Gamma/2\hbar)t}) \tag{19}$$

and

$$\begin{aligned}c_{\mathbf{k}}^{(1)}(t)=&-\frac{V_{\mathbf{k}}^*}{2\hbar^2}\frac{\gamma_R}{[\omega_{\mathbf{k}}-\omega_0+i(\Gamma/2\hbar)]^2}\sum_m e^{-i\mathbf{k}\cdot\mathbf{r}_m}\frac{e^{i\omega_{\mathbf{k}}r_m/c}}{\omega_0 r_m/c}\\&\times\{1+[i(\omega_{\mathbf{k}}-\omega_0+i\Gamma/2\hbar)(t-r_m/c)\\&\times e^{i(\omega_{\mathbf{k}}-\omega_0+i\Gamma/2\hbar)(t-r_m/c)}\\&-e^{i(\omega_{\mathbf{k}}-\omega_0+i\Gamma/2\hbar)(t-r_m/c)}]\}.\end{aligned} \tag{20}$$

$c_{\mathbf{k}}^{(0)}(t)$ is the amplitude to have a photon with wave vector $\mathbf{k}$ produced by the source nucleus alone. $c_{\mathbf{k}}^{(1)}(t)$ is the amplitude to have a photon with wave vector $\mathbf{k}$ due to scattering nuclei, having scattered the photon produced by the source nucleus. In the next section, the radiated intensity will be calculated with the aid of expressions (19) and (20).

IV. RADIATED INTENSITY AND DISCUSSION

A. Radiated intensity

The probability of having a photon with wave vector $\mathbf{k}$ present at time t, is given by

$$\begin{aligned}P_{\mathbf{k}}(t)=|c_{\mathbf{k}}(t)|^2=&|c_{\mathbf{k}}^{(0)}(t)|^2+|c_{\mathbf{k}}^{(1)}(t)|^2\\&+2\,\mathrm{Re}[c_{\mathbf{k}}^{(0)}(t)c_{\mathbf{k}}^{(1)*}(t)].\end{aligned} \tag{21}$$

The first term $|c_{\mathbf{k}}^{(0)}(t)|^2$ is the probability to have a photon with wave vector $\mathbf{k}$, due to the presence of the source nucleus alone. The second term $|c_{\mathbf{k}}^{(1)}(t)|^2$ is the probability to have a photon with wave vector $\mathbf{k}$, due to the presence of the scattering nuclei, having scattered the photon produced by the source nucleus. For a small number of scattering nuclei, this term is much smaller then $|c_{\mathbf{k}}^{(0)}(t)|^2$ so that it can be neglected. The last term on the right-hand side of expression (21) is an interference term between the amplitude due to the source nucleus and all amplitudes due to the scattering nuclei.

The interference term $I_{\mathbf{k}}^{01}(t)$ is given explicitly by

$$I_{\mathbf{k}}^{01}(t)=-\mathrm{Re}\left\{\frac{|V_{\mathbf{k}}|^2\gamma_R}{\hbar^3}\frac{1}{\omega_{\mathbf{k}}-\omega_0+i(\Gamma/2\hbar)}(1-e^{i(\omega_{\mathbf{k}}-\omega_0+i\Gamma/2\hbar)t})\frac{1}{[\omega_{\mathbf{k}}-\omega_0-i(\Gamma/2\hbar)]^2}\sum_m e^{i\mathbf{k}\cdot\mathbf{r}_m}\frac{e^{-i\omega_{\mathbf{k}}r_m/c}}{\omega_0 r_m/c}\right.$$
$$\left.\times\{1+[-i(\omega_{\mathbf{k}}-\omega_0-i\Gamma/2\hbar)(t-r_m/c)e^{-i(\omega_{\mathbf{k}}-\omega_0-i\Gamma/2\hbar)(t-r_m/c)}-e^{-i(\omega_{\mathbf{k}}-\omega_0-i\Gamma/2\hbar)(t-r_m/c)}]\}\right\}. \quad (22)$$

It is clear from expression (22) that the count rate depends on the direction, this because of the presence of the factors $e^{i\mathbf{k}\cdot\mathbf{r}_m}$. The count rate will thus change as a function of the emission direction. Expression (22) contains therefore information on the position of the scattering nuclei with respect to the source nucleus.

We will consider the probability that a photon has been emitted in direction $\mathbf{k}$ for long times ($t\rightarrow\infty$). The interference term for long times becomes

$$I_{\mathbf{k}}^{01}(\infty)=-\mathrm{Re}\left\{\frac{|V_{\mathbf{k}}|^2\gamma_R}{\hbar^3}\frac{1}{\omega_{\mathbf{k}}-\omega_0+i(\Gamma/2\hbar)}\right.$$
$$\left.\times\frac{1}{[\omega_{\mathbf{k}}-\omega_0-i(\Gamma/2\hbar)]^2}\sum_m e^{i\mathbf{k}\cdot\mathbf{r}_m}\frac{e^{-i\omega_{\mathbf{k}}r_m/c}}{\omega_0 r_m/c}\right\}. \quad (23)$$

At resonance, it can be shown that

$$I_{\mathbf{k}}^{01}(\infty)=\frac{8|V_{\mathbf{k}}|^2\gamma_R}{\Gamma^3}\sum_m\frac{\sin(\mathbf{k}\cdot\mathbf{r}_m-\omega_0 r_m/c)}{\omega_0 r_m/c}. \quad (24)$$

The contrast function $F_{\mathbf{k}}$ (still corresponding to the resonance condition), is given by the ratio of the interference term $I_{\mathbf{k}}^{01}(\infty)$ given by expression (24), and the square of the absolute value of expression (19), evaluated at resonance and for long times. One has

$$F_{\mathbf{k}}=\frac{I_{\mathbf{k}}^{01}(\infty)}{|c_{\mathbf{k}}^{(0)}(\infty)|^2}=2\frac{\gamma_R}{\Gamma}\sum_m\frac{\sin\left(\mathbf{k}\cdot\mathbf{r}_m-\frac{\omega_0 r_m}{c}\right)}{\omega_0 r_m/c}. \quad (25)$$

The detector, positioned in a certain direction, does not register a single energy, of course. One has to integrate the interference term, Eq. (23), and the direct term over all photon energies. When doing so, the new contrast function $F_{\mathbf{k}}$ will depend solely on the direction, given by the unit vector $\hat{k}=\mathbf{k}/k$. The explicit expression results in

$$F_{\hat{k}}=-\frac{\gamma_R}{\Gamma}\sum_m e^{-(1-\hat{k}\cdot\hat{r}_m)\Gamma r_m/2\hbar c}\frac{\sin[(1-\hat{k}\cdot\hat{r}_m)\omega_0 r_m/c]}{\omega_0 r_m/c}, \quad (26)$$

where $\hat{r}_m$ is a unit vector in the direction $\mathbf{r}_m$. It can be verified easily that $e^{-(1-\hat{k}\cdot\hat{r}_m)\Gamma r_m/2\hbar c}\approx 1$, so that finally one has

$$F_{\hat{k}}=-\frac{\gamma_R}{\Gamma}\sum_m\frac{\sin[(1-\hat{k}\cdot\hat{r}_m)\omega_0 r_m/c]}{\omega_0 r_m/c}. \quad (27)$$

This contrast function has similar features as the one for an x-ray fluorescence hologram[7] or photoelectron hologram.[8] In the next section this function will be discussed.

B. Discussion

The contrast function $F_{\hat{k}}$ depends evidently on the direction $\hat{k}$ with respect to the positions $\mathbf{r}_m$ of the nuclei. Measuring as a function of the direction will thus give information on the positions of the nuclei with respect to the source nucleus, whence the term nuclear hologram. Superimposed on a constant background, due to the direct radiation of the source nucleus, there will be fluctuations in the measured intensity. The reconstruction of the real image from the radiated intensity can be done according to certain techniques (see, e.g., Ref. 9).

The order of magnitude of the ‘‘contrast’’ as well as of the ‘‘oscillations’’ as a function of the emission direction, will be estimated for one scattering nucleus at a distance of 3 Å from the source nucleus. For the ratio γ_R/Γ we take $\frac{1}{10}$. The energy is taken as 14.4 keV (^{57}Fe nucleus). A simple calculation shows that the contrast, defined here as the difference between the maximum and the minimum value of the ratio given by expression (27), is of the order of 1%. Although expression (27), applied to the simple case of one source nucleus and one scattering nucleus, is not really a periodic function, it shows a series of (nonequidistant) maxima and minima. Figure 1 shows the simulated contrast for this simple case. The angular distance between a maximum and its adjacent minimum is about 8° for radiation emitted in the vicinity of 90° with respect to the line joining source and scattering nucleus. For smaller or larger angles, this angular distance increases. For example, a maximum occurs around 37° and its adjacent minimum around 20°.

Figure 2 displays the intensity pattern of the 14.4 keV radiation produced by an excited ^{57}Fe nucleus at a center of a cube (of length 2.866 Å) surrounded by eight ground state ^{57}Fe nuclei at the corners of the cube. The intensity is shown as a function of the spherical angular coordinates θ and φ, where the axes are defined in Fig. 2. The spots with bright intensity correspond to directions with high radiated intensity and, inversely, the darker a spot, the lower the intensity corresponding to this direction. For any specific configuration, the contrast function can be easily evaluated numerically.

Experiments can be performed with the use of a position-sensitive detector. Given the numerical values of the simulation displayed in Fig. 2, a rough estimate of the needed angular resolution can be made for the cluster of nuclei presented in Fig. 2. The angular distance between a maximum and a minimum is of the order of 10°. If we choose for the detection a pixel definition of one tenth of this, we would need an angular resolution of 1°, which is easily feasible. For

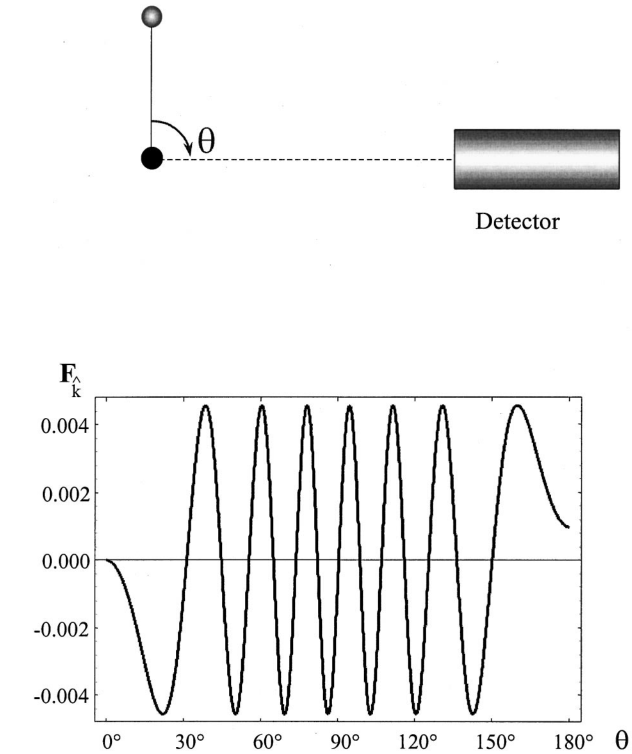

FIG. 1. Contrast function [Eq. (27)] of the 14.4 keV radiation produced by an excited ^{57}Fe nucleus at which neighborhood a ground state ^{57}Fe nucleus is situated at 3 Å. θ is the angle between the line defined by the source and scattering nuclei and the line defined by the source nucleus and the (far away) detector.

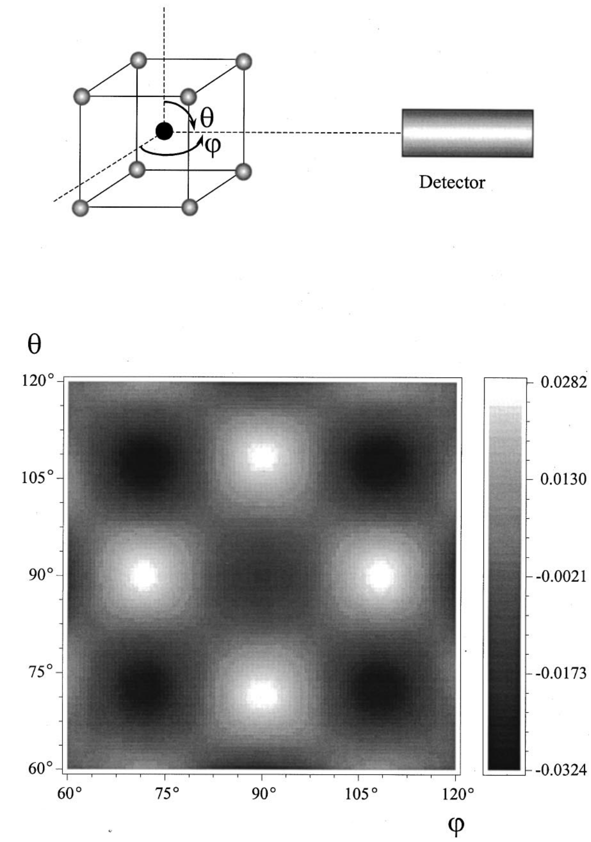

FIG. 2. Intensity pattern of radiation produced by an excited nucleus on a substitutional site in a bcc cell with lattice constant 2.866 Å, surrounded by eight ground state nuclei.

a microcurie γ source a total counting time of the order of a few days would already be sufficient.

Finally, it has to be mentioned that a real holographic image will not look as simple as the one we considered in Fig. 2, this due to the contribution of nuclei situated further away from the source nuclei. It can be seen from expression (27) that the further the nuclei are away from the source nucleus, the higher the frequencies of the oscillations corresponding to these nuclei are. Applying a low-pass filtering procedure on the contrast function, one can filter out the high frequency oscillations, and thus keep only the contributions of the near-neighbor atoms.

V. CONCLUSIONS

Radiation emitted by a radioactive nucleus incorporated in a solid state lattice can go directly to a (far-field) 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 in the intensity reaching the detector if measurements are performed as a function of the emission angle. These oscillations contain information about the relative position of the nuclei involved in the scattering processes. This is the idea of holography with nuclear radiation. The direct radiation is the reference wave and the scattered radiation is the object wave in the holography. The real space reconstruction has to be performed. The quantum mechanical theory of the scattering processes has been developed using perturbation theory in the frequency domain. Allowing for only single resonant scattering, the system of equations can be solved exactly. We have developed the theory for a single line source. The extension to the case where there is hyperfine splitting has still to be done. Nuclear emission holography has the advantage that it could be applied to study structures that are too small for the usual x-ray diffraction.

To obtain a single hologram, some conditions have to be fulfilled:[11] the environment of every source nucleus has to be the same, every environment has to be oriented in the same way, the size of the sample has to be much smaller than the distance sample-detector, the radiation emitted by different source nuclei has to be incoherent (this is always the case for radioactive nuclei). The problem of sample preparation for nuclear emission holography is not trivial. Nuclear emission holography is only applicable if enriched single crystalline samples are available. The most useful radioactive isotope for emission holography is probably ^{57}Fe. The problem of sample preparation is the introduction of ^{57}Co (which decays to ^{57}Fe) into a sample containing ^{57}Fe in the ground state. Ion implantation of ^{57}Co into a previously prepared enriched sample of ^{57}Fe is a possibility. Co-deposition of ^{57}Co and ^{57}Fe by means of special techniques such as molecular beam epitaxy (e.g., at the ion and molecular beam laboratory[12] of the University of Leuven) is another possibility.

As already has been mentioned, the crucial part is the ^{57}Fe environment of every ^{57}Co. If all environments are identical and oriented the same way, then separate but identical holograms are added. If there exist a small number of inequivalent source sites, then the hologram will be a superposition of the neighboring environments surrounding each source nucleus. If the environments have completely random orientations with respect to each other, the structure in the hologram will disappear.

If the problem of sample preparation is solved, the specific applications of nuclear emission holography could be the study of nuclei on a surface or of nuclei belonging to clusters or precipitates. A final word could be said about the influence of Thomson electronic scattering. The Thomson scattering occurs not only for the scattering nuclei, but also for the atoms of the matrix. The Thomson cross section[4] is of the order of 6×10^{-25} cm^2, while the resonant nuclear cross section for ^{57}Fe is 2.56×10^{-18} cm^2. If the concentration of the resonant scattering nuclei is low (as is the case for nonenriched samples), the Thomson scattering will increase the background, reducing the effect of the holographic oscillations. For enriched, thin samples, Thomson scattering will be negligible because of the large difference between the resonant nuclear and Thomson cross sections.

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APPENDIX A

The general Eqs. (12) and (13) will be derived in this appendix, starting from Eqs. (7)–(11). Substituting Eq. (8) in Eq. (7)

$$(\omega-\omega_0+i\varepsilon)A(\omega)=\sum_{\mathbf{k}}\frac{V_{\mathbf{k}}}{\hbar}C_{\mathbf{k}}(\omega)+\sum_{\mathbf{p}}\frac{1}{\omega-\omega_{\mathbf{p}}+i\varepsilon}\frac{|V_{\mathbf{p}}|^2}{\hbar^2}A(\omega)+1. \tag{A1}$$

Analogously, with Eqs. (9) and (10), one has

$$(\omega-\omega_0+i\varepsilon)B_m(\omega)=\sum_{\mathbf{k}}\frac{V_{\mathbf{k}}}{\hbar}e^{i\mathbf{k}\cdot\mathbf{r}_m}C_{\mathbf{k}}(\omega)+\sum_{\mathbf{p}}\frac{1}{\omega-\omega_{\mathbf{p}}+i\varepsilon}\frac{|V_{\mathbf{p}}|^2}{\hbar^2}B_m(\omega). \tag{A2}$$

By converting the sums on $\mathbf{p}$ in Eqs. (A1) and (A2) into an integral[4] and expressing $|V_{\mathbf{p}}|^2/\hbar^2(\omega-\omega_{\mathbf{p}}+i\varepsilon)$ in terms of a principal part and a delta function, according to the relation[4]

$$\frac{1}{x+i\varepsilon}=P\frac{1}{x}-i\pi\delta(x) \tag{A3}$$

one has

$$\sum_{\mathbf{p}}\frac{|V_{\mathbf{p}}|^2}{\hbar^2(\omega-\omega_{\mathbf{p}}+i\varepsilon)}A(\omega)=\frac{V}{(2\pi\hbar)^3}\frac{1}{\hbar^2}P\int\int\int\frac{|V_{\mathbf{p}}|^2}{\omega-\omega_{\mathbf{p}}}p^2dp\,d\Omega A(\omega)-i\times\frac{V}{(2\pi\hbar)^3}\frac{\pi}{\hbar^2}\int\int\int|V_{\mathbf{p}}|^2p^2\delta(\omega-\omega_{\mathbf{p}})dp\,d\Omega A(\omega) \tag{A4}$$

and an analogous expression for $B_m(\omega)$. P stands for the principal value of the integral. The presence of the volume V in Eq. (A4) and in the others resulting from the conversion of a sum into an integral in three dimensions, is only apparent, because the matrix elements such as $|V_{\mathbf{p}}|^2$ contain[10] $1/V$.

When the expression in Eq. (A4) is taken to the left-hand side of Eq. (A1), the principal value term corresponds to a shift in the frequency, which can be incorporated into ω_0. The second term of Eq. (A4) gives a width due to the interaction of a nucleus with its conversion electron. This width γ_c is defined by

$$\gamma_c=\frac{2\pi V}{(2\pi\hbar)^3\hbar}\int\int\int|V_{\mathbf{p}}|^2p^2\delta(\omega-\omega_{\mathbf{p}})dp\,d\Omega. \tag{A5}$$

Rewriting Eq. (A1) gives then

$$\left(\omega-\omega_0+i\frac{\gamma_c}{2\hbar}\right)A(\omega)=\sum_{\mathbf{k}}\frac{V_{\mathbf{k}}}{\hbar}C_{\mathbf{k}}(\omega)+1. \tag{A6}$$

The corresponding equation for $B_m(\omega)$ is

$$\left(\omega-\omega_0+i\frac{\gamma_c}{2\hbar}\right)B_m(\omega)=\sum_{\mathbf{k}}\frac{V_{\mathbf{k}}}{\hbar}e^{i\mathbf{k}\cdot\mathbf{r}_m}C_{\mathbf{k}}(\omega). \tag{A7}$$

Solving now Eq. (11) of the main text for $C_{\mathbf{k}}(\omega)$ and substituting into Eq. (A6) gives

$$\left(\omega-\omega_0+i\frac{\gamma_c}{2\hbar}\right)A(\omega)=\sum_{\mathbf{k}}\frac{1}{\omega-\omega_{\mathbf{k}}+i\varepsilon}\frac{|V_{\mathbf{k}}|^2}{\hbar^2}A(\omega)+\sum_m\sum_{\mathbf{k}}\frac{1}{\omega-\omega_{\mathbf{k}}+i\varepsilon}\frac{|V_{\mathbf{k}}|^2}{\hbar^2}\times e^{-i\mathbf{k}\cdot\mathbf{r}_m}B_m(\omega)+1. \tag{A8}$$

Considering the first term on the right-hand side of Eq. (A8), the sum on $\mathbf{k}$ can be converted into an integral, analogously to what has been done before. This results again in a principal value term and a delta-function term. The principal value term corresponds again to a frequency shift when brought to the left-hand side. The delta function term corresponds to the usual radiative width[13] γ_R, where

$$\gamma_R=\frac{2\pi V}{(2\pi)^3\hbar}\int\int\int|V_{\mathbf{k}}|^2\delta(\omega-\omega_{\mathbf{k}})k^2dk\,d\Omega. \tag{A9}$$

Collecting terms on the left-hand side of Eq. (A8), using Eq. (A9), gives

$$\left(\omega-\omega_0+i\frac{\Gamma}{2\hbar}\right)A(\omega)=1+\sum_m\sum_{\mathbf{k}}\frac{1}{\omega-\omega_{\mathbf{k}}+i\varepsilon}\frac{|V_{\mathbf{k}}|^2}{\hbar^2}e^{-i\mathbf{k}\cdot\mathbf{r}_m}B_m(\omega)\tag{A10}$$

which is Eq. (12) of the main text.

Γ is the total width, equal to the sum of the conversion-electron and radiative widths. Solving again Eq. (11) of the main text for $C_{\mathbf{k}}(\omega)$ and substituting into Eq. (A7) gives

$$\left(\omega-\omega_0+i\frac{\gamma_c}{2\hbar}\right)B_m(\omega)=A(\omega)\sum_{\mathbf{k}}\frac{1}{\omega-\omega_{\mathbf{k}}+i\varepsilon}\frac{|V_{\mathbf{k}}|^2}{\hbar^2}e^{i\mathbf{k}\cdot\mathbf{r}_m}+\sum_{\mathbf{k}}\sum_{m'}B_{m'}(\omega)\frac{1}{\omega-\omega_{\mathbf{k}}+i\varepsilon}\times\frac{|V_{\mathbf{k}}|^2}{\hbar^2}e^{i\mathbf{k}\cdot(\mathbf{r}_m-\mathbf{r}_{m'})}.\tag{A11}$$

The second series of the right-hand side of Eq. (A11) can be divided in two parts: one with $m'\neq m$ and the other with $m'=m$. Then Eq. (A11) can be rewritten as

$$\left(\omega-\omega_0+i\frac{\gamma_c}{2\hbar}\right)B_m(\omega)=A(\omega)\sum_{\mathbf{k}}\frac{1}{\omega-\omega_{\mathbf{k}}+i\varepsilon}\frac{|V_{\mathbf{k}}|^2}{\hbar^2}e^{i\mathbf{k}\cdot\mathbf{r}_m}+B_m(\omega)\sum_{\mathbf{k}}\frac{1}{\omega-\omega_{\mathbf{k}}+i\varepsilon}\frac{|V_{\mathbf{k}}|^2}{\hbar^2}+\sum_{\mathbf{k}}\sum_{m'\neq m}B_{m'}(\omega)\frac{1}{\omega-\omega_{\mathbf{k}}+i\varepsilon}\frac{|V_{\mathbf{k}}|^2}{\hbar^2}e^{i\mathbf{k}\cdot(\mathbf{r}_m-\mathbf{r}_{m'})}.\tag{A12}$$

The second term on the right-hand side of Eq. (A12) can again be transformed into an integral. When this term is brought to the left-hand side, there will again be a frequency shift and a radiative width. The equation for $B_m(\omega)$ then becomes

$$\left(\omega-\omega_0+i\frac{\Gamma}{2\hbar}\right)B_m(\omega)=A(\omega)\sum_{\mathbf{k}}\frac{1}{\omega-\omega_{\mathbf{k}}+i\varepsilon}\frac{|V_{\mathbf{k}}|^2}{\hbar^2}e^{i\mathbf{k}\cdot\mathbf{r}_m}+\sum_{\mathbf{k}}\sum_{m'\neq m}B_{m'}(\omega)\frac{1}{\omega-\omega_{\mathbf{k}}+i\varepsilon}\frac{|V_{\mathbf{k}}|^2}{\hbar^2}e^{i\mathbf{k}\cdot(\mathbf{r}_m-\mathbf{r}_{m'})}\tag{A13}$$

which is Eq. (13) of the main text.

APPENDIX B

The sum on $\mathbf{k}$ in expression (16)

$$\left(\omega-\omega_0+i\frac{\Gamma}{2\hbar}\right)B_m(\omega)=\frac{1}{\omega-\omega_0+i(\Gamma/2\hbar)}\sum_{\mathbf{k}}\frac{1}{\omega-\omega_{\mathbf{k}}+i\varepsilon}\frac{|V_{\mathbf{k}}|^2}{\hbar^2}e^{i\mathbf{k}\cdot\mathbf{r}_m}\tag{B1}$$

of the main text will be evaluated now. After converting the sum into an integral, as has been done in Appendix A one has

$$\sum_{\mathbf{k}}\frac{1}{\omega-\omega_{\mathbf{k}}+i\varepsilon}\frac{|V_{\mathbf{k}}|^2}{\hbar^2}e^{i\mathbf{k}\cdot\mathbf{r}_m}=\frac{V}{(2\pi)^3\hbar^2}\int\int\int\frac{|V_{\mathbf{k}}|^2}{\omega-\omega_{\mathbf{k}}+i\varepsilon}e^{i\mathbf{k}\cdot\mathbf{r}_m}d^3\mathbf{k}.\tag{B2}$$

The integral in expression (B2) is of special interest, because it contains the factor $e^{i\mathbf{k}\cdot\mathbf{r}_m}$. It will be evaluated in Appendix C. One finds

$$\sum_{\mathbf{k}}\frac{1}{\omega-\omega_{\mathbf{k}}+i\varepsilon}\frac{|V_{\mathbf{k}}|^2}{\hbar^2}e^{i\mathbf{k}\cdot\mathbf{r}_m}=-\frac{V}{2\pi\hbar^2c^2}\frac{e^{i\omega r_m/c}}{r_m}|V(\omega)|^2\omega.\tag{B3}$$

The radiative width γ_R can be shown [after evaluation of the integral in expression (A9)] to be equal to

$$\gamma_R=\frac{V}{\pi\hbar c^3}|V(\omega)|^2\omega^2.\tag{B4}$$

Substituting Eq. (B4) into Eq. (B3) gives

$$\sum_{\mathbf{k}}\frac{1}{\omega-\omega_{\mathbf{k}}+i\varepsilon}\frac{|V_{\mathbf{k}}|^2}{\hbar^2}e^{i\mathbf{k}\cdot\mathbf{r}_m}=-\frac{\gamma_R}{2\hbar}\frac{e^{i\omega r_m/c}}{\omega r_m/c}\tag{B5}$$

which is a Hankel function of the first kind.

Substituting Eq. (B5) into Eq. (B1) gives

$$\left(\omega-\omega_0+i\frac{\Gamma}{2\hbar}\right)B_m(\omega)=-\frac{1}{\omega-\omega_0+i(\Gamma/2\hbar)}\frac{\gamma_R}{2\hbar}\frac{e^{i\omega r_m/c}}{\omega r_m/c}.\tag{B6}$$

Substituting finally Eqs. (14) of the main text and (B6) into Eq. (11) of the main text gives us

$$C_{\mathbf{k}}(\omega)=\frac{1}{(\omega-\omega_{\mathbf{k}}+i\varepsilon)}\frac{V_{\mathbf{k}}^*}{\hbar}\frac{1}{\omega-\omega_0+i(\Gamma/2\hbar)}-\frac{\gamma_RV_k^*}{2\hbar^2}\frac{1}{(\omega-\omega_{\mathbf{k}}+i\varepsilon)}\frac{1}{[\omega-\omega_0+i(\Gamma/2\hbar)]^2}\times\sum_m e^{-i\mathbf{k}\cdot\mathbf{r}_m}\frac{e^{i\omega r_m/c}}{\omega r_m/c}.\tag{B7}$$

Going back to the time domain, applying the inverse Fourier transformation of expression (B7) [Eq. (4)], gives $c_{\mathbf{k}}(t)$

$$c_{\mathbf{k}}(t)=-\frac{1}{2\pi i}\frac{V_{\mathbf{k}}^{*}}{\hbar}\int_{-\infty}^{+\infty}\frac{e^{i(\omega_{\mathbf{k}}-\omega)t}}{(\omega-\omega_{\mathbf{k}}+i\varepsilon)[\omega-\omega_0+i(\Gamma/2\hbar)]}\times d\omega+\frac{1}{2\pi i}\frac{V_k^{*}}{2\hbar^2}\sum_m e^{-i\mathbf{k}\cdot\mathbf{r}_m}\times\int_{-\infty}^{+\infty}\frac{e^{i(\omega_{\mathbf{k}}-\omega)t}}{(\omega-\omega_{\mathbf{k}}+i\varepsilon)[\omega-\omega_0+i(\Gamma/2\hbar)]^2}\times\gamma_R\frac{e^{i\omega r_m/c}}{\omega r_m/c}d\omega. \tag{B8}$$

The expression containing the first integral will be denoted $c_{\mathbf{k}}^{(0)}(t)$. After contour integration one finds

$$c_{\mathbf{k}}^{(0)}(t)=\frac{V_{\mathbf{k}}^{*}}{\hbar}\left[\frac{1}{\omega_{\mathbf{k}}-\omega_0+i(\Gamma/2\hbar)}+\frac{e^{i(\omega_{\mathbf{k}}-\omega_0+i\Gamma/2\hbar)t}}{\omega_0-\omega_{\mathbf{k}}-i(\Gamma/2\hbar)}\right]=\frac{V_{\mathbf{k}}^{*}}{\hbar}\frac{1}{\omega_{\mathbf{k}}-\omega_0+i(\Gamma/2\hbar)}(1-e^{i(\omega_{\mathbf{k}}-\omega_0+i\Gamma/2\hbar)t}). \tag{B9}$$

The expression containing the second integral on the right-hand side of expression (B8) will be denoted $c_{\mathbf{k}}^{(1)}(t)$, defined by

$$c_{\mathbf{k}}^{(1)}(t)=\frac{1}{2\pi i}\frac{V_k^{*}}{2\hbar^2}\sum_m e^{-i\mathbf{k}\cdot\mathbf{r}_m}\times\int_{-\infty}^{\infty}\frac{e^{i(\omega_{\mathbf{k}}-\omega)t}}{(\omega-\omega_{\mathbf{k}}+i\varepsilon)[\omega-\omega_0+i(\Gamma/2\hbar)]^2}\gamma_R\times\frac{e^{i\omega r_m/c}}{\omega r_m/c}d\omega. \tag{B10}$$

It has to be noticed that $\omega=0$ is not a pole because of the presence of γ_R [Eq. (B4)]. E.g., for a dipole transition $\gamma_R\propto\omega^3$ (see, e.g., Ref. 13). Because $\omega_{\mathbf{k}}$ will always be close to the nuclear resonance frequency ω_0, the factor $1/\omega$ can be written as $1/\omega_0$ in all residues arising in the integration over ω. This can be confirmed by a straightforward analysis. So γ_R/ω can be put in front of the integral and replaced by γ_R/ω_0. Therefore

$$c_{\mathbf{k}}^{(1)}(t)=\frac{1}{2\pi i}\frac{V_{\mathbf{k}}^{*}}{2\hbar^2}\frac{\gamma_R}{\omega_0 r_m/c}\sum_m e^{-i\mathbf{k}\cdot\mathbf{r}_m}\times\int_{-\infty}^{+\infty}\frac{e^{i(\omega_{\mathbf{k}}-\omega)t}}{(\omega-\omega_{\mathbf{k}}+i\varepsilon)[\omega-\omega_0+i(\Gamma/2\hbar)]^2}\times e^{i\omega r_m/c}d\omega. \tag{B11}$$

This integral can also be calculated using the residue theorem. For $t<r_m/c$, the contour has to be closed in the upper half plane, where there are no poles. Consequently, for $t<r_m/c$, $c_{\mathbf{k}}^{(1)}(t)=0$. This is nothing but the principle of causality which states that there cannot be any radiation coming from a scattering nucleus before it is excited by radiation coming from the source. For $t>r_m/c$ the contour has to be closed in the lower half plane, where there are two poles $\omega=\omega_{\mathbf{k}}-i\varepsilon$ and $\omega=\omega_0-i\Gamma/2\hbar$ (which is of second order). After calculation one finds

$$c_{\mathbf{k}}^{(1)}(t)=-\frac{V_{\mathbf{k}}^{*}}{2\hbar^2}\frac{\gamma_R}{[\omega_{\mathbf{k}}-\omega_0+i(\Gamma/2\hbar)]^2}\sum_m e^{-i\mathbf{k}\cdot\mathbf{r}_m}\frac{e^{i\omega_{\mathbf{k}}r_m/c}}{\omega_0 r_m/c}\times\{1+[i(\omega_{\mathbf{k}}-\omega_0+i\Gamma/2\hbar)(t-r_m/c)\times e^{i(\omega_{\mathbf{k}}-\omega_0+i\Gamma/2\hbar)(t-r_m/c)}-e^{i(\omega_{\mathbf{k}}-\omega_0+i\Gamma/2\hbar)(t-r_m/c)}]\}. \tag{B12}$$

Substituting expressions (B9) and (B12) into Eq. (B8) finally gives

$$c_{\mathbf{k}}(t)=\frac{V_{\mathbf{k}}^{*}}{\hbar}\frac{1}{\omega_{\mathbf{k}}-\omega_0+i(\Gamma/2\hbar)}(1-e^{i(\omega_{\mathbf{k}}-\omega_0+i\Gamma/2\hbar)t})-\frac{V_{\mathbf{k}}^{*}}{2\hbar^2}\frac{\gamma_R}{[\omega_{\mathbf{k}}-\omega_0+i(\Gamma/2\hbar)]^2}\sum_m e^{-i\mathbf{k}\cdot\mathbf{r}_m}\times\frac{e^{i\omega_{\mathbf{k}}r_m/c}}{\omega_0 r_m/c}\{1+[i(\omega_{\mathbf{k}}-\omega_0+i\Gamma/2\hbar)(t-r_m/c)\times e^{i(\omega_{\mathbf{k}}-\omega_0+i\Gamma/2\hbar)(t-r_m/c)}-e^{i(\omega_k-\omega_0+i\Gamma/2\hbar)(t-r_m/c)}]\} \tag{B13}$$

which is Eq. (17) of the main text.

APPENDIX C

The integral in expression (B2), $I(\mathbf{r}_m,\omega)$, is defined by

$$I(\mathbf{r}_m,\omega)=\int\int\int\frac{|V_{\mathbf{k}}|^2}{\omega-\omega_{\mathbf{k}}+i\varepsilon}e^{i\mathbf{k}\cdot\mathbf{r}_m}d^3\mathbf{k}. \tag{C1}$$

Choosing $\mathbf{r}_m$//OZ, so $e^{i\mathbf{k}\cdot\mathbf{r}_m}=e^{ikr_m\cos\theta}$, one has

$$I(\mathbf{r}_m,\omega)=\int\int\int\frac{|V_{\mathbf{k}}|^2}{\omega-\omega_{\mathbf{k}}+i\varepsilon}e^{ikr_m\cos\theta}k^2dk\sin\theta d\theta d\phi. \tag{C2}$$

Supposing that $|V_{\mathbf{k}}|^2$ does not depend on θ and ϕ (no polarization effects), the ϕ integration gives 2π. The θ integration is also simple

$$\int_0^{\pi}e^{ikr_m\cos\theta}\sin\theta d\theta=\frac{e^{ikr_m}-e^{-ikr_m}}{ikr_m}. \tag{C3}$$

Therefore

$$I(\mathbf{r}_m,\omega)=2\pi\int_0^{\infty}\frac{|V_{\mathbf{k}}|^2}{\omega-\omega_{\mathbf{k}}+i\varepsilon}\left(\frac{e^{ikr_m}-e^{-ikr_m}}{ikr_m}\right)k^2dk. \tag{C4}$$

With

$$k=\frac{\omega_{\mathbf{k}}}{c}\rightarrow dk=\frac{1}{c}d\omega_{\mathbf{k}} \tag{C5}$$

the integral becomes

$$I(\mathbf{r}_m,\omega)=\frac{2\pi}{c^3}\int_0^\infty\frac{|V_\mathbf{k}|^2}{\omega-\omega_\mathbf{k}+i\varepsilon}\left(\frac{e^{i\omega_\mathbf{k}r_m/c}-e^{-i\omega_\mathbf{k}r_m/c}}{i\omega_\mathbf{k}r_m/c}\right)\times\omega_\mathbf{k}^2 d\omega_\mathbf{k}. \tag{C6}$$

This integral cannot be evaluated in a simple manner using contour integration, because of the lower bound 0. It will be shown now how the lower bound can be extended to $-\infty$, without changing the value of the integral. $e^{\pm i\omega_\mathbf{k}r_m/c}$ are oscillating functions of $\omega_\mathbf{k}$. For r_m large (of the order of a few Å), $\omega_\mathbf{k}r_m/c\gg 1$, except for small values of $\omega_\mathbf{k}$. But for small values of $\omega_\mathbf{k}$, $\omega_\mathbf{k}^2/(\omega-\omega_\mathbf{k}+i\varepsilon)$ is negligible. For appreciable values of $\omega_\mathbf{k}$, $e^{\pm i\omega_\mathbf{k}r_m/c}$ oscillates rapidly, so there will be no contribution to the integral unless one of the other factors shows a comparatively fast variation, compensating this due to $e^{\pm i\omega_\mathbf{k}r_m/c}$. This occurs near $k=\omega/c$. As a consequence, the range of $\omega_\mathbf{k}$ values that contribute to the integral could be reduced to a small region $c\Delta k_0$ around ω and there, all factors are practically constant except those which have just been mentioned. So we can extend the integral from $-\infty$ to $+\infty$. Therefore

$$I(\mathbf{r}_m,\omega)=\frac{2\pi}{ir_mc^2}\int_{-\infty}^{\infty}\frac{|V_\mathbf{k}|^2e^{i\omega_\mathbf{k}r_m/c}}{\omega-\omega_\mathbf{k}+i\varepsilon}\,\omega_\mathbf{k}d\omega_\mathbf{k}-\frac{2\pi}{ir_mc^2}\int_{-\infty}^{\infty}\frac{|V_\mathbf{k}|^2e^{-i\omega_\mathbf{k}r_m/c}}{\omega-\omega_\mathbf{k}+i\varepsilon}\,\omega_\mathbf{k}d\omega_\mathbf{k}. \tag{C7}$$

These two integrals, contrary to the one in expression (C6), can be calculated easily using the residue theorem. The contour of the first integral has to be closed in the upper half plane. The pole is within the contour. The contour of the second integral has to be closed in the lower half plane and the pole is outside of the contour, so that the second integral is zero. Therefore we have

$$I(\mathbf{r}_m,\omega)=-\frac{(2\pi)^2e^{i\omega r_m/c}|V(\omega)|^2\omega}{r_mc^2}. \tag{C8}$$

This expression is used in expression (B2).

[1] M. Tegze and G. Faigel, Europhys. Lett. **16**, 41 (1991).
[2] M. Tegze and G. Faigel, Nature (London) **380**, 49 (1996).
[3] P. Korecki, J. Korecki, and T. Slezak, Phys. Rev. Lett. **79**, 3518 (1997).
[4] W. Heitler, *The Quantum Theory of Radiation*, 3rd ed. (Oxford University Press, Oxford, 1954), pp. 163–174.
[5] S. M. Harris, Phys. Rev. **124**, 1178 (1961).
[6] G. R. Hoy, J. Phys.: Condens. Matter **9**, 8749 (1997).
[7] A. Szöke, in *Short Wavelength Coherent Radiation: Generation and Applications*, AIP Conf. Proc. No. 147, edited by T. Attwood and J. Baker (AIP, New York, 1986), p. 361.
[8] J. J. Barton, S. W. Robey, and D. A. Shirley, Phys. Rev. B **34**, 778 (1986).
[9] J. J. Barton, Phys. Rev. Lett. **61**, 1356 (1988).
[10] L. Mandel and E. Wolf, *Optical Coherence and Quantum Optics* (Cambridge University Press, Cambridge, 1995).
[11] G. Faigel and M. Tedge, Europhys. News **27**, 179 (1996).
[12] J. Dekoster (private communication).
[13] W.H. Louisell, *Quantum Statistical Properties of Radiation* (Wiley, New York, 1973).