Temporal theory of resonant gamma-ray scattering from a nuclear isomer: calculation of scattering interaction time

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We carry out a temporal analysis of resonance fluorescence, taking accurate account of the energy dependence of the width of the initial state. This enables us to more completely determine the formation dynamics and spatial propagation of scattered-particle wave packets. We calculate the lifetime of the isomeric excited state of the scattering nucleus as a function of the energy of the scattered radiation. © *1995 American Institute of Physics.*

1. INTRODUCTION

In recent years, experimental techniques—particularly those employing high-power pulsed synchrotron sources of highly monochromatic resonance gamma rays—have made it possible to measure the energy and temporal distribution of resonantly scattered photons.[1] As a consequence, it has become necessary to derive a comprehensive description of the temporal and spatial dynamics of the resonance scattering of particles by a quantum system in a discrete quasistationary state.

The quantum theory of the resonance scattering of photons by a quasistationary excited level of an atom (nucleus) was developed by Weisskopf and Wigner.[2] Later results[3–8] determined the lifetime of the nucleus in the excited state, or as it is often put, the scattering collision time. According to the latter work, the lifetime of a virtual excited state can be defined to be the mean time delay of an incoming particle in the nuclear interaction region. The collision time was predicted to depend on the energy of the scattered particle. For an isolated resonance, that dependence is given by

$$\tau=2\tau_0\frac{(\gamma/2)^2}{(\Delta E)^2+(\gamma/2)^2}, \tag{1}$$

where ΔE is the difference between the energy of the incident particle and the nuclear transition energy, and $\gamma=1/\tau_0$ is the width of the metastable state. Note that Eq. (1) was derived under the assumption that the energy distribution of the original photons is much narrower than that of the nuclear level (γ). Subsequent work has been based on similar approximations, either implicit or explicit.

Experimentally, however, the most frequently encountered situation is one in which the width of the energy distribution for the scattered-particle wave packet is comparable to that of the quasistationary state. Hence, there is considerable practical interest in developing a theoretical description of the temporal and spatial laws governing the formation of a wave packet in resonance scattering, incorporating the actual spectrum of scattered radiation, and with no assumptions about the form taken by the distribution function for τ. Furthermore, it is important that the theoretical description of such a wave packet be consistent with the initial experimental conditions.

Our results are, on the one hand, intended to describe real experiments, while on the other they are of a rather general nature and are not tied to any specific experimental scheme. The resulting theory, despite being based specifically on the scattering of resonant photons, is general both in content and applicability.

2. THEORY

We first consider the physical statement of the problem of the interaction time during resonance scattering of resonant particles by a quasistationary level. With no loss of generality, we can for the sake of definiteness analyze nuclear resonance scattering of Mössbauer gamma rays. Since the resonance scattering will depend heavily on the energy distribution of the incoming radiation in the range of the natural width of the excited nuclear state, we assume a rather general expression for the energy distribution of the scattered radiation—a Lorentzian with arbitrary width γ_0. It follows from Wigner's definition[2] that to calculate the interaction time in the general case, we must find the wave function for the state (including the scattered photon) as a function of time and location in space $\mathbf{r}$, and then determine the wave packet's propagation time delay, as compared with the usual delay r/c, in the nuclear interaction region. To do a thorough job, we must solve the time-dependent Schrödinger equation for resonance scattering as a function of the spectral distribution of the incident flux. In the resonance approximation and at times $t\gg\omega_0^{-1}$, where ω_0 is the energy of the resonant nuclear transition, Heitler's method[5] enables us to solve this problem for an arbitrary spectral width of the scattered radiation.

The wave function for the system consisting of the nucleus plus electromagnetic field looks like

$$\Psi(t)=A(t)\varphi_{\mathrm{n}}^{(o)}|n_\lambda\rangle+\sum_\lambda B_\lambda(t)\varphi_{\mathrm{n}}^{*}|n_\lambda-1\rangle$$

$$+\sum_\lambda C_{\lambda\sigma}(t)\varphi_{\mathrm{n}}^{(o)}|n_\lambda-1,1_\sigma\rangle, \tag{2}$$

where $\varphi_n^{(o)}$ and φ_{n}^{*} are the nuclear wave functions in the ground and excited states, with energies E_0 and E_{n}, respectively; $|n_\lambda\rangle$ is the wave function of the electromagnetic field

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in an n_λ-photon state; $|n_\lambda-1, 1_\sigma\rangle$ is the wave function of the field in a state with one absorbed photon of frequency ω_λ and an emitted photon of frequency ω_σ. The Hamiltonian of the nucleus+field system can be written in the form[5]

$$H=H_n^{(o)}+H_f+\int d\mathbf{r}A(\mathbf{r},t)J(\mathbf{r},t), \tag{3}$$

where $H_n^{(o)}$ is the Hamiltonian describing the nuclear state, H_f is the electromagnetic field Hamiltonian, and the third term represents the interaction between the nucleus and the field, whose vector potential is $A(\mathbf{r},t)$. $J(\mathbf{r},t)$ is the nuclear transition current. We seek time-dependent functions (2) for the initial conditions $A(0)=1$, $B_\lambda(0)=C_{\lambda\sigma}(0)=0$. According to Ref. 5, we have for the Fourier transforms of these amplitudes

$$A(E)=\frac{1}{E-E_0+i\Gamma(E)/2},$$

$$B_\lambda(E)=\frac{H_{\lambda|o}}{(E-E_0+i\Gamma(E)/2)(E-E_\lambda+i\gamma/2)},$$

$$C_{\lambda\sigma}(E)$$

$$=\frac{H_{\lambda\sigma|\lambda}\times H_{\lambda|o}}{(E-E_{\lambda\sigma}+i\varepsilon)(E-E_0+i\Gamma(E)/2)(E-E_\lambda+i\gamma/2)},$$

$$\Gamma(E)=2i\sum_\lambda \frac{|H_{\lambda|o}|^2}{E-E_\lambda+i\gamma/2}. \tag{4}$$

Here $H_{\lambda|o}$ is the nuclear transition matrix element involving absorption of a photon λ from the incident flux; $H_{\lambda\sigma|\lambda}$ is the matrix element for a transition to a state with spontaneous emission of a photon σ, while the photon λ remains absorbed; γ is the total width of the excited nuclear state ($\hbar=c=1$).

Note that in the expression for the width $\Gamma(E)$ of the initial state—which consists of the incident radiation and a scattering nucleus in its ground state—the sum over λ denotes summation over frequencies and angles of the incident gamma-ray flux. The finite width of the scattered-particle spectrum can therefore be accommodated by evaluating this sum in the appropriate manner.

The matrix element $H_{\lambda|o}$ corresponds to a transition with absorption of a photon λ from the incident flux, so $|H_{\lambda|o}|^2$ is proportional to the number of photons n_λ. The summation Σ_λ can be replaced by an integral, averaging over all oscillators of the field. Then $\Gamma(E)$ becomes

$$\Gamma(E)=2i\int dE_\lambda \int d\Omega \frac{\rho(\omega_\lambda)\bar{n}_\lambda|H_{\lambda|0}(\omega_\lambda)|^2}{E-E_\lambda+i\gamma/2}, \tag{5}$$

where $E_\lambda=E_n-\omega_\lambda$ is the energy of the excited nuclear state, and a photon of frequency ω_λ has been absorbed. The mean number $\bar{n}_\lambda$ of photons of frequency ω_λ arriving per oscillator can be expressed in terms of the incident intensity:

$$\bar{n}_\lambda\omega_\lambda\rho(\omega_\lambda)d\Omega=I(\omega_\lambda). \tag{6}$$

We assume that the incoming radiation has a Lorentzian spectrum peaked at energy $\omega_0+\Delta$, with a width of γ_0:

$$I(\omega_\lambda)=\frac{I_0\gamma_0}{2\pi}\frac{1}{(\omega_\lambda-\omega_0-\Delta)^2+\gamma_0^2/4}, \tag{7}$$

where Δ is the offset between the center of the incident spectrum and that of the absorption spectrum, and $\omega_0=E_n-E_0$. Integrating (5) over energy E_λ, we obtain $\Gamma(E)$

$$\Gamma(E)=\frac{2i\alpha}{E-E_0+\Delta+i\Gamma_+/2}, \tag{8}$$

where $\alpha=|H_{\lambda|o}(\omega_\lambda)|^2(I_0/\omega_0)$, and $\Gamma_+=\gamma+\gamma_0$. Substituting this result into (4), taking Fourier transforms, and calculating squared moduli of the corresponding time-dependent amplitudes, we obtain (for $\gamma=\gamma_0$)

$$\begin{aligned}
|A(t)|^2=&e^{-2\eta t}[1-2f(\Delta^2-\gamma^2)/p]+2f\cos[(\Delta\\
&+2\xi)t]e^{-\gamma t}(\Delta^2-\gamma^2)/p+4f\sin[(\Delta\\
&+2\xi)t]e^{-\gamma t}\Delta\gamma/p,\\
|B(t)|^2=&\sum_\lambda|B_\lambda(t)|^2=2f\{e^{-2\eta t}-e^{-\gamma t}-e^{-\gamma t}\cos[(\Delta\\
&+2\xi)t]+e^{-\gamma t+\eta t}\cos(\xi t)+(\gamma/\Delta)\\
&\times\sin(\xi t)e^{-\gamma t+\eta t}-(\gamma/\Delta)\sin[(\Delta+2\xi)t]e^{-\gamma t}\},\\
|C(t)|^2=&\sum_{\lambda,\sigma}|C_{\lambda,\sigma}(t)|^2=1-e^{-2\eta t}(t+4f\gamma^2/p)\\
&+4f\cos[(\Delta+2\xi)t]e^{-\gamma t}\gamma^2/p+2fe^{-\gamma t}\\
&-2f\cos(\xi t)e^{-\gamma t+\eta t}+2f\gamma(\gamma^2-\Delta^2)/(\Delta/p)\\
&\times\sin[(\Delta+2\xi)t]e^{-\gamma t}-(2f\gamma/\Delta)\\
&\times\sin(\xi t)e^{-\gamma t+\eta t},
\end{aligned} \tag{9}$$

where $p=\Delta^2+\gamma^2$, $f=\alpha/p$, $\eta=\gamma f$, and $\xi=\Delta f$. In integrating over E_λ in (9), we have allowed for the spectral distribution of the incident radiation. To make the meaning of these expressions (9) clear, we have plotted them in Figs. 1–3 for various values of Δ.

In deriving (9), it should be noted that in addition to the resonance approximation, we have taken advantage of the smallness of η relative to γ. This corresponds to assuming low probability of resonance fluorescence per unit time compared with the inverse of the total lifetime of the isomeric nuclear state. Since the scattering probability is proportional to the mean number of incident photons $\bar{n}_\lambda$, this assumption is equivalent to assuming $\bar{n}_\lambda\ll 1$, which will hold unless the incoming photons come from a laser.

The above expressions completely describe resonance scattering by an isolated level, and imply that the probability of finding the system in a given state depends on the time in a nonexponential fashion—the time dependence of all three probabilities is basically dictated by two exponentials and a set of oscillatory functions. In two cases, the time dependence simplifies: when the width of the energy distribution of the incident photon flux is much larger than that of the isomeric level, and when scattering takes place far from resonance ($\gamma\ll\Delta$). For an illustration of the first case, consider Eq. (5) for $\Gamma(E)$. Since we are dealing with values of the

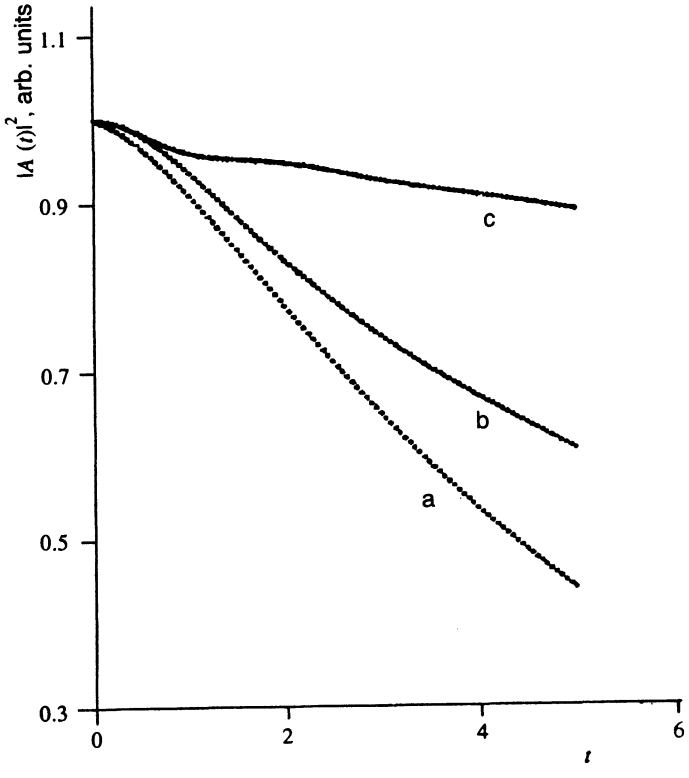

FIG. 1. Plots of $|A(t)|^2$ at various values of shift a) $\Delta=0$; b) $\Delta=\gamma$; c) $\Delta=3\gamma$.

energy E close to E_0, only photons of energy ω_λ close to ω_0 (to within γ) can be absorbed; moreover, since H and ρ depend weakly on ω_λ, we can substitute $\omega_\lambda=\omega_0$ in both. Then Γ will be independent of E. In the second case, $\Delta^2/\gamma^2\gg 1$ (as before, we assume an isolated resonance with $\Delta\ll E_0$). We see from (9) that for $t\gg 1/\gamma$ in this limiting case, the expressions for the probabilities, which were derived to order α/Δ^2, retain the terms proportional to an exponential with exponent η, which agrees with the transition probability per unit time for the process usually referred to as Rayleigh scattering:

$$|A(t)|^2=(1-2\alpha/\Delta^2)e^{-2\eta t},\quad |B(t)|^2=2\alpha/\Delta^2 \times e^{-2\eta t},\quad |C(t)|^2=1-e^{-2\eta t}. \tag{10}$$

This shows that $|B(t)|^2$, which gives the probability of finding the nucleus in the excited state at time t, is a factor α/Δ^2 down from $|A(t)|^2$ and $|C(t)|^2$.

For arbitrary Δ and t, we can use (9) and neglect terms proportional to α^2 to obtain the probability per unit time of finding the nucleus in the corresponding state:

$$A'=\frac{d|A(t)|^2}{dt}=-2\eta+2f\gamma\cos(\Delta t)e^{-\gamma t} -2f\Delta\sin(\Delta t)e^{-\gamma t},$$

$$B'=\frac{d|B(t)|^2}{dt}=(2fp/\Delta)\sin(\Delta t)e^{-\gamma t},$$

$$C'=\frac{d|C(t)|^2}{dt}=2\eta-2f\gamma\cos(\Delta t)e^{-\gamma t}-(2f\gamma^2/\Delta) \times\sin(\Delta t)e^{-\gamma t}. \tag{11}$$

Here C' is proportional to the experimentally observed intensity of the scattered radiation or the intensity of converted electrons. It can be shown that to order α, $C'=\gamma\Sigma_\lambda|B_\lambda(t)|^2$, i.e., it is proportional to the population of the excited state. The observed intensity can also be calculated by starting with B' and integrating it over all nuclear illumination onset times from 0 to t.

We now examine in more detail the calculation of the collision time. According to the classic work of Weisskopf and Wigner[2] and Smith,[8] the collision time can be defined as the difference between the time spent by an incident particle in the interaction region and the time needed to traverse that region in the absence of interaction, in the limit as the region over which all possible particle-scattering trajectories are summed goes to infinity. This definition of the collision time corresponds uniquely to the time-independent statement of the scattering problem, and cannot fully accommodate the dynamics of wave packet formation during the collision process. At the same time, the time-dependent theory enables one to allow for the spatial and temporal dynamics of wave packet formation, and thus more accurately to determine the delay associated with excitation of an isomeric nuclear state of the scatterer.

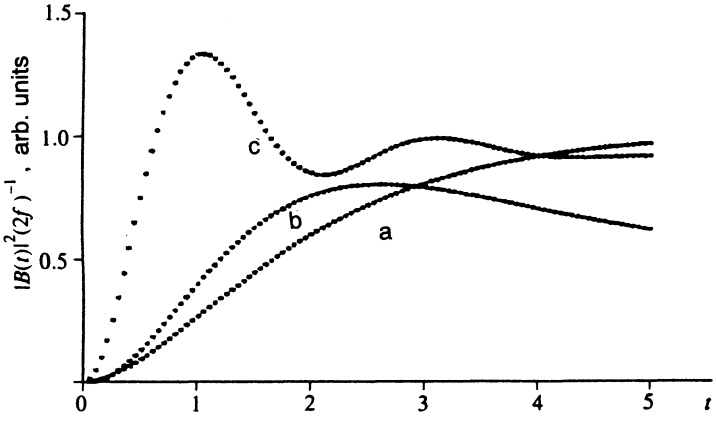

FIG. 2. Plots of $|B(t)|^2(2f)^{-1}$ at various values of shift a) $\Delta=0$; b) $\Delta=\gamma$; c) $\Delta=3\gamma$.

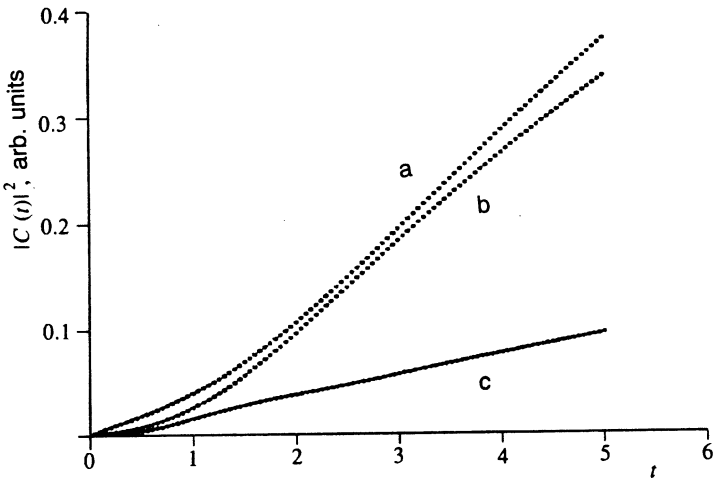

FIG. 3. Plots of $|C(t)|^2$ at various values of shift a) $\Delta=0$; b) $\Delta=\gamma$; c) $\Delta=3\gamma$.

To obtain an expression for the collision time, we make use of the wave function $\Psi_{\lambda\sigma}(\mathbf{r},t)$ of the scattered radiation, which is related to the amplitude $C_{\lambda\sigma}(t)$ by a Fourier transform:

$$\Psi_{\lambda\sigma}(\mathbf{r},t)=\int d\mathbf{k}_\sigma \exp[i(\mathbf{k}_\sigma\mathbf{r}-\omega_\sigma t)]C_{\lambda\sigma}(t). \tag{12}$$

Calculating this integral, we can show that $\Psi_{\lambda\sigma}(\mathbf{r},t)$ takes the form

$$\Psi_{\lambda\sigma}(\mathbf{r},t)=\begin{cases}2\pi/rH_{\lambda\sigma|\lambda}B_\lambda(\tau), & \tau\geqslant 0,\\ 0, & \tau<0\end{cases} \tag{13}$$

where $\tau=t-r/c$. Since $\Sigma_\lambda|\Psi_{\lambda\sigma}(\mathbf{r},t)|^2$ is the probability of detecting a scattered photon at a distance r from the nucleus at time t, the mean collision time is given by

$$\bar{t}=\int_0^\infty t\sum_\lambda |\Psi_{\lambda\sigma}(\mathbf{r},t)|^2dt \Big/ \int_0^\infty \sum_\lambda |\Psi_{\lambda\sigma}(\mathbf{r},t)|^2dt. \tag{14}$$

By virtue of (13), we can transform from $\Psi_{\lambda\sigma}(\mathbf{r},t)$ to $B_\lambda(\tau)$ in (14). As in (9), for an arbitrary ratio between the width of the incident spectrum and that of the isomeric level of the scattering nucleus, we have

$$\begin{aligned}|B(t)|^2=F+\{&(e^{-2\eta t}-e^{-\gamma t})\Gamma_+(\Delta^2+\Gamma_-^2/4)/\gamma-2(\Delta^2\\ &+\Gamma_+\Gamma_-/4)[\cos[(\Delta+2\xi)t]e^{-\Gamma_+ t/2}\\ &-e^{-\gamma t+\eta t}\cos(\xi t)]+2\gamma_0\Delta[e^{-\gamma t+\eta t}\sin(\xi t)\\ &-e^{-\Gamma_+ t/2}\sin[(\Delta+2\xi)t]]\},\\ F=\alpha&[(\Delta^2+\Gamma_+\Gamma_-/4)^2+\Delta^2\gamma_0^2]^{-1},\end{aligned} \tag{15}$$

where $\Gamma_-=\gamma-\gamma_0$. For arbitrary γ and γ_0, the expression for $|A(t)|^2$ looks like (9) if we replace γ with $\Gamma_+/2$. Making use of (15), we also obtain an expression for $\bar{t}$ from (14) to zeroth order in α:

$$\begin{aligned}\bar{t}=&\frac{\Gamma_+}{\Delta^2+\Gamma_+^2/4}\\ &+\frac{\Gamma_-}{2}\frac{\Delta^4+\Delta^2(\gamma+\gamma_0^2)/2+(\Gamma_+^2/2)(\Gamma_-^2/4)}{(\Gamma_+/2)(\Delta^2+\Gamma_+^2/4)(\Delta^2+\Gamma_-^2/4)\gamma}.\end{aligned} \tag{16}$$

For $\gamma=\gamma_0$,

$$\bar{t}=2\frac{\gamma}{\Delta^2+\gamma^2}-\alpha\frac{2\Delta^2(\Delta^2+5\gamma^2)}{\gamma(\Delta^2+\gamma^2)^3}. \tag{17}$$

The first term in (17) is a generalized expression for $\bar{t}$.[8] We emphasize here that (16) and (17) represent not the total duration of the scattering process but the time delay due to resonant excitation of the scattering nucleus. This comes about because in using (15) we have subtracted the term proportional to $\exp(-2\eta t)$ from $|B(t)|^2$.

Equation (13) tells us that the probability of detecting a scattered photon at a point $\mathbf{r}$ and time t is proportional to $\Sigma_\lambda|B(t)|^2$, so from here on we analyze the temporal dependence in terms of this quantity. For simplicity, we assume the $\gamma=\gamma_0$. From (9), the monotonic dependence of $|B(t)|^2$ on time is dictated, as noted above, by the two exponentials with arguments proportional to η and γ. Since we are also assuming that $\eta\ll\gamma$, we can identify a time interval $\gamma^{-1}\ll t\ll\eta^{-1}$ over which the temporal evolution of $|B(t)|^2$ is governed solely by $\exp(-2\eta t)$. At times $t\leqslant\gamma^{-1}$, where $\exp(-2\eta t)\sim 1$, the dominant influence is $\exp(-\gamma t)$.

In temporal experiments in which the initial state of the system is fixed at $t=0$, one measures the total duration of the resonance scattering process (which we denote by $\bar{T}$). The above analysis of the present solution shows that the duration of measurements Δt and the calibration accuracy of the temporal origin of those measurements can have a significant influence on the results. The experimental mean duration of resonance scattering is given by an integral over Δt:

$$\bar{T}=\int_{\Delta t} t\sum_\lambda |\Psi_{\lambda\sigma}(\mathbf{r},t)|^2dt \Big/ \int_0^\infty \sum_\lambda |\Psi_{\lambda\sigma}(\mathbf{r},t)|^2dt. \tag{18}$$

Changing the limits of integration in this expression can alter the result significantly. For example, if the lower limit is greater than γ^{-1}, the integrand of (18) will be dominated by the exponential with argument $\sim\eta$, while the contribution due to terms $\sim\exp(-\gamma t)$ will be relatively minor. This implies that the temporal origin must be specified to an accuracy of at least γ^{-1}. On the other hand, if we make the upper limit of the interval Δt much greater than $\gamma^{-1}(\sim\eta^{-1}$, for

example), terms $\sim\exp(-2\eta t)$ will also figure prominently. As a result, the optimal Δt is of order γ^{-1}. We then have $\bar{T}\approx\bar{t}$, and we can actually measure the duration of the resonance process itself in such an experiment.

Interest in the direct experimental determination of the resonance-scattering interaction time using nonmonochromatic radiation has risen in recent years.[1,9] The theoretical calculations in Refs. 1 and 9 were largely qualitative, and in our view the analysis of experimental considerations was incomplete, although the derived values of $\bar{t}$ do depend on the energy of the incident radiation, and they differ from γ^{-1} in the case of strict resonance, which is consistent with our own results.

We now calculate the probability amplitude for detecting a nucleus in an excited state at point $\mathbf{r}$ and time t. The wave function of that state is

$$\Psi_\lambda(\mathbf{r},t)=\int d\mathbf{k}_\lambda B_\lambda(t)\exp[i(\mathbf{k}_\lambda\mathbf{r}-E_\lambda t)]. \tag{19}$$

We assume that the incoming flux is incident along the Z axis (i.e., $k_x=k_y=0$, $k_z\neq 0$) and that its spectrum is given by Eq. (7). Carrying out the integration in (19), we find

$$\Psi_\lambda(z,t)=-\frac{\xi-i\eta}{\Delta+i\Gamma_+/2+2\xi-2i\eta}\exp[i\tau\Delta-\tau\gamma_0/2 - \gamma t/2]-\exp[-i\xi\tau-\eta\tau-(\gamma z/2)/c], \tag{20}$$

where c, as usual, is the speed of light, and $\tau=t-z/c$. To a first approximation in α, the probability is

$$|\Psi_\lambda(z,t)|^2=(\Delta^2+\Gamma_+^2/4)^{-1}\{(\Delta^2+\Gamma_+^2/4+2\Delta\xi - \Gamma_+\eta)\exp[-\gamma z/c-2\eta\tau]+2[(\Delta\xi - \eta\Gamma_+/2)\cos(\Delta\tau)+(\Delta\eta + \xi\Gamma_+/2)\sin(\Delta\tau)]\exp[-\tau\Gamma_+/2-\gamma t]\}. \tag{21}$$

The argument z in (21) runs from Z_1 to Z_2 ($Z_2-Z_1=d$, where d is the target thickness). The probability vanishes at all other values of Z. To zeroth order in α, we clearly obtain the usual exponential dependence on the depth of penetration of radiation in matter.

3. CONCLUSION

To conclude, we briefly discuss the principal results. The expressions derived here for the overall wave function describing the resonance scattering of a photon by a quasistationary level indicate that its temporal behavior depends strongly on the energy distribution of the incident photon flux. We especially emphasize that averaging over the energy spectrum must take place in two steps: we first average over the width $\Gamma(E)$ of the initial state, and then, transforming from the E-representation to the t-representation of the amplitudes, we average the squared modulus of the wave function over the spectrum of the incident radiation. Since the energy dependence of $\Gamma(E)$ for an arbitrary width of the incident spectrum is pole-like (which only ceases to be the case for a very broad distribution, $\gamma_0\gg\gamma$), additional exponential functions of the time enter into the amplitudes when one goes from the E-representation to the t-representation.

Finally, we have what in our view is the most important conclusion, which follows from an analysis of Eq. (21). The probability at time t of finding a nucleus in the excited state at some point z inside the scattering target, as we have already pointed out, is governed by two exponentials with arguments of order γt and $\eta\tau$. Since it is essentially always true that $\gamma\gg\eta$, the probability of detecting the excited state in the target at $t\gg\gamma^{-1}$ as well is appreciably greater than zero. This can have an extremely important impact on the dynamics of wave function formation and propagation in a resonant medium. This is particularly so as the results derived here imply that long-lived space-time oscillations are superimposed on the exponential decay.

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