Quantum-mechanical theory of enhanced resolution in Mössbauer spectroscopy using a resonant detector

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The quantum-mechanical theory for Mössbauer spectroscopy using a resonant detector, in which the conversion electrons are recorded, is presented. The resonant detector consists of nuclei having the same microscopic environment as the source nuclei. Fundamental equations, in the frequency domain, describing the system formed by a source nucleus, an ensemble of absorber nuclei, and a resonant detector nucleus are solved. The absorber is allowed to be of arbitrary thickness. The analysis includes the presence of conversion electrons and γ radiation. The full width at half maximum of a Mössbauer spectrum, using the resonant-detector scheme, is calculated and found to be appreciably smaller than the width found using a conventional Mössbauer setup.

I. INTRODUCTION

A conventional Mössbauer-effect apparatus[1] consists of a radioactive source, an absorber containing the same type of nuclei in the ground state and a γ radiation detector (e.g., a proportional counter). The intensity of the transmitted electromagnetic radiation, as a function of the relative velocity of the source with respect to the absorber, is measured. It is well known that the minimum linewidth in a conventional Mössbauer experiment is two times the natural linewidth of the excited state. It is also possible to count the conversion electrons produced by the absorber nuclei instead of detecting the γ radiation that is transmitted through the absorber. This is particularly useful if the conversion coefficient is high. However, in that case also, the spectral linewidth still has a lower limit of two times the natural linewidth of the excited state.

In a previous paper[2] it has been shown that if, instead of a conventional γ radiation detector (proportional counter, NaI detector), one uses a ''resonant'' detector, the full width at half maximum of the Mössbauer spectrum is appreciably smaller than two times the natural linewidth when the absorber is thin. The resonant detector contains nuclei whose environment is identical with that of the source nuclei. This insures that the isomer shift[1] of the detector's nuclear levels with respect to the source's nuclear levels is zero. In this setup, as also often used in the conventional setup, the absorber is moved with respect to the source and the detector which are both stationary and hence in resonance. One detects conversion electrons coming from the resonant-detector nuclei as a function of the velocity of the absorber. In the paper[2] mentioned above, the quantum-mechanical system, consisting of a source nucleus, an absorber nucleus, and a resonant detector nucleus, was studied. In order to obtain the minimum linewidth, a very thin absorber was considered. This corresponds to the linear absorption approximation, which simplifies the mathematics involved. In practice however, the absorber usually has a thickness that does not correspond to the thin absorber limit. If a thick absorber is considered, multiple photon scattering processes occur in the absorber. These processes complicate the solution of the equations describing the system. In this paper we present the general solution of the problem for an absorber of arbitrary thickness.

II. MATHEMATICAL FRAMEWORK

The general method used in this paper is discussed in Heitler,[3] Harris,[4] and in a more recent paper.[5] The method applies time-dependent quantum mechanics 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 part describing the evolution of the nuclei, the free radiation field and the conversion electrons in the absence of any coupling. The eigenstates of H_0 correspond to nuclear states of an ensemble of noninteracting nuclei, the states of the free radiation field, taken here as plane waves, and the states of the conversion electrons, also taken as plane waves. Only two nuclear states are considered, namely the nuclear ground state and the nuclear first-excited state. The part of the Hamiltonian describing transitions between the eigenstates of H_0 is denoted by V.

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 is 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 $\omega_l - \omega_q = (E_l - E_q)/\hbar$. It is necessary to obtain a solution of this system of coupled differential equations that

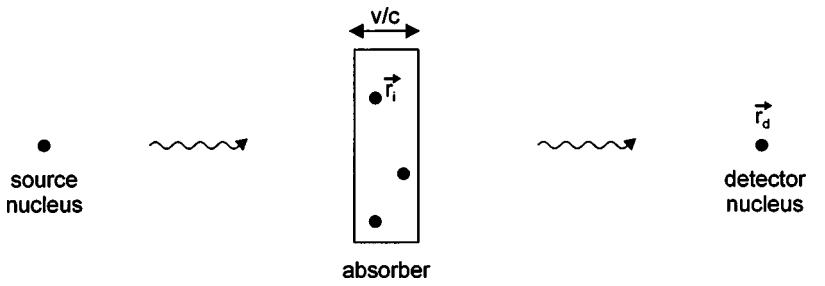

FIG. 1. Geometry of the Mössbauer setup using a resonant detector consisting of resonant nuclei. The source is at the origin of the coordinate system. The absorber, consisting of resonant nuclei i at positions $\mathbf{r}_i$, is Doppler modulated. The position of a particular resonant-detector nucleus, labeled d, is at $\mathbf{r}_d$.

satisfies the initial condition that at $t=0$ the system is in a well-defined state, say $|\varphi_n\rangle$, and all other probability amplitudes are zero. This means that for $t=0$, $a_l(0)=0$ for l not equal to n. On the other hand, $a_n(+0)=1$, which indicates the result for a_n when t approaches zero from the positive side. Although a physically meaningful solution only involves positive times ($t\geqslant 0$), for analytical reasons, following Heitler,[3] the solution will be extended to the negative time axis. The a_l's are chosen such that $a_l(t)=a_n(t)=0$ for $t<0$. It follows that $a_n(t)$ has a discontinuity at $t=0$, which can be represented by a step function $\theta(t)$, that must be dealt with. Heitler[3] has shown that by adding an inhomogeneous term, to the right-hand side of Eq. (2), the initial conditions and the discontinuity are handled correctly. The resulting equation is

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

where $\delta_{\ln}$ is the Kronecker delta and $\delta(t)$ is the Dirac delta function. Next introducing the Fourier transform[3]

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

allows Eq. (2) to 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 time-independent matrix element inducing a transition from the qth state to the lth state of H_0, i.e.,

$$V_{lq}=\langle\varphi_l(0)|V|\varphi_q(0)\rangle. \tag{6}$$

The integral representation of the Dirac delta function has also been used. To obtain an equation for $A_l(\omega)$ we would have to divide Eq. (5) by $(\omega-\omega_l)$. This division will not be unique[3] 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 represented by a factor

$$\lim_{\varepsilon\to 0^+}\frac{1}{\omega-\omega_l+i\varepsilon}.$$

This is the proper procedure to avoid infinities in the integrals and to choose the direction of time so that ''cause'' precedes ''effect.'' (See below.) In fact, the replacement of $(\omega-\omega_l)$ by $(\omega-\omega_l+i\varepsilon)$ (ε being an infinitesimal, positive number) defines the right path of integration guaranteeing causality.[3] This is completely analogous to the definition of integration paths in the propagator concept of 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

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

The advantage of the set of equations, Eq. (7), is that it is a linear system of coupled algebraic, not differential, equations. In the next section we apply the formalism to our study of the interaction of radiation with nuclei embedded in a lattice.

III. FUNDAMENTAL EQUATIONS

Suppose we have an excited nucleus, the ''source'' nucleus, with energy $\hbar\omega_0$ at the origin of a coordinate system at time $t=0$. We also have an ensemble of identical nuclei i ($i=1$ to N), initially in the ground state, situated at positions $\mathbf{r}_i$. These N nuclei constitute the ''absorber.'' The absorber nuclei have a first-excited state at energy $\hbar\omega_0'$. Finally, we consider another ground-state nucleus, the ''detector'' nucleus, located at position $\mathbf{r}_d$. The detector nucleus has a first-excited state at energy $\hbar\omega_0$ as does the source nucleus. The detector nucleus represents the ''resonant detector,'' which is not part of the absorber. The geometry of the setup is schematized in Fig. 1. The setup corresponds to a thin source, an absorber of arbitrary thickness, and a thin resonant detector. As a consequence, we can neglect multiple photon scattering in the source and in the resonant detector, but must allow for multiple scattering in the absorber. The absorber nuclei are, obviously, situated between the source nucleus and the resonant-detector nucleus. The evolution of the quantum system composed of the nuclei, the radiation field, and the conversion electrons will be investigated. In the following, we will assume that the recoil-free fractions,[1] of the emission and absorption processes, are one. This is not an essential hypothesis.

The mathematical formalism, briefly sketched above, can be applied to our system. The following amplitudes can be defined:

(i) $A(\omega)$ is the amplitude corresponding to the source nucleus excited ($\hbar\omega_0$), the other nuclei in the ground state, and no photon or conversion electron is present;

(ii) $B_{\mathbf{k}}(\omega)$ is the amplitude corresponding to all nuclei in the ground state, a photon of wave number $\mathbf{k}$ and energy $\hbar\omega_{\mathbf{k}}$ present, and no conversion electron;

(iii) $C_i(\omega)$ ($i=1$ to N) is the amplitude corresponding to absorber nucleus i at position $\mathbf{r}_i$ excited ($\hbar\omega_0'$), all other nuclei in the ground state, and no photon or conversion electron is present;

(iv) $A_d(\omega)$ is the amplitude corresponding to the resonant detector nucleus d excited at energy $\hbar\omega_0$, all other nuclei in the ground state, and no photon or conversion electron is present;

(v) $D_{\mathbf{p}}(\omega)$ is the amplitude for having a conversion electron from the source nucleus with momentum $\mathbf{p}$ present, all nuclei in the ground state, and no photon present;

(vi) $E_{i\mathbf{p}}(\omega)$, $i=1$ to N, is the amplitude for having a conversion electron from absorber nucleus i present, all nuclei in the ground state, and no photon present;

(vii) $F_{d\mathbf{p}}(\omega)$ is the amplitude for having only a conversion electron from the detector nucleus present, all nuclei in the ground state, and no photon present.

At $t=0$ only the source nucleus is excited. Thus we have the following set of coupled equations which satisfy the initial conditions:

$$(\omega-\omega_0+i\varepsilon)A(\omega)=1+\sum_{\mathbf{k}}\frac{H_{\mathbf{k}}}{\hbar}B_{\mathbf{k}}(\omega)+\sum_{\mathbf{p}}\frac{H_{\mathbf{p}}}{\hbar}D_{\mathbf{p}}(\omega), \tag{8}$$

$$(\omega-\omega_{\mathbf{k}}+i\varepsilon)B_{\mathbf{k}}(\omega)=\frac{H_{\mathbf{k}}^*}{\hbar}A(\omega)+\sum_{i=1}^{N}\frac{H_{\mathbf{k}}^*}{\hbar}e^{-i\mathbf{k}\cdot\mathbf{r}_i}C_i(\omega)+\frac{H_{\mathbf{k}}^*}{\hbar}e^{-i\mathbf{k}\cdot\mathbf{r}_d}A_d(\omega), \tag{9}$$

$$(\omega-\omega_0'+i\varepsilon)C_i(\omega)=\sum_{\mathbf{k}}\frac{H_{\mathbf{k}}}{\hbar}e^{i\mathbf{k}\cdot\mathbf{r}_i}B_{\mathbf{k}}(\omega)+\sum_{\mathbf{p}}\frac{H_{\mathbf{p}}}{\hbar}e^{i[(\mathbf{p}\cdot\mathbf{r}_i)/\hbar]}E_{i\mathbf{p}}(\omega), \tag{10}$$

$$(\omega-\omega_0+i\varepsilon)A_d(\omega)=\sum_{\mathbf{k}}\frac{H_{\mathbf{k}}}{\hbar}e^{i\mathbf{k}\cdot\mathbf{r}_d}B_{\mathbf{k}}(\omega)+\sum_{\mathbf{p}}\frac{H_{\mathbf{p}}}{\hbar}e^{i[(\mathbf{p}\cdot\mathbf{r}_d)/\hbar]}F_{d\mathbf{p}}(\omega), \tag{11}$$

$$(\omega-\omega_{\mathbf{p}}+i\varepsilon)D_{\mathbf{p}}(\omega)=\frac{H_{\mathbf{p}}^*}{\hbar}A(\omega), \tag{12}$$

$$(\omega-\omega_{\mathbf{p}}+i\varepsilon)E_{i\mathbf{p}}(\omega)=\frac{H_{\mathbf{p}}^*}{\hbar}e^{-i[(\mathbf{p}\cdot\mathbf{r}_i)/\hbar]}C_i(\omega)\quad i=1,N, \tag{13}$$

$$(\omega-\omega_{\mathbf{p}}+i\varepsilon)F_{d\mathbf{p}}(\omega)=\frac{H_{\mathbf{p}}^*}{\hbar}e^{-i[(\mathbf{p}\cdot\mathbf{r}_d)/\hbar]}A_d(\omega), \tag{14}$$

where we have written $H_{\mathbf{k}}$ for the matrix element coupling the nuclear ground state to the excited state through absorption of a photon having wave vector $\mathbf{k}$. $H_{\mathbf{k}}^*$ is the complex conjugate of $H_{\mathbf{k}}$. For the processes where conversion electrons are involved, there are analogous matrix elements $H_{\mathbf{p}}$ and $H_{\mathbf{p}}^*$.

Equations (8)–(14) can be interpreted in a straightforward manner. For example, Eq. (9) describes the production of a photon having wave vector $\mathbf{k}$. This can occur through the emission of such a photon by the source nucleus. This is the first term at the right-hand side of Eq. (9). This can also occur through photon emission by one of the absorber nuclei at positions $\mathbf{r}_i$ ($i=1$ to N). This is represented by the series on the right-hand side of Eq. (9). Finally, this can occur through photon emission by the resonant-detector nucleus. This is represented by the last term on the right-hand side of Eq. (9). The last two terms on the right-hand side of Eq. (9) represent re-emission (or scattering) processes because the absorber and detector nuclei are not excited at $t=0$, according to the initial condition. The functions $e^{-i\mathbf{k}\cdot\mathbf{r}_i}$, $i=1$ to N, and $e^{-i\mathbf{k}\cdot\mathbf{r}_d}$ designate the phase according to the position where the photon emission takes place, i.e., at $\mathbf{r}_i$, the position of absorber nucleus i, or at $\mathbf{r}_d$, the position of the resonant-detector nucleus. The source nucleus, being at the origin, does not have such a factor. The other equations can be interpreted in a similar manner. The set of Eqs. (8)–(14) describes the system completely.

IV. SOLUTION OF THE EQUATIONS

A. Fundamental equations in one dimension

First we will treat the conversion electrons to reduce the number of equations. Substituting Eq. (12) into Eq. (8) and converting the sum over $\mathbf{p}$ into an integral using the well-known prescription[3]

$$\sum_{\mathbf{p}}\rightarrow\frac{V}{(2\pi\hbar)^3}\int\int\int d^3\mathbf{p} \tag{15}$$

gives

$$\sum_{\mathbf{p}}\frac{|H_{\mathbf{p}}|^2}{(\omega-\omega_{\mathbf{p}}+i\varepsilon)\hbar^2}A(\omega)\rightarrow\frac{V}{(2\pi\hbar)^3}\int\int\int\frac{|H_{\mathbf{p}}|^2}{(\omega-\omega_{\mathbf{p}}+i\varepsilon)\hbar^2}d^3\mathbf{p}A(\omega). \tag{16}$$

Using the relation[3]

$$\frac{1}{x-a\pm i\varepsilon}=P\frac{1}{x-a}\mp i\pi\delta(x-a), \tag{17}$$

where P indicates the principal part of an integral, one has

$$\sum_{\mathbf{p}}\frac{|H_{\mathbf{p}}|^2}{(\omega-\omega_{\mathbf{p}}+i\varepsilon)\hbar^2}A(\omega)\rightarrow\frac{V}{(2\pi\hbar)^3\hbar^2}\left[P\int\int\int\frac{|H_{\mathbf{p}}|^2}{\omega-\omega_{\mathbf{p}}}d^3\mathbf{p}-i\pi\times\int\int\int|H_{\mathbf{p}}|^2\delta(\omega-\omega_{\mathbf{p}})d^3\mathbf{p}\right]A(\omega). \tag{18}$$

Equation (8) becomes then

$$(\omega-\omega_0+i\varepsilon)A(\omega)=1+\sum_{\mathbf{k}}\frac{H_{\mathbf{k}}}{\hbar}B_{\mathbf{k}}(\omega)+\left(\delta-i\frac{\gamma_C}{2\hbar}\right)A(\omega), \tag{19}$$

where δ is defined by

$$\delta = \frac{V}{(2\pi\hbar)^3\hbar^2} P \int\int\int \frac{|H_{\mathbf{p}}|^2}{\omega-\omega_{\mathbf{p}}} d^3\mathbf{p}, \tag{20}$$

and γ_C by

$$\gamma_C = \frac{2\pi V}{(2\pi\hbar)^3\hbar} \int\int\int |H_{\mathbf{p}}|^2 \delta(\omega-\omega_{\mathbf{p}}) d^3\mathbf{p}. \tag{21}$$

The presence of V, a volume, in Eqs. (20) and (21), as well as in subsequent expressions, is only apparent because the matrix elements $|H_{\mathbf{p}}|^2$ contain[6] $1/V$. When the last term on the right-hand side of Eq. (19) is brought to the left-hand side, δ corresponds to a shift in the nuclear frequency, which can be incorporated into ω_0, and γ_C gives a width due to the interaction of the nucleus with its conversion electron. The shift and the width are apparently functions of ω. A careful analysis performed by Heitler[3] shows that for the width and the shift the value ω can be replaced by ω_0. This shows up only after going back to the time domain for times such that $t \gg 1/\omega_0$. In the nuclear realm $\omega_0 \cong 10^{18}\,\mathrm{s}^{-1}$ or larger, so 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. With the shift incorporated into ω_0, Eq. (19) becomes

$$\left(\omega-\omega_0+i\frac{\gamma_C}{2\hbar}\right) A(\omega) = 1 + \sum_{\mathbf{k}} \frac{H_{\mathbf{k}}}{\hbar} B_{\mathbf{k}}(\omega). \tag{22}$$

The conversion electrons, from the absorber nuclei, can be treated in a similar way. Solving Eq. (13) for $E_{i\mathbf{p}}(\omega)$ and substituting into Eq. (10), making use of the same procedure as for $D_{\mathbf{p}}(\omega)$, gives

$$\left(\omega-\omega_0'+i\frac{\gamma_C}{2\hbar}\right) C_i(\omega) = \sum_{\mathbf{k}} \frac{H_{\mathbf{k}}}{\hbar} e^{i\mathbf{k}\cdot\mathbf{r}_i} B_{\mathbf{k}}(\omega). \tag{23}$$

Analogously, with Eqs. (14) and (11), one obtains

$$\left(\omega-\omega_0+i\frac{\gamma_C}{2\hbar}\right) A_d(\omega) = \sum_{\mathbf{k}} \frac{H_{\mathbf{k}}}{\hbar} e^{i\mathbf{k}\cdot\mathbf{r}_d} B_{\mathbf{k}}(\omega). \tag{24}$$

The sole effect of considering the conversion electrons is to introduce the conversion-electron width γ_C in the equations for the amplitudes $A(\omega)$, $C_i(\omega)$, and $A_d(\omega)$. The fundamental equations describing the interaction of radiation with the nuclei are reduced to Eqs. (9) and (22)–(24). In order to solve this system of equations the approach of Ref. 5 will be adopted. Reference 5 treats the problem of an excited source nucleus and N resonant absorber nuclei in the ground state. It has been shown[5] that this problem has a closed-form solution if one restricts the calculation to forward scattering. A simple argument can be given to show that it is reasonable to consider radiation only in the forward direction. If one considers scattering in directions other than forward, one must account for the difference in the optical paths for all radiation reaching the detector. Since we must sum over all coherent amplitudes, representing the different scattering paths the radiation takes from the source to the absorber and then to the detector, the difference in the various optical path lengths must be considered. For polycrystalline material having small grains only the forward direction exhibits special features, because only in this direction all optical path lengths are equal.

The fundamental equations, specialized to the forward direction, can be written as

$$(\omega-\omega_k+i\varepsilon) B_k(\omega) = \frac{H_k^*}{\hbar} A(\omega) + \sum_{i=1}^{N} \frac{H_k^*}{\hbar} e^{-ikx_i} C_i(\omega) + \frac{H_k^*}{\hbar} e^{-ikx_d} A_d(\omega), \tag{25}$$

$$\left(\omega-\omega_0+i\frac{\gamma_C}{2\hbar}\right) A(\omega) = 1 + \sum_k \frac{H_k}{\hbar} B_k(\omega), \tag{26}$$

$$\left(\omega-\omega_0'+i\frac{\gamma_C}{2\hbar}\right) C_i(\omega) = \sum_k \frac{H_k}{\hbar} e^{ikx_i} B_k(\omega), \tag{27}$$

$$\left(\omega-\omega_0+i\frac{\gamma_C}{2\hbar}\right) A_d(\omega) = \sum_k \frac{H_k}{\hbar} e^{ikx_d} B_k(\omega), \tag{28}$$

where the positions of the absorber nuclei are denoted by x_i and the position of the resonant-detector nucleus by x_d. The positions are ordered such that $0 < x_1 < x_2 < \cdots < x_{N-1} < x_N$. In our geometry we also have $x_i < x_d$ for all values of i.

B. Transformation of the equations

Solving Eq. (25) for $B_k(\omega)$ and substituting into Eq. (27) gives

$$\begin{aligned}\left(\omega-\omega_0'+i\frac{\gamma_C}{2\hbar}\right) C_i(\omega) &= \sum_k \frac{|H_k|^2}{\hbar^2} \frac{e^{ikx_i}}{\omega-\omega_k+i\varepsilon} A(\omega) \\ &+ \sum_k \frac{|H_k|^2}{\hbar^2} \frac{1}{\omega-\omega_k+i\varepsilon} C_i(\omega) \\ &+ \sum_{j=1,j\neq i}^{N} \sum_k \frac{|H_k|^2}{\hbar^2} \frac{1}{\omega-\omega_k+i\varepsilon} e^{ik(x_i-x_j)} C_j(\omega) \\ &+ \sum_k \frac{|H_k|^2}{\hbar^2} \frac{1}{\omega-\omega_k+i\varepsilon} e^{ik(x_i-x_d)} A_d(\omega).\end{aligned} \tag{29}$$

By the same substitution one obtains from Eq. (28)

$$\left(\omega-\omega_0+i\frac{\gamma_C}{2\hbar}\right)A_d(\omega)$$
$$=\sum_k \frac{|H_k|^2}{\hbar^2}\frac{e^{ikx_d}}{\omega-\omega_k+i\varepsilon}A(\omega)$$
$$+\sum_k \frac{|H_k|^2}{\hbar^2}\frac{1}{\omega-\omega_k+i\varepsilon}A_d(\omega)$$
$$+\sum_{i=1}^{N}\sum_k \frac{|H_k|^2}{\hbar^2}\frac{1}{\omega-\omega_k+i\varepsilon}e^{ik(x_d-x_i)}C_i(\omega). \tag{30}$$

The common term

$$\sum_k \frac{|H_k|^2}{\hbar^2}\frac{1}{\omega-\omega_k+i\varepsilon}.$$

in Eqs. (29) and (30) can be transformed into an integral. The prescription in the one-dimensional geometry is given[4,5] by

$$\sum_k \rightarrow \frac{L}{2\pi}\int dk, \tag{31}$$

where L is a normalizing length appearing when the one-dimensional sums are transformed into one-dimensional integrals. Again, using Eq. (17), this sum will give a shift which can be incorporated into ω_0 or ω_0', and a width γ_R, which is called the radiative width. The radiative width and the width due to the conversion electron defines the total width Γ

$$\Gamma=\gamma_C+\gamma_R. \tag{32}$$

Equation (29) now becomes

$$\left(\omega-\omega_0'+i\frac{\Gamma}{2\hbar}\right)C_i(\omega)$$
$$=\sum_k \frac{|H_k|^2}{\hbar^2}\frac{e^{ikx_i}}{\omega-\omega_k+i\varepsilon}A(\omega)$$
$$+\sum_{j=1,j\neq i}^{N}\sum_k \frac{|H_k|^2}{\hbar^2}\frac{1}{\omega-\omega_k+i\varepsilon}e^{ik(x_i-x_j)}C_j(\omega)$$
$$+\sum_k \frac{|H_k|^2}{\hbar^2}\frac{1}{\omega-\omega_k+i\varepsilon}e^{ik(x_i-x_d)}A_d(\omega), \tag{33}$$

and Eq. (30) becomes

$$\left(\omega-\omega_0+i\frac{\Gamma}{2\hbar}\right)A_d(\omega)$$
$$=\sum_k \frac{|H_k|^2}{\hbar^2}\frac{e^{ikx_d}}{\omega-\omega_k+i\varepsilon}A(\omega)$$
$$+\sum_{i=1}^{N}\sum_k \frac{|H_k|^2}{\hbar^2}\frac{1}{\omega-\omega_k+i\varepsilon}e^{ik(x_d-x_i)}C_i(\omega). \tag{34}$$

Equation (33) gives us, after transforming the remaining Σ_k into integrals with the same prescription as before

$$\left(\omega-\omega_0'+i\frac{\Gamma}{2\hbar}\right)C_i(\omega)$$
$$=\frac{L}{2\pi\hbar^2}\int_{-\infty}^{+\infty}\frac{|H_k|^2e^{ikx_i}}{\omega-\omega_k+i\varepsilon}dkA(\omega)+\frac{L}{2\pi\hbar^2}$$
$$\times\sum_{j=1,j\neq i}^{N}\int_{-\infty}^{+\infty}\frac{|H_k|^2}{\omega-\omega_k+i\varepsilon}e^{ik(x_i-x_j)}dkC_j(\omega)$$
$$+\frac{L}{2\pi\hbar^2}\int_{-\infty}^{+\infty}\frac{|H_k|^2}{\omega-\omega_k+i\varepsilon}e^{ik(x_i-x_d)}dkA_d(\omega). \tag{35}$$

It will be shown, in the Appendix, that this equation can be transformed into

$$\left(\omega-\omega_0'+i\frac{\Gamma}{2\hbar}\right)C_i(\omega)$$
$$=-\frac{i\gamma_R}{2\hbar}e^{ix_i\omega/c}A(\omega)-\frac{i\gamma_R}{2\hbar}\sum_{j=1}^{i-1}e^{i(x_i-x_j)\omega/c}C_j(\omega)$$
$$+\frac{i\gamma_R}{2\hbar}\sum_{j=i+1}^{N}e^{-i(x_i-x_j)\omega/c}C_j(\omega)$$
$$+\frac{i\gamma_R}{2\hbar}e^{-i(x_i-x_d)\omega/c}A_d(\omega). \tag{36}$$

The contribution, due to the nuclei ''upstream'' from nucleus i, is given by the first sum on the right-hand side of Eq. (36). The second series in Eq. (36) and the last single term give the contribution from the nuclei ''downstream'' from the nucleus labeled by i. Each contribution has a different form. This is important as will be seen below.

The equation for $A_d(\omega)$ can be calculated form Eq. (34) by applying the same procedure. One finds

$$\left(\omega-\omega_0+i\frac{\Gamma}{2\hbar}\right)A_d(\omega)=-\frac{i\gamma_R}{2\hbar}e^{ix_d\omega/c}A(\omega)$$
$$-\frac{i\gamma_R}{2\hbar}\sum_{i=1}^{N}e^{i(x_d-x_i)\omega/c}C_i(\omega). \tag{37}$$

With our geometry all absorber nuclei are ''upstream'' with respect to the resonant-detector nucleus, which is located at a distance x_d from the source nucleus.

The equation for $A(\omega)$ can be calculated by substituting Eq. (25) into Eq. (26). After similar algebraic manipulations one obtains

$$\left(\omega-\omega_0+i\frac{\Gamma}{2\hbar}\right)A(\omega)=1+\sum_{i=1}^{N}\frac{i\gamma_R}{2\hbar}e^{ix_i\omega/c}C_i(\omega)$$
$$+\frac{i\gamma_R}{2\hbar}e^{ix_d\omega/c}A_d(\omega). \tag{38}$$

With respect to the source nucleus, all other nuclei are of course ''downstream.'' The fundamental equations are Eqs. (36)–(38) together with Eq. (25).

C. Solution of the equations

Equations (36)–(38) can be formally solved by means of iteration. The first-order approximation gives

$$A^{(1)}(\omega)=\frac{1}{\omega-\omega_0+i(\Gamma/2\hbar)}, \tag{39}$$

$$C_i^{(1)}(\omega)=-\frac{i\gamma_R}{2\hbar}e^{ix_i\omega/c}\frac{1}{\left(\omega-\omega_0+i\frac{\Gamma}{2\hbar}\right)\left(\omega-\omega_0'+i\frac{\Gamma}{2\hbar}\right)}, \tag{40}$$

$$A_d^{(1)}(\omega)=-\frac{i\gamma_R}{2\hbar}e^{ix_d\omega/c}\frac{1}{\left(\omega-\omega_0+i\frac{\Gamma}{2\hbar}\right)^2}. \tag{41}$$

Substituting Eqs. (40) and (41) into Eq. (38) gives the second-order approximation for $A(\omega)$. One finds

$$A^{(2)}(\omega)=\frac{1}{\omega-\omega_0+i\frac{\Gamma}{2\hbar}}+\sum_{i=1}^{N}\left(\frac{\gamma_R}{2\hbar}\right)^2\times\frac{1}{\left(\omega-\omega_0+i\frac{\Gamma}{2\hbar}\right)^2\left(\omega-\omega_0'+i\frac{\Gamma}{2\hbar}\right)}e^{2ix_i\omega/c}+\left(\frac{\gamma_R}{2\hbar}\right)^2\frac{1}{\left(\omega-\omega_0+i\frac{\Gamma}{2\hbar}\right)^3}e^{2ix_d\omega/c}. \tag{42}$$

The first term on the right-hand side of Eq. (42) is the dominant one. This can be seen easily on resonance, where $\omega=\omega_0$ or $\omega=\omega_0'$. The absolute value of the ratio of a term of the series and the first term is of the order of γ_R^2/Γ^2, which, in the case of ^{57}Fe, is about 1/100. We have the same order of magnitude for the absolute value of the ratio of the last term of Eq. (42) and the first term. Furthermore, and more importantly, the factors $e^{2ix_i\omega/c}$ and $e^{2ix_d\omega/c}$ are rapidly oscillating functions, compared to the constant 1. When going back to the time domain one must integrate over ω, where ω will eventually be replaced by its value at the poles. Due to the rapid oscillations of these phase factors, the corresponding integrals will be negligible. Higher-order approximations will contain even more rapidly oscillating phase factors. Hence they can also be neglected.

The physical explanation corresponding to this approximation is as follows. The radiation, coming from the absorber nuclei after they have been excited by radiation coming from the source nucleus, has a very small probability of being reabsorbed by the source nucleus. The solid angle subtended by the source nucleus with respect to any absorber nucleus, or the resonant-detector nucleus for that matter, is extremely small. Thus, also radiation coming from the resonant detector nucleus does not re-excite the source at any time greater than zero. Using this approximation the equation for $A(\omega)$ becomes

$$A(\omega)=\frac{1}{\omega-\omega_0+i(\Gamma/2\hbar)}. \tag{43}$$

Analogously, the second approximation for $C_i(\omega)$ can be calculated. One finds

$$C_i^{(2)}(\omega)=-\frac{i\gamma_R}{2\hbar}e^{ix_i\omega/c}\frac{1}{\left(\omega-\omega_0+i\frac{\Gamma}{2\hbar}\right)\left(\omega-\omega_0'+i\frac{\Gamma}{2\hbar}\right)}+\left(-\frac{i\gamma_R}{2\hbar}\right)^2\sum_{j=1}^{i-1}e^{ix_i\omega/c}\times\frac{1}{\left(\omega-\omega_0+i\frac{\Gamma}{2\hbar}\right)\left(\omega-\omega_0'+i\frac{\Gamma}{2\hbar}\right)^2}-\left(\frac{i\gamma_R}{2\hbar}\right)^2\sum_{j=i+1}^{N}e^{-i(x_i-2x_j)\omega/c}\times\frac{1}{\left(\omega-\omega_0+i\frac{\Gamma}{2\hbar}\right)\left(\omega-\omega_0'+i\frac{\Gamma}{2\hbar}\right)^2}-\left(\frac{i\gamma_R}{2\hbar}\right)^2\times\frac{1}{\left(\omega-\omega_0'+i\frac{\Gamma}{2\hbar}\right)\left(\omega-\omega_0+i\frac{\Gamma}{2\hbar}\right)^2}e^{-i(x_i-2x_d)\omega/c}. \tag{44}$$

The exponential functions $e^{-i(x_i-2x_j)\omega/c}$ and $e^{-i(x_i-2x_d)\omega/c}$ are rapidly oscillating phase factors compared to $e^{ix_i\omega/c}$. When going back to the time domain, the contribution of the more rapidly oscillating functions again will be small compared to the other ones. Higher-order approximations will contain even more rapidly oscillating functions that can also be neglected. The approximation amounts to neglecting the second series and the last term on the right-hand side of Eq. (36). The physical explanation behind this approximation is that the radiation coming from the nuclei ''downstream'' (with respect to the positive direction of the x axis) from absorber nucleus i does not ''return'' to excite nucleus i. The physics of this will be explained briefly below. When calculating the second and higher-order approximations for $A_d(\omega)$, one finds that there are no terms that can be neglected. The conclusion of this analysis is that only radiation coming from nuclei ''upstream'' (with respect to a given nucleus) gives a contribution.

We will briefly explain the physics behind this approximation. If we consider the absorber nuclei, one may argue that the solid angle that one absorber nucleus subtends with respect to another absorber nucleus may not be very small if the two are close together. However, suppose we consider an absorber nucleus located by i and another absorber nucleus located at j ''downstream'' such that $j>i$. If we consider all the downstream absorber nuclei labeled by j, relative to the absorber nucleus located at i, we see that the optical path length for each path from j to i will be different. These different ''optical paths'' correspond to different phase factors for the radiation coming back to the ith absorber nucleus. When summing over all these downstream absorber nuclei, the contribution to re-excitation of the ith absorber nucleus by such processes is negligible. On the other hand, for absorber nuclei located upstream from i, i.e., $j<i$, all optical

paths from the source nucleus to the ith absorber nucleus are equal. Naturally these processes are very important and must be included in the calculation. It can be shown that the approximation made here consists of replacing, everywhere, ω_k by kc. This has already been done before,[4,5] without a thorough justification for this procedure.

Let us recall the equations resulting from the approximations noted above

$$A(\omega)=\frac{1}{\omega-\omega_0+i\frac{\Gamma}{2\hbar}}, \tag{45}$$

$$C_i(\omega)=-\frac{i\gamma_R}{2\hbar}\frac{1}{\omega-\omega_0'+i\frac{\Gamma}{2\hbar}}e^{ix_i\omega/c}A(\omega) -\frac{i\gamma_R}{2\hbar}\frac{1}{\omega-\omega_0'+i\frac{\Gamma}{2\hbar}}\sum_{j=1}^{i-1}e^{i(x_i-x_j)\omega/c}C_j(\omega), \tag{46}$$

$$A_d(\omega)=-\frac{i\gamma_R}{2\hbar}\frac{1}{\omega-\omega_0+i\frac{\Gamma}{2\hbar}}e^{ix_d\omega/c}A(\omega) -\frac{i\gamma_R}{2\hbar}\frac{1}{\omega-\omega_0+i\frac{\Gamma}{2\hbar}}\sum_{i=1}^{N}e^{i(x_d-x_i)\omega/c}C_i(\omega). \tag{47}$$

Going back to the time domain, using the definition Eq. (4), it can be shown easily from Eq. (45) that

$$a(t)=e^{-\Gamma t/2\hbar}. \tag{48}$$

This is nothing but the normal exponential decay of the source nucleus. The source should, of course, have this natural time behavior because, essentially, no radiation from the excited absorber nuclei ever returns to re-excite the source nucleus.

In the next section the probability of having the resonant-detector nucleus excited will be calculated. This leads to the probability of detecting and recording a conversion electron event in our conversion-electron resonant detector.

V. RECORDING A CONVERSION ELECTRON IN THE CONVERSION-ELECTRON RESONANT DETECTOR

Applying a recursion relation to Eq. (46), it can be shown[5] that

$$C_i(\omega)=-\frac{i\gamma_R}{2\hbar}\frac{1}{\omega-\omega_0'+i\frac{\Gamma}{2\hbar}}e^{ix_i\omega/c}A(\omega) \times\left[1-\frac{i\gamma_R}{2\hbar}\frac{1}{\omega-\omega_0'+i\frac{\Gamma}{2\hbar}}\right]^{i-1}. \tag{49}$$

By defining $\alpha'(\omega)$ as

$$\alpha'(\omega)=-\frac{i\gamma_R}{2\hbar}\frac{1}{\omega-\omega_0'+i\frac{\Gamma}{2\hbar}}, \tag{50}$$

Eq. (49) can be rewritten as

$$C_i(\omega)=\alpha'(\omega)e^{ix_i\omega/c}A(\omega)[1+\alpha'(\omega)]^{i-1}. \tag{51}$$

Substituting Eq. (51), together with Eq. (45) into Eq. (47) gives

$$A_d(\omega)=\alpha(\omega)\frac{1}{\omega-\omega_0+i\frac{\Gamma}{2\hbar}}e^{ix_d\omega/c} \times\left[1+\alpha'(\omega)\sum_{i=1}^{N}[1+\alpha'(\omega)]^{i-1}\right], \tag{52}$$

where $\alpha(\omega)$ is defined by

$$\alpha(\omega)=-\frac{i\gamma_R}{2\hbar}\frac{1}{\omega-\omega_0+i\frac{\Gamma}{2\hbar}}. \tag{53}$$

The probability of having a conversion electron produced by the resonant-detector nucleus is proportional to the probability of having the resonant-detector nucleus excited, namely $\int_{-\infty}^{+\infty}|A_d(\omega)|^2d\omega$. After some manipulations, this can be transformed into a series of integrals

$$I(\omega_0,\omega_0') =\int_{-\infty}^{+\infty}|A_d(\omega)|^2d\omega =\frac{\gamma_R^2}{4\hbar^2}\sum_{p=0}^{N}\binom{N}{p}C^p \times\int_{-\infty}^{+\infty}\frac{1}{\left[(\omega-\omega_0)^2+\frac{\Gamma^2}{4\hbar^2}\right]^2}\frac{1}{\left[(\omega-\omega_0')^2+\frac{\Gamma^2}{4\hbar^2}\right]^p}d\omega, \tag{54}$$

where $\binom{N}{p}$ is the binomial coefficient and where the constant C is defined by

$$C=\frac{\gamma_R}{4\hbar^2}(\gamma_R-2\Gamma). \tag{55}$$

The first integral corresponds to $p=0$. Its value is $\pi\hbar\gamma_R^2/\Gamma^3$. This constant value does not depend on ω_0', the frequency corresponding to the absorber nucleus. It therefore gives the contribution, to the conversion-electron production in the resonant detector, due to excitation of the resonant-detector nucleus by radiation coming directly from the source nucleus without exciting any absorber nuclei. In the detection signal this will give a constant background analogous to the background due to source radiation in a conventional Mössbauer experiment. The integral with $p=1$ corresponds to the contribution, of conversion-electron production in the resonant detector, if the absorber could be represented as having only one ''effective'' nucleus. This would correspond to the case when the absorber is very thin. It has been shown previously[2] that the Mössbauer spectrum is given by

$$I^{(1)}(\omega_0,\omega_0')=\frac{\pi\gamma_R^3(\gamma_R-2\Gamma)}{4\hbar\Gamma^3}\frac{12a^2+\Delta\omega_0^2}{(4a^2+\Delta\omega_0^2)^2}, \tag{56}$$

where a and $\Delta\omega_0$ are defined by the following expressions:

$$a=\frac{\Gamma}{2\hbar}, \tag{57}$$

$$\Delta\omega_0=\omega_0-\omega_0'. \tag{58}$$

The full width at half maximum of this distribution is 1.463 Γ. This value is close to 1.47Γ advanced in Ref. 7, where a heuristic approach has been used based on the calculation of the transmission integral. Also experiments[7] are presented where this narrowing effect was confirmed using ^{119}Sn. More recently this narrowing has been observed[8] again with ^{119}Sn. Thus there is experimental and theoretical evidence that, when using the conversion-electron resonant-detector scheme outlined above, the observed linewidth is appreciably smaller than the minimum linewidth obtained in a conventional Mössbauer experiment which is, as is well known, equal to 2Γ.

The other integrals, corresponding to thicker absorbers, can be calculated by means of contour integration. However, it is not possible to find a general recursion relation to calculate all these integrals. Their expressions become very lengthy and cumbersome even for small values of p. We have calculated them numerically. Figure 2 gives the spectrum for a few selected values of N.

It has been shown[5] that there is a relation between the parameter N representing the absorber thickness in the one-dimensional model and β, the actual resonant thickness of the absorber, which is

$$N=\frac{\beta\Gamma}{2f\gamma_R}. \tag{59}$$

The actual absorber thickness parameter β is given by $N_0 f\sigma_0 d$ where N_0 is the number of resonant nuclei/cm^3, f is the recoilless fraction, σ_0 is the maximum cross section evaluated on resonance and d is the thickness of the sample. Figure 3 gives the full width at half maximum of $I(\omega_0,\omega_0')$ as a function of the resonant thickness β of the absorber

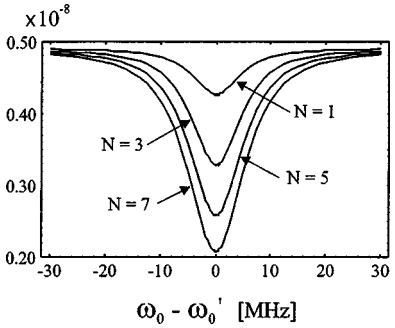

FIG. 2. Spectrum, obtained by counting the conversion electrons produced in the resonant detector using the geometry shown in Fig. 1, as a function of the difference in frequency $\omega_0-\omega_0'$. The resonant frequency of the source nucleus is ω_0 and the resonant frequency of the absorber nuclei is ω_0'.

using the one-dimensional model parameter N. One can see that line broadening, due to thickness effects, occurs as it does in conventional Mössbauer spectroscopy. However, the lines are narrower, using the conversion-electron resonant-detector method, than those obtained using the conventional Mössbauer setup for the same absorber. If high resolution is required in a Mössbauer experiment, it is advantageous to use the setup with a resonant detector.

VI. CONCLUSIONS

The quantum-mechanical theory for a particular Mössbauer-effect setup that uses a conventional source, a conventional absorber, and a nuclear-resonant detector is developed. The nuclear-resonant detector is assumed to consist of ground-state nuclei having the same environment as the source nuclei. Thus the source nuclei and the resonant-detector nuclei are in resonance when both are stationary. The absorber is moved relative to the source and detector.

Starting from the Schrödinger equation and applying a specific Fourier transformation, one obtains a set of linear algebraic equations for the complete system of nuclei, radiation, and conversion electrons. This system of equations can be solved analytically. The probability of having a nucleus in the resonant detector excited is calculated. This probability is proportional to the emission of γ rays or conversion electrons by the resonant-detector nucleus. When one counts the

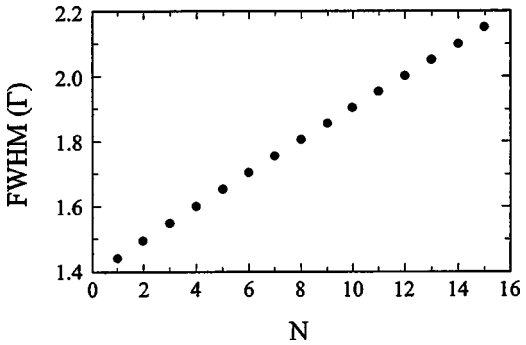

FIG. 3. The full width at half maximum of the spectrum obtained using the geometry shown in Fig. 1 as a function of the one-dimensional-model absorber-thickness parameter N.

conversion electrons produced in the resonant detector, as a function of the Doppler velocity of the absorber with respect to the stationary source and resonant detector, the minimum full width at half maximum of the spectrum is 1.463 Γ. This is appreciably less than the minimum linewidth, 2Γ, found using a conventional Mössbauer experimental setup. This minimum linewidth naturally corresponds to the case of a very thin absorber. As one considers thicker absorbers, lines broaden as in conventional Mössbauer spectroscopy. However, the lines obtained using the resonant-detector method are narrower than those found using the conventional Mössbauer setup. For experiments requiring very high resolution, the resonant-detector method outlined here is recommended.

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APPENDIX

Let us consider Eq. (35)

$$\left(\omega-\omega_0'+i\frac{\Gamma}{2\hbar}\right)C_i(\omega)=\frac{L}{2\pi\hbar^2}\int_{-\infty}^{+\infty}\frac{|H_k|^2e^{ikx_i}}{\omega-\omega_k+i\varepsilon}dk\,A(\omega)+\frac{L}{2\pi\hbar^2}\sum_{j=1,j\neq i}^{N}\int_{-\infty}^{+\infty}\frac{|H_k|^2}{\omega-\omega_k+i\varepsilon}\times e^{ik(x_i-x_j)}dk\,C_j(\omega)+\frac{L}{2\pi\hbar^2}\int_{-\infty}^{+\infty}\frac{|H_k|^2}{\omega-\omega_k+i\varepsilon}\times e^{ik(x_i-x_d)}dk\,A_d(\omega). \tag{A1}$$

Let us consider first the integral

$$I_{ij}=\frac{L}{2\pi\hbar^2}\int_{-\infty}^{+\infty}\frac{|H_k|^2e^{ik(x_i-x_j)}}{\omega-\omega_k+i\varepsilon}dk. \tag{A2}$$

With

$$\omega_k=|k|c, \tag{A3}$$

one has $\omega_k=-kc$ for $k<0$ and $\omega_k=kc$ for $k>0$. So Eq. (A2) becomes

$$I_{ij}=\frac{L}{2\pi c\hbar^2}\int_0^{+\infty}d\omega_k\frac{|H_k|^2e^{-i(x_i-x_j)\omega_k/c}}{\omega-\omega_k+i\varepsilon}+\frac{L}{2\pi c\hbar^2}\int_0^{+\infty}d\omega_k\frac{|H_k|^2e^{i(x_i-x_j)\omega_k/c}}{\omega-\omega_k+i\varepsilon}. \tag{A4}$$

Clearly $e^{\pm i(x_i-x_j)\omega_k/c}$ are rapidly oscillating functions of ω_k. Thus, there will not be an appreciable contribution to the integral unless one of the other factors shows a comparatively rapid variation, compensating for that due to $e^{\pm i(x_i-x_j)\omega_k/c}$. This occurs when $\omega_k=\omega$. As a consequence, the effective integration range can be reduced to a small region around ω. Therefore, both integrals can be extended from $-\infty$ to $+\infty$ and one has

$$I_{ij}=\frac{L}{2\pi c\hbar^2}\int_{-\infty}^{+\infty}d\omega_k\frac{|H_k|^2e^{-i(x_i-x_j)\omega_k/c}}{\omega-\omega_k+i\varepsilon}+\frac{L}{2\pi c\hbar^2}\int_{-\infty}^{+\infty}d\omega_k\frac{|H_k|^2e^{i(x_i-x_j)\omega_k/c}}{\omega-\omega_k+i\varepsilon}. \tag{A5}$$

These integrals depend on the positions of the nuclei. If $x_i<x_j$, the contour of the first integral has to be closed in the upper half plane and this integral gives

$$I_{ij}=\frac{Li}{c}\frac{|H_k|^2}{\hbar^2}e^{-i(x_i-x_j)\omega/c}. \tag{A6}$$

The second integral in Eq. (A5) is zero because the pole is not situated inside the contour, which has to be closed in the lower half plane. If $x_i>x_j$, the contours have to be closed differently, so that the first integral in Eq. (A5) is zero and the second one is

$$I_{ij}=-\frac{Li}{c}\frac{|H_k|^2}{\hbar^2}e^{i(x_i-x_j)\omega/c}. \tag{A7}$$

It can be shown easily that the radiative width in this approach is given by

$$\gamma_R=\frac{2L}{\hbar c}|H_k(\omega)|^2. \tag{A8}$$

So

$$I_{ij}=\frac{i\gamma_R}{2\hbar}e^{-i(x_i-x_j)\omega/c}\quad\text{for}\quad x_i<x_j, \tag{A9}$$

or

$$I_{ij}=-\frac{i\gamma_R}{2\hbar}e^{i(x_i-x_j)\omega/c}\quad\text{for}\quad x_i>x_j. \tag{A10}$$

The first integral on the right-hand side of Eq. (A1) can be calculated in a similar way. One finds

$$I_i=\int_{-\infty}^{+\infty}\frac{|H_k|^2 e^{ikx_i}}{\omega-\omega_k+i\varepsilon}dk=-\frac{i\gamma_R}{2\hbar}e^{ix_i\omega/c}. \qquad \text{(A11)}$$

The last integral, on the right-hand side of Eq. (A1), can be calculated similarly remembering that $x_i<x_d$. One finds

$$I_{id}=\frac{L}{2\pi\hbar^2}\int_{-\infty}^{+\infty}\frac{|H_k|^2}{\omega-\omega_k+i\varepsilon}e^{ik(x_i-x_d)}dk=\frac{i\gamma_R}{2\hbar}e^{-i(x_i-x_d)\omega/c}. \qquad \text{(A12)}$$

Substituting Eqs. (A9)–(A12) into Eq. (A1) gives

$$\left(\omega-\omega_0'+i\frac{\Gamma}{2\hbar}\right)C_i(\omega)=-\frac{i\gamma_R}{2\hbar}e^{ix_i\omega/c}A(\omega)-\frac{i\gamma_R}{2\hbar}\sum_{j=1}^{i-1}e^{i(x_i-x_j)\omega/c}C_j(\omega)+\frac{i\gamma_R}{2\hbar}\sum_{j=i+1}^{N}e^{-i(x_i-x_j)\omega/c}C_j(\omega)+\frac{i\gamma_R}{2\hbar}e^{-i(x_i-x_d)\omega/c}A_d(\omega). \qquad \text{(A13)}$$

This is Eq. (36) in the main text.

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