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Theory of nuclear resonant scattering of synchrotron radiation in the presence of diffusive motion of nuclei

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A general theory of the time dependence of nuclear resonant forward scattering (spatially coherent) as well as 4π scattering (incoherent) is presented for a system where resonant nuclei are moving diffusively. As in Mössbauer absorption spectroscopy the diffusive motion is taken into account by employing the Van Hove correlation function. However, in the case of scattering the role of correlations turns out to be more complicated. The general formulas are obtained and used to analyze the case of free diffusion.

I. INTRODUCTION

It was immediately realized after the discovery of the Mössbauer effect that it provides a new opportunity to explore the dynamics of atomic motions. The high energy resolution of Mössbauer spectroscopy allows it to detect very slow motion, unattainable by other techniques. It is especially applicable to a diffusive motion which occurs in liquid and solid materials. The analysis of Mössbauer spectra affected by diffusion is based on early work by Singwi and Sjölander.[1] The γ-ray absorption cross section for a single nucleus diffusing in a system contains the γ-ray phase factor $\exp(i\boldsymbol{k}\cdot\boldsymbol{r})$, ($\boldsymbol{k}$ is the γ-ray propagation vector, $\boldsymbol{r}$ is a position of the absorbing nucleus) and the self-correlation function $G(\boldsymbol{r},t)$. This function describes the probability for finding the absorbing nucleus at a position $\boldsymbol{r}$ at time t, if it was at the origin at time $t=0$. The phase of the incident wave seen by a nucleus is sensitive to the nuclear motion $\boldsymbol{r}(t)$. The explicit form of the self-correlation function has to be found in each particular case of diffusive motion. This might be an isotropic and quasicontinuous process as in liquids or a unisotropic jumpy motion from one site to another as in solids. The motion might be spatially unlimited (free) or might be confined to a certain region. In the latter case it is decribed as bounded diffusion within a cage. Diffusive motion might be both translational and rotational.

In any case diffusion manifests itself in the profile of the Mössbauer absorption spectra through the broadening and changes in a shape of the resonance lines (for a review, see Refs. 2–5). As for free diffusion it is a simple broadening of the Lorentzian line.[3] In the case of bounded diffusion a superposition of different broadened lines may be expected.[6,7] Such spectra are especially characteristic for biological samples.

Today a new powerful technique for studying nuclear γ resonances is rapidly developing, namely, the spectroscopy in time domain with the use of nuclear scattering of synchrotron radiation (SR). A wide range of nuclear and solid-state parameters can be explored by studying the time dependencies of the intensity of the delayed products of nuclear deexcitation.[8,9]

The goal of the present work is to develop the theory of resonant scattering of SR by a system where resonant nuclei are moving diffusively. The new elements of the theory are related to the character of the scattering process. Regarding the process of scattering one has to take into account both the process of nuclear resonant absorption and the process of emission. In addition, because of the interference of the wavelets scattered by different nuclei the spatial correlation of the excitation over the ensemble of nuclei should also be considered.

The advantage of the use of SR is that one is able to detect both a coherent signal in the forward direction and an incoherent signal into 4π. The incoherent channel should be especially informative about the presence of intensive diffusive motion. The latter leads to strong broadening of nuclear resonance which cannot be revealed by the usual Mössbauer spectroscopy.

In Sec. II the basic wave equation is presented with accounting for diffusive motion in the general form. As an example we consider the most simple case of free diffusion. In Sec. III the time dependence of the forward scattering is evaluated for the general case and in particular for the model of free diffusion. In Sec. IV the time dependence of the 4π scattering intensity is found. Section V presents the results of computer simulation of the time dependence of the intensity of forward and 4π scattering and their analysis.

II. WAVE EQUATION FOR THE TRANSMITTED BEAM

To decribe the electromagnetic field of γ radiation inside the matter we use Maxwell's wave equation.[10] In terms of a space and time Fourier component of the electric-field vector $\boldsymbol{E}(\boldsymbol{k},\omega)$, which presents the amplitude of a plane monochromatic wave having the wave vector $\boldsymbol{k}$ and frequency ω, this equation can be written in the form

$$[\,k^2 - K^2\,]\,\boldsymbol{E}(\boldsymbol{k},\omega) - \boldsymbol{k}\,(\,\boldsymbol{k}\cdot\boldsymbol{E}\,(\boldsymbol{k},\omega)\,) = \frac{4\pi i\omega}{c^2}\,\boldsymbol{j}\,(\boldsymbol{k},\omega)\,, \tag{1}$$

where $K = \omega/c$, c is the light velocity in the vacuum, $\boldsymbol{j}\,(\boldsymbol{k},\omega)$ is the Fourier component of the induced current density which is a quantum-mechanical average of the current density operator. It has contributions from both the electronic and the nuclear subsystems. The nuclear part is directly related to the nuclear transition current which has the form of an oscillating multipole. The Fourier component of the nuclear current is determined by the following expression:

$$\boldsymbol{j}_n\,(\boldsymbol{k},\omega) = \int dt \sum_a \exp\,(-i\boldsymbol{k}\cdot\boldsymbol{r}_a + i\omega t) \times \langle\psi_a(t)\,|\,\hat{\boldsymbol{j}}\,(\boldsymbol{k})\,|\,\psi_a(t)\rangle\,. \tag{2}$$

Here $\langle\psi_a(t)\,|\,\hat{\boldsymbol{j}}\,(\boldsymbol{k})\,|\,\psi_a(t)\rangle$ is the time-dependent current density for the ath nucleus with $\psi_a(t)$ as the wave function in the transition state, that is the superposition of the excited and the ground states, and $\hat{\boldsymbol{j}}\,(\boldsymbol{k})$ as the current density operator in $\boldsymbol{k}$ space; summing is performed over the nuclei in a unit volume.

The current density for an individual nucleus in the first nonvanishing order of perturbation theory[10] is equal to

$$\langle\psi_a(t)\,|\,\hat{\boldsymbol{j}}\,(\boldsymbol{k})\,|\,\psi_a(t)\rangle = -\frac{i}{(2I_0+1)\hbar}\sum_{ge}\int_{-\infty}^{t} dt'\,\exp\{i\,(-\omega_{eg} + i\Gamma_0/2\hbar)\,(t-t')\}\langle g\,|\,\hat{\boldsymbol{j}}\,(\boldsymbol{k})\,|\,e\rangle\langle e\,|\,\hat{V}_a(t')\,|\,g\rangle\,. \tag{3}$$

Equation (3) presents the convolution of the excitation and deexcitation processes, with t' and $t > t'$ as the excitation and deexcitation times, respectively. Here $|\,g\rangle$ and $|\,e\rangle$ are space parts of the wave functions of the ground and excited states of a nucleus; summing over transitions between the hyperfine levels of the ground and the excited states is performed; I_0 is the nuclear spin in the ground state; ω_{eg} is the resonance frequency of the corresponding transition; Γ_0 is the natural energy width of the nuclear level. The time-dependent Hamiltonian of the interaction of the ath nucleus with the γ ray, $\hat{V}_a(t)$, is defined in a standard way where the Coulomb gauge with a zero scalar potential is used so that the vector potential $\boldsymbol{A}(\omega) = -ic\omega^{-1}\boldsymbol{E}(\omega)$. In order to evaluate $\boldsymbol{j}\,(\boldsymbol{k},\omega)$ the operator $V_a(t')$ should be presented in the form of the expansion of the current density operator and the vector potential over their space and time Fourier components

$$\hat{V}_a(t') = -\frac{1}{c}\int d\boldsymbol{r}\,\hat{\boldsymbol{j}}\,(\boldsymbol{r}-\boldsymbol{r}_a)\,\boldsymbol{A}(\boldsymbol{r},t') = i\int\frac{d\omega'}{2\pi\omega'}\int\frac{d\boldsymbol{k}'}{(2\pi)^3}\,\exp\,(i\boldsymbol{k}'\cdot\boldsymbol{r}_a - i\omega' t')\,\boldsymbol{j}^*(\boldsymbol{k}')\,\boldsymbol{E}(\boldsymbol{k}',\omega'). \tag{4}$$

Substituting (4) in (3) and then (3) in (2) yields

$$j_n^s(\boldsymbol{k},\omega) = \int\frac{d\boldsymbol{k}'}{(2\pi)^3}\int\frac{d\omega'}{2\pi}\sum_{ges'}\frac{C^s_{ge}(\boldsymbol{k})\,C^{s'*}_{ge}(\boldsymbol{k}')}{\hbar\omega'\,(2I_0+1)}\,E^{s'}(\boldsymbol{k}',\omega') \times\int dt\,\exp\{i(\omega-\omega')\,t\}\int_{-\infty}^{t} dt'\,e\,(\omega'-\omega_{eg},t-t')\,F(\boldsymbol{k},\boldsymbol{k}',t,t')\,, \tag{5}$$

where the indexes s', s denote the polarization states of the wave field before and after scattering and

$$F(\boldsymbol{k},\boldsymbol{k}',t,t') = \sum_a \exp\{-i\boldsymbol{k}\cdot\boldsymbol{r}_a(t) + i\boldsymbol{k}'\cdot\boldsymbol{r}_a(t')\}\,. \tag{6}$$

Here the time dependence of a nuclear position is taken into account in the explicit form. The following notations are used in (5) and below:

$$\begin{aligned} C^s_{ge}(\boldsymbol{k}) &= \langle g\,|\,\hat{j}^s(\boldsymbol{k})\,|\,e\rangle\,, \\ e\,(\omega,t) &= \exp\{i(\omega + i\Gamma_0/2\hbar)\,t\}\,. \end{aligned} \tag{7}$$

Before proceeding to develop this outline one must evaluate the sum over nuclear positions in (6). The result of this evaluation depends on the particular system considered. In this paper we restrict ourselves to the most simple system, namely, the irregular nuclear ensemble where the positions of different nuclei are not correlated in space (liquids, glasses, amorphous metals, etc.). The motion of nuclei may be decomposed into the fast motion near the mean position (thermal vibrations) and the slow motion of the mean position due to diffusion, as compared to the lifetime of the excited state of the nucleus. The account of the fast motion yields the Lamb-Mössbauer factor. As for the slow motion let us consider first the very slow one so that $\boldsymbol{r}_a(t') \approx \boldsymbol{r}_a(t)$ during the lifetime of the intermediate excited state. In this case we can use the well-known random-phase approximation

$$\sum_a \exp\{-i(\boldsymbol{k}-\boldsymbol{k}')\cdot\boldsymbol{r}_a\} = N\Delta_{\boldsymbol{k}\boldsymbol{k}'} \approx \frac{(2\pi)^3}{V_0}\delta(\boldsymbol{k}-\boldsymbol{k}'), \tag{8}$$

where $N = 1/V_0$ is the number of nuclei in the unit volume, V_0 is the target volume corresponding to one nucleus, $\Delta_{\boldsymbol{k}\boldsymbol{k}'}$ is the delta symbol which differs from zero only for $\boldsymbol{k}=\boldsymbol{k}'$, $\delta(\boldsymbol{k}-\boldsymbol{k}')$ is the delta function. In (8) the standard transformation from summing to integrating is used.

This formula shows that the coherent scattering of γ rays by the irregular nuclear ensemble exists only in the forward direction. We shall keep the result concerning the spatial structure of the coherent field also in the case where the characteristic time of diffusive motion is comparable with the nuclear lifetime and consider only $\boldsymbol{k}=\boldsymbol{k}'$. Then

$$F(\boldsymbol{k},\boldsymbol{k}',t,t') = \frac{(2\pi)^3}{V_0}\,\delta(\boldsymbol{k}-\boldsymbol{k}')\,f_{\mathrm{LM}}(\boldsymbol{k})\,F(\boldsymbol{k},t,t'), \quad (9)$$

where $f_{\mathrm{LM}}(\boldsymbol{k})$ is the Lamb-Mössbauer factor accounting for the fast motion of nuclei and the function

$$F(\boldsymbol{k},t,t') = \frac{1}{N}\sum_a \exp\{-i\boldsymbol{k}[\boldsymbol{r}_a(t)-\boldsymbol{r}_a(t')]\}, \quad (10)$$

accounting for the slow motion.

Thus, in this approximation we have the following expression for the current density:

$$j_n^s(\boldsymbol{k},\omega) = \int \frac{d\omega'}{2\pi}\,E^s(\boldsymbol{k},\omega')\,f_{\mathrm{LM}}(\boldsymbol{k})\sum_{ge}\frac{|C^s_{ge}(\boldsymbol{k})|^2}{\hbar\omega'(2I_0+1)V_0}\int dt\exp\{i(\omega-\omega')t\}\int_{-\infty}^{t} dt'e(\omega'-\omega_{eg},t-t')F(\boldsymbol{k},t,t'). \quad (11)$$

Here we assume additionally that the polarization state is conserved in the forward scattering. As a consequence of our assumption the "fast" and the "slow" nuclei motions are factorized and enter in a different way into the nuclear current density. Later we shall see how the slow motion will affect the time dependence of the scattering intensity. The function (10) describes the correlation between the positions of one and the same particle at different times averaged over all particles in the unit volume. This averaging is equivalent to the thermodynamic averaging of the motion of one particle.

In the following we shall assume that the function F depends only on the time difference $t-t'$, so that $F=F_s(\boldsymbol{k},t-t')$. This function can be associated with the well-known Van Hove function[11] $G_s(\boldsymbol{r},t)$ (see also Ref. 1) in the following way:

$$F_s(\boldsymbol{k},t) = \int d\boldsymbol{r}\,\mathrm{e}^{-i\mathbf{k}\cdot\mathbf{r}}\,G_s(\boldsymbol{r},t),$$

$$G_s(\boldsymbol{r},t) = \int \frac{d\boldsymbol{k}}{(2\pi)^3}\,\mathrm{e}^{i\mathbf{k}\cdot\mathbf{r}}\,F_s(\boldsymbol{k},t). \quad (12)$$

This assumption is appropriate when there are no correlations between the positions of different particles so that the mean position is the same at all times. A quite different situation arises when all nuclei of the system are vibrating in unison.[12] In this case the assumption is not correct.

Using the approximation $F(t,t')=F_s(t-t')$ one can immediately deduce that the frequency is conserved in the scattering. This result follows from the fact that the integral over t' does not depend on time [see (16)]. Then the integral over t equals $2\pi\,\delta(\omega'-\omega)$. As a consequence $E^s(\boldsymbol{r},t)$ and $j^s(\boldsymbol{r},t)$ have the identical time dependencies. Finally, from (11) we arrive at the following relation between the nuclear current density and the field:

$$j_n^s(\boldsymbol{k},\omega) = \frac{c^2K^2}{4\pi i\omega}\,g_{00}^{ss(n)}(\boldsymbol{k},\omega)\,E^s(\boldsymbol{k},\omega), \quad (13)$$

where

$$g_{00}^{ss(n)}(\boldsymbol{k},\omega) = i\,\frac{\Gamma_0}{2\hbar}\sum_{ge} B_{00}^{ss}(ge)\,\varphi(\boldsymbol{k},\omega-\omega_{eg}),$$

$$B_{00}^{ss}(ge) = \frac{8\pi f_{\mathrm{LM}}(\boldsymbol{k})}{\omega^2V_0(2I_0+1)\,\Gamma_0}\,|\,C^s_{ge}(\boldsymbol{k})\,|^2, \quad (14)$$

where we introduce the universal function $\varphi(\boldsymbol{k},\omega)$ which takes into account the diffusive motion. It is defined by

$$\varphi(\boldsymbol{k},\omega) = \int_{-\infty}^{t} dt'\exp\{i(\omega+i\Gamma_0/2\hbar)(t-t')\}F_s(\boldsymbol{k},t-t').$$

The factor $g_{00}^{ss(n)}$ represents the nuclear resonant susceptibility which is closely related to the scattering amplitude.

We note here that the electronic current density is given by Eq. (13) as well by means of replacing the resonant susceptibility $g_{00}^{ss(n)}(\boldsymbol{k},\omega)$ by the nonresonant one. The total susceptibility for scattering into the forward direction is the sum of the two contributions

$$g_{00}^{ss}(\omega) = g_{00}^{ss(n)}(\omega) + \chi_{00},$$

$$\chi_{00} = \frac{1}{V_0K}\left(-\frac{4\pi}{K}r_0 + i\sigma_e\right), \quad (15)$$

where $r_0=e^2/mc^2$ is the classical radius of electron, σ_e is the total cross section of electonic inelastic scattering.

Thus, the diffusive motion is described in our theory by the function $\varphi(\boldsymbol{k},\omega)$. This function can be rewritten in a more simple form by changing the variable of integration $t''=t-t'$, $t''\to t$

$$\varphi(\boldsymbol{k},\omega) = \int_0^{\infty} dt\,\exp(i\omega t-\Gamma_0 t/2\hbar)\,F_s(\boldsymbol{k},t). \quad (16)$$

Actually it describes the resonant factor averaged by the correlation function. In order to see it we expand the function $F_s(\boldsymbol{k},t)$ in the Fourier integral

$$F_s(\boldsymbol{k},t) = \int \frac{d\tilde{\omega}}{2\pi}\,\mathrm{e}^{-i\tilde{\omega}t}\,F_s(\boldsymbol{k},\tilde{\omega}),$$

$$F_s(\boldsymbol{k},\tilde{\omega}) = \int dt\,\mathrm{e}^{i\tilde{\omega}t}\,F_s(\boldsymbol{k},t), \quad (17)$$

then we put $F_s(\boldsymbol{k},t)$ in (16) and perform the integration over time. The result can be written in the following form:

$$\varphi\,(\boldsymbol{k},\omega) = \int \frac{d\tilde{\omega}}{2\pi}\, F_s(\boldsymbol{k},\tilde{\omega})\, G_0(\omega-\tilde{\omega})\,,$$

$$G_0(\omega) = \frac{i}{\omega + i\Gamma_0/2\hbar}\,. \tag{18}$$

Here $G_0(\omega)$ is the standard resonant scattering factor.

We conclude that the main result of accounting for diffusion is the averaging of the scattering amplitude over Doppler shifts of resonance frequency caused by diffusive motion of nuclei. The influence of this motion on the spectra is presented by the correlation function $F_s(\boldsymbol{k},t)$ [see (16)] and can also be described with the help of the Van Hove function $G_s(\boldsymbol{r},t)$ and the spectral function $F_s(\boldsymbol{k},\omega)$.

As mentioned above we restrict our analysis in this paper to the case of free diffusion only. More complicated cases will be considered later. In the present case the Van Hove function is the solution of the simple diffusion equation $\partial G_s/\partial t = D\,\nabla^2\,G_s$ with the diffusion coefficient D. The substitution of the Fourier expansion (12) in this equation allows us to find the correlation function $F_s(\boldsymbol{k},t)$ satisfying the boundary condition $F_s(\boldsymbol{k},0) = 1$

$$F_s(\boldsymbol{k},t) = \exp\left(-D\,k^2\,t\right)\,, \quad t > 0\,. \tag{19}$$

Substituting this expression in (16) we immediately arrive at the following result:

$$\varphi\,(\boldsymbol{k},\omega) = \frac{i}{\omega + i\Gamma_0/2\hbar + iDk^2}\,. \tag{20}$$

In Refs. 1 and 6 the real part of this expression is employed to describe the Mössbauer absorption spectra.

As for the spectral function $F_s(\boldsymbol{k},\omega)$, its determination via the Fourier transformation requires also the knowledge of the function $F_s(\boldsymbol{k},t)$ in the region $t < 0$. However, because $F_s(\boldsymbol{k},-t) = F_s^*(\boldsymbol{k},t)$ by definition (10), one should use $|t|$ instead of t. It leads to the following well-known formulas:

$$F_s(\boldsymbol{k},\omega) = \frac{2Dk^2}{\omega^2 + (Dk^2)^2}\,,$$

$$G_s(\boldsymbol{r},t) = \frac{1}{(4\pi D\,|t|)^{3/2}}\,\exp\left(-\frac{r^2}{4D\,|t|}\right)\,. \tag{21}$$

We note that only the time interval $t > 0$ has a physical meaning in the case of coherent scattering but it is not so in the case of incoherent scattering as will be shown in Sec. IV.

III. TIME DEPENDENCE OF THE FORWARD SCATTERING

The incident radiation can be decomposed into the plane monochromatic wave components. A typical component with the propagation vector $\boldsymbol{k}_0$ has the form

$$\boldsymbol{E}_i(\boldsymbol{r},t) = \boldsymbol{e}_s\,E^s_{\omega i}\,\exp\{i\,(\boldsymbol{k}_0\cdot\boldsymbol{r} - \omega t)\}\,. \tag{22}$$

Let this radiation fall on a slab of matter with the surface perpendicular to the Z axis and placed at $z = 0$. In (22) $\boldsymbol{e}_s$ is a unit vector of the wave polarization and $E^s_{\omega i}$ is the scalar wave amplitude. Let the vector $\boldsymbol{k}_0$ be oriented along $\boldsymbol{Z}$ where $\boldsymbol{Z}$ is a unit vector of the Z axis. The solution of the wave equation (1) with the current density (13) is given by the same exponent function as the plane monochromatic wave. However this function has the complex wave vector $\boldsymbol{k}_0' = \boldsymbol{k}_0 + (\varepsilon/2)\,\boldsymbol{Z}$, where ε is a small complex and frequency-dependent correction due to the refraction effect. Because of the very small magnitude of the interaction between the γ ray and the target we can account for the correction ε only in the linear approximation. Using the explicit form of the vector $\boldsymbol{k}_0'$ we easily obtain from (1) and (13) that $\varepsilon = K\,g^{ss}_{00}(\boldsymbol{k}_0,\omega)$. Assuming that the polarization state is not changed in scattering we shall omit below the polarization indexes. Thus, the solution of the wave equation in the case of forward scattering has the form of an incident plane wave but with a slowly variable scalar amplitude as the function of depth

$$E_\omega(z) = \exp\left\{i\,\frac{K}{2}\,g_{00}(\omega)\,z\right\}\,E_{\omega i}\,. \tag{23}$$

In the case of incident monochromatic wave with the spectral density $I_0(\omega)$ the transmitted beam has the intensity

$$I(\omega,z) = I_0(\omega)\,\exp\{-\mu(\omega)\,z\}, \quad \mu(\omega) = K\,g''_{00}(\omega)\,, \tag{24}$$

where g'' is an imaginary part of g. It can be verified directly that (24) contains only the real part of the function $\varphi\,(\omega)$ [see Eq. (14)].

Let us consider the transmission of synchrotron radiation through a resonant target. A short pulse of synchrotron radiation can be decomposed into the continuous set of monochromatic spectral components within the frequency interval near ω_0, well exceeding the width of the resonance range. One may assume that all spectral components have equal weights, i.e., $E_{\omega i} = E_{\omega_0 i}$. In order to find the wave packet of the transmitted radiation one should integrate all spectral components of the transmitted radiation (23)

$$E(z,t) = E_{\omega_0}(z)\int\frac{d\omega}{2\pi}\,\exp\left(-i\omega t\right)$$

$$\times\exp\left\{i\frac{K}{2}\,g^{(n)}_{00}(\omega)\,z\right\}\,. \tag{25}$$

where the function $E_{\omega_0}(z)$ has the modulus $(I_0/\Delta\omega)^{\frac{1}{2}}\,\exp\left(-\mu_e z/2\right)$, $\mu_e = K\,\chi''_{00}$, and I_0 is the intensity of SR within the frequency band $\Delta\omega$ as determined by a monochromator system. As follows from (25) *the complex function* $\varphi(\omega)$ *(via* $g^{(n)}_{00}$*) will enter in the expression for the intensity entirely.*

According to (23) the interaction with electrons influences all spectral components equally contributing only a constant factor to the wave packet (25). So in considering the frequency and time dependence of the transmitted intensity we deal with

$$g_{00}^{(n)}(\omega) = -\sum_{m} \frac{B_{00}(m)\,\Gamma_0/2\hbar}{\omega - \omega_m + i\Gamma_0/2\hbar + iD\,k_0^2}\,, \tag{26}$$

where summing over $'ge'$ in (14) is transformed into summing over the transitions between ground and excited states of the nucleus $'m'$, taking into account the selection rules (we consider only the case of free diffusion). If a hyperfine splitting of nuclear levels is absent we arrive at the expression for the transmitted wave packet similar to that obtained in Ref. 13

$$E(z,\tau) = E_{\omega_0}(z,\tau)\,\frac{1}{t_0}\left\{\delta(\tau) - \exp(-q\tau/2)\,\sqrt{\mu_n z/4\tau}\,J_1(\sqrt{\mu_n z\tau}\,)\right\}\,, \tag{27}$$

where $\tau = t/t_0$, $t_0 = \hbar/\Gamma_0$ is the lifetime of excited nuclear state, J_1 is the Bessel function, and $E_{\omega_0}(z,\tau) = E_{\omega_0}(z)\,\exp(-i\omega_0 t_0\tau)$,

$$\mu_n = K\sum_m B_{00}(m) = N\,\sigma_0\,\eta\,f_{\rm LM}\,,\quad q = 1 + \frac{2\hbar D\,k_0^2}{\Gamma_0}\,. \tag{28}$$

Here σ_0 is the nuclear cross section at the resonance. The enrichment by resonance isotope $\eta \leq 1$ is also taken into account. For the forward scattering intensity one has

$$\begin{aligned} I_{\rm FS}(z,\tau) = {}& \frac{I_0}{\Delta\omega t_0}\,\exp(-\mu_e z) \\ & \times \exp(-q\tau)\,\frac{\mu_n\,z}{4\,t}\,J_1^2(\sqrt{\mu_n\,z\,\tau}\,)\,, \end{aligned} \tag{29}$$

It follows from (29) that the broadening caused by diffusive motion results only in a faster decay of the exponential factor. It happens because dephasing the spectral components emerging from the target proceeds faster due to a broader spread of their frequencies. The latter yields the additional destroying the coherent signal. It is of interest to mention the faster decay is described exactly by the correlation function $F_s^2(\boldsymbol{k},t)$ [see (19)] as a multiplier. At the same time the effective dimensionless target thickness $\mu_n z$ stays the same; therefore both the initial intensity and the dynamical beats (expressed by the Bessel function J_1) are not changed. Just the same situation took place in the study of coherent scattering in crystals excited by vibrations when all nuclei were vibrated in unison.[14] It was found that the effective target thickness stayed just the same as in the case without vibrations.

IV. TIME DEPENDENCE OF THE 4π SCATTERING

A full picture of scattering includes both spatially coherent and incoherent channels. Along with the directed coherent beams of γ radiation the 4π shine of γ rays, electrons, and fluorescence x rays appears around a target due to incoherent scattering of the primary radiation. In this section we shall regard the influence of the diffusive atomic motion on this channel.

There are many paths of scattering into 4π. We consider here the channel where the fluorescence radiation is generated. Each atom being excited by the coherent field propagating through the target [see Eq. (27)] becomes a source of a secondary radiation. In particular, due to the internal conversion and photoeffect an electron is ejected from the atomic shell. The subsequent filling a hole is accompanied by the emission of fluorescence radiation.

In the process of nuclear resonant scattering of SR the two possibilities should be distinguished: the first stage of nuclear scattering, γ-ray absorption (excitation), can proceed both with and without recoil (see also Refs. 15 and 16). We begin our analysis with the recoilless channel which shall be denoted as the A channel.

The internal conversion happens at the second stage of the scattering process and it brings the standard exponential time dependence in the whole process with the stationary probability amplitude B_{eg}. More complicated time dependence of the scattering amplitude by a separate nucleus is determined by the amplitude of the nuclear excitation, so that the total amplitude has the form

$$A_{age}(t) = -\frac{i}{\hbar}\,B_{eg}\int_{-\infty}^{t} dt'\,\exp\{i(-\omega_{eg} + i\Gamma_0/2\hbar)\,(t-t')\}\langle e\,|\,V_a(t')\,|\,g\rangle. \tag{30}$$

The interaction potential for a nucleus with number $'a'$, $V_a(t)$, has just the same nature as that for the coherent channel [see Eq. (4)] but now considering the incoherent channel we shall assume the incident radiation in the form of plane wave with the variable amplitude (23),

$$V_a(t) = i\hat{j}^{s*}(k_0)\exp[i\boldsymbol{k}_0\cdot\boldsymbol{r}_a(t)]\int\frac{d\omega}{2\pi}\,\exp(-i\omega t)\,\frac{E_\omega^s(z)}{\omega}\,. \tag{31}$$

Substituting (31) in (30) and changing the order of integration one has for the time-dependent amplitude of the incoherent scattering

$$\begin{aligned} A_{age}(t,z) = {}& C_{ge}^{*}B_{eg}\int\frac{d\omega'}{2\pi}\,\frac{E_{\omega'}(z)}{\hbar\omega'}\,\exp(-i\omega' t) \\ & \times \int_{-\infty}^{t} dt'\,e\,(\omega' - \omega_{eg}, t - t') \\ & \times \exp[i\boldsymbol{k}_0\cdot\boldsymbol{r}_a(t')]\,, \end{aligned} \tag{32}$$

where C_{ge}^{*} and $e\,(\omega,t)$ are defined by (7). The phase factor $\exp[i\boldsymbol{k}_0\cdot\boldsymbol{r}_a(t)]$ accounting for the motion of scattering nucleus (the diffusional one included) explicitly enters the scattering amplitude. When the nuclei are at rest, $\boldsymbol{r}_a(t)$ =const, one can calculate the integral over t' directly. As a result one obtains

$$A_{age}(t,z) = \frac{C^*_{ge}B_{eg}}{\hbar\omega} \exp{(i\boldsymbol{k}_0 \cdot \boldsymbol{r}_a)} \times \int \frac{d\omega'}{2\pi} \exp(-i\omega' t) \frac{E_{\omega'}(z)}{\omega' - \omega_{eg} + i\Gamma_0/2\hbar} . \tag{33}$$

Here we took into account that near the resonance $\omega' \approx \omega_{eg} \approx \omega$ and the factor $1/\omega$ does not influence the result of integration, therefore it can be taken out of the integral. This formula has a simple physical meaning: the amplitude of the probability of nuclear excitation is proportional to the sum over all frequency components of the field with the weight that is a resonance factor for a given nuclear transition.

The probability of the incoherent scattering of the transmitted radiation at the depth z as a function of time can be found as the square modulus of the amplitude which is summed over all nuclei in the unit volume and over all excited states and averaged over ground states

$$P(t,z) = (2I_0+1)^{-1} \sum_{age} |\, A_{age}(t,z) \,|^2 . \tag{34}$$

When the nuclei take part in a diffusive motion the problem becomes more complicated because we have to account for the correlation between the positions of nuclei at different times. Substituting (32) in (34) we obtain

$$P = \frac{f_{\mathrm{LM}}(\boldsymbol{k}_0)}{(\hbar\omega)^2 V_0 (2I_0+1)} \sum_{ge} |\, C_{ge}B_{eg} \,|^2 \int \frac{d\omega'}{2\pi} E_{\omega'} \exp(-i\omega' t) \int \frac{d\omega''}{2\pi} E^*_{\omega''} \exp(i\omega'' t) \times \int_{-\infty}^{t} dt' \int_{-\infty}^{t} dt''\, e\,(\omega' - \omega_{eg}, t - t')\, e^*(\omega'' - \omega_{eg}, t - t'')\, F(\boldsymbol{k}_0, t'', t') , \tag{35}$$

where the correlation function $F(\boldsymbol{k}_0, t'', t')$ is determined by (10). We have to notice that in incoherent scattering the correlations arise only in the intensity but not the amplitude.

As before we can replace F by $F_s(\boldsymbol{k}_0, t'' - t')$. Introducing the Fourier expansion of the correlation function (17) we can perform the integration over time. Finally we pass from the probability P to the intensity of secondary radiation $I^A_{4\pi}$ for a depth interval from zero to d. Here the index A refers to the nuclear recoilless interaction. As a result we obtain the following expression:

$$I^A_{4\pi}(t) = \frac{f_{\mathrm{LM}}}{(\hbar\omega)^2 V_0 (2I_0+1)} \sum_{ge} |\, C_{ge}B_{eg} \,|^2 \int_0^d dz \int \frac{d\tilde{\omega}}{2\pi} F_s(\boldsymbol{k}_0, \tilde{\omega})\, |\, D_{eg}(t, z, \tilde{\omega}) \,|^2 , \tag{36}$$

where

$$D_{eg}(t, z, \tilde{\omega}) = \int \frac{d\omega}{2\pi} \exp{(-i\omega t)} \frac{E_\omega(z)}{\omega - \tilde{\omega} - \omega_{eg} + i\Gamma_0/2\hbar} . \tag{37}$$

We note that Eq. (37) represents the time response of the separate nucleus having resonance frequency $\omega_{eg} + \tilde{\omega}$ shifted due to diffusive motion [compare with (33)]. The integral over $\tilde{\omega}$ in (36) leads to averaging the relevant time dependencies.

Let us consider once again the simple case when hyperfine splitting is absent. Then Eqs. (36) and (37) can be written in the explicit form

$$I^A_{4\pi}(\tau) = \frac{I_0}{\Delta\omega t_0} \frac{\Gamma_e}{\hbar} N \sigma_0 \eta f_{\mathrm{LM}} \int_0^d dz \exp{(-\mu_e z)} \int \frac{d\tilde{w}}{\pi} \frac{W}{\tilde{w}^2 + W^2} \left| \int \frac{dw}{2\pi} \frac{\exp{(-iw\tau)}}{(w - \tilde{w} + i/2)} \exp\left(-i \frac{\mu_n z}{2\,(2w + iq)}\right) \right|^2 . \tag{38}$$

Here and below we use dimensionless variable $w = \omega t_0$ and $W = (q-1)/2$, Γ_e is the nonradiative (internal conversion) part of the width of nuclear level $\Gamma_e = \Gamma_0 \alpha/(1+\alpha)$ where α is the conversion coefficient.

When $D = 0$ $(q = 1)$, the diffusive motion of nuclei is absent and one can obtain from (38) a more simple analytical expression for the intensity of recoilless incoherent scattering in the form

$$I^A_{4\pi}(\tau) = \frac{I_0}{\Delta\omega t_0} \frac{\Gamma_e}{\hbar} N \sigma_0 \eta f_{\mathrm{LM}} \exp(-\tau) \times \int_0^d dz \exp{(-\mu_e z)}\, J_0^2(\sqrt{\mu_n z \tau}\,), \tag{39}$$

where J_0 is the Bessel function of order zero. Please note that the time dependence of recoilless incoherent scattering at a fixed depth in the target [Eq. (39) without the integration] is much different from the time dependence of the coherent field intensity at the same depth [see Eq. (29)]. But in Ref. 15, simply the integration of the coherent field intensity over the sample thickness was performed to get the recoilless incoherent scattering intensity. In this way the resonance nature of the nuclear incoherent scattering was not taken into account.

Now we turn to the nuclear resonant scattering of the transmitted radiation by a nucleus with recoil. This process is described by the same formulas (36) and (37). However now we must replace the transition energy ω_{eg} by $\omega_{eg} + \omega_{\rm ph}$ where $\omega_{\rm ph}$ is a phonon frequency and integrate over the phonon spectrum. We suppose that the spectrum of SR is wide enough to include these processes. In principle, the two frequency intervals must be distinguished while integrating (38) in order to get the time-dependent result. In the frequency range where $\omega \approx \omega_{eg} + \omega_{\rm ph}$ a nucleus is resonantly excited. In this frequency range we can neglect the frequency dependence of the amplitude $E_\omega(z)$ because $\omega_{\rm ph}$ is much larger than Γ_0 and the characteristic scale of the diffusive broadening. In the frequency range $\omega \approx \omega_{eg} + \tilde{\omega}$, on the contrary $E_\omega(z)$ is strongly dependent on frequency, while the denominator in (37) is practically constant and equals $-\omega_{\rm ph}$ with good accuracy. As a result we have the following two contributions into the nuclear scattering intensity with recoil:

$$I^B_{4\pi}(\tau) = \frac{I_0}{\Delta\omega t_0}\,\frac{\Gamma_e}{\hbar}\,N\,\sigma_0\,\eta\,(1 - f_{\rm LM})\,\exp\,(-\tau) \times \int_0^d dz\,\exp\,(-\mu_e z)\,,$$

$$I^D_{4\pi}(\tau) = N\,\sigma_0\,\frac{\Gamma_e}{\Gamma_0}\int \frac{d\omega_{\rm ph}}{(\omega_{\rm ph} t_0)^2}\,\xi\,(\omega_{\rm ph})\int_0^d dz\,I_{\rm FS}\,(z,\tau), \tag{40}$$

where $\xi(\omega_{\rm ph})$ is the normalized weight function of a phonon spectrum. The first term in (40) is characterized by the natural decay time, while the second term has just the same time dependence as the forward scattering intensity. We note that the second term is of order $(\Gamma_0/\hbar\omega_D)^2$ smaller than the first one where ω_D is Debye frequency, and we shall neglect it in calculations.

The last contribution to the 4π scattering (C channel) is related to the scattering of the coherent field by the electronic shell.[15,16] In considering it we shall assume that the frequency region of the incident radiation is far away from the resonance frequency for the electronic photoeffect. So in this case the time dependence is determined by the intensity of forward scattering

$$I^C_{4\pi}(\tau) = N\,\sigma_{\rm pho}\int_0^d dz\,I_{\rm FS}\,(z,\tau), \tag{41}$$

where $\sigma_{\rm pho}$ is a cross section of electronic photoabsorption corresponding to the resonance frequency of the Mössbauer effect.

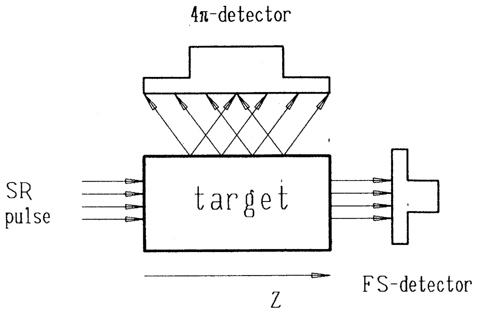

FIG. 1. Possible experimental arrangement.

V. ANALYSIS OF THE FREE DIFFUSION CASE

We shall illustrate the theory developed in the previous sections by calculations of the time dependencies of the forward and 4π scattering for a simple model system. We assume that the very small particles containing resonant nuclei ^{57}Fe perform a diffusive motion in a viscous medium. The arrangement of the possible experiment is shown in Fig. 1. Let the motion be unconfined, quasicontinuous, and isotropic. We shall consider the values of the diffusion coefficient D in the range $10^{-15} - 10^{-14}$ m^2/s causing the nuclear γ resonance broadening up to 10 natural linewidths. To single out the effect of broadening we assume that the Lamb-Mössbauer factor stays constant while the diffusion coefficient is changing.

The forward scattering intensity as well as the B and C channels of 4π scattering were calculated directly by means of Eqs. (29) and (38) – (41). For calculation of the A channel the integral over frequency in (38) was transformed into the integral over time, so that

$$I^A_{4\pi}(\tau) = \frac{I_0}{\Delta\omega t_0}\,\frac{\Gamma_e}{\hbar}\,N\,\sigma_0\,\eta\,f_{\rm LM}\,\exp\,(-\tau)\int_0^d dz\,\exp\,(-\mu_e z)\int \frac{d\tilde{\omega}}{\pi}\,\frac{W}{\tilde{\omega}^2 + W^2} \times \left| 1 - \int_0^\tau d\tau'\,\exp\,(-W\tau' + i\tilde{\omega}\tau')\sqrt{\frac{\mu_n z}{4\tau'}}\,J_1\,(\sqrt{\mu_n z\tau'}\,)\right|^2 . \tag{42}$$

The results of direct numerical calculations are presented in Figs. 2–4. In every figure the time dependencies are shown illustrating the evolution of SR scattering into the spatially coherent (Fig. 2) and incoherent (Figs. 3 and 4) channels for the three magnitudes of the parameter of broadening q = 1, 4, and 10 corresponding to the val-

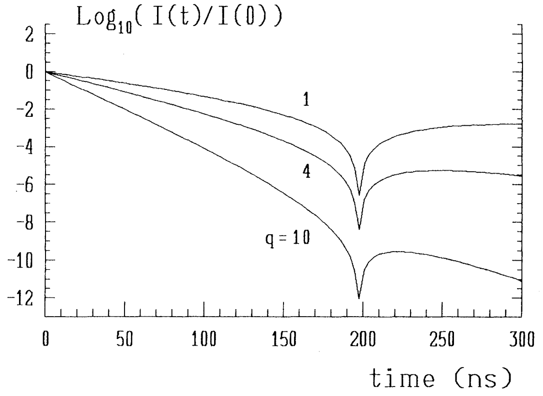

FIG. 2. Forward scattering intensity, $\mu_n d = 10.5$.

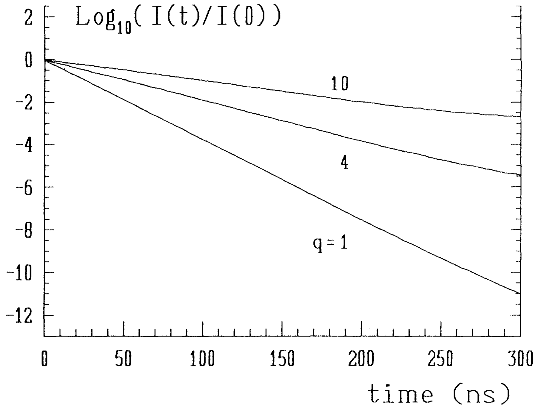

FIG. 4. Incoherent scattering intensity, $\mu_n d = 10.5$, Process C (electronic scattering).

ues of the diffusion coefficient (0, 2, 6) $10^{-15}\,\mathrm{m}^2/\mathrm{s}$. It is seen in Fig. 2 that the increase of the diffusion coefficient causes the progressive acceleration of the coherent signal decay. At the initial stage the decay follows closely to the exponential law $\exp\left[-\tau\,(1+2\hbar Dk^2/\Gamma_0+\mu_n d/4)\right]$. There are two different physical reasons for the faster decay of the coherent signal. One reason is the speed-up of the nuclear deexcitation which is related to the effect of enhancement of the radiative channel.[17,18] The relevant speed-up is given by the exponential index term $\mu_n d/4$ proportional to the target thickness.

Another reason for the dropping down of the coherent signal is diffusional broadening of the nuclear resonance which leads to more rapid dephasing of the spectral components of the forward scattered radiation. The relevant acceleration is determined by the exponential index term $2\hbar Dk^2/\Gamma_0$.

We note that the diffusion coefficient variation does not influence the initial forward scattered intensity and produces no effect on the position of the dynamical beating minimum. This is because the effective dimensionless thickness of the target $\mu_n d$ stays the same irrespective of the diffusion state. The time-integrated intensity of the forward scattering is a decreasing function of the diffusion coefficient.

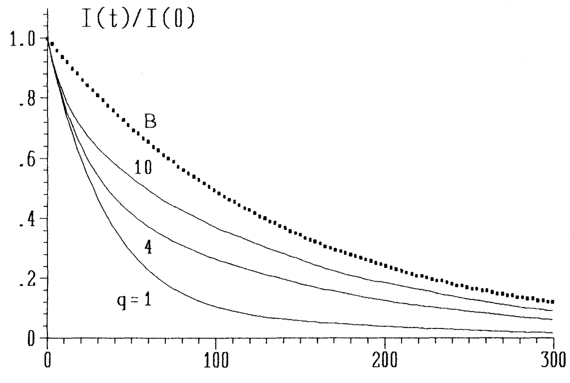

FIG. 3. Incoherent scattering intensity, $\mu_n d = 10.5$, solid lines: Process A (nuclear recoilless scattering) for q = 1, 4, 10, dotted line: Process B (nuclear scattering with recoil) for q = ∞.

Regarding the A contribution to the 4π scattering (Fig. 3) one can notice the two clearly distinguished stages of the decay evolution: the initial fast and the later slow ones. One can also notice that the duration of the initial stage is close to the characteristic duration of the coherent signal (Fig. 2). From this observation and from the corresponding analysis of Eqs. (29) and (42) it is possible to deduce that at the initial stage the decay of separate nuclei excited by the prompt part of the transmitted wave packet [δ function in Eq. (27)] proceeds under strong influence of the delayed part of the coherent radiation (which plays the role of external radiation for the downstream nuclei). At a later stage when the coherent signal is vanishing the nuclei are deexciting spontaneously in accordance with the natural decay law $\exp(-\tau)$. The more intense the diffusive motion the shorter the initial stage. In the limit of very large diffusion coefficient D, where the broadening reaches several hundreds, the initial stage is negligible and nuclei radiate into 4π spontaneously for almost all times.

This conclusion can be also derived from the approximate analytical expression for the integrals of Eq. (38). Indeed, expanding the resonant exponent in a power series and calculating each term by means of the residue theorem one can obtain the integrand over z in the form of an expansion over the parameters $h = \mu_n z/4W$ and $g = W\tau$ as follows:

$$I^A_{4\pi}(\tau) = C\int_0^d dz\,\exp(-\mu_e z)\left[\exp(-\tau)\,F(z,W)\right.$$
$$\left.+\exp(-q\tau)\,G(z,W,\tau)\right], \tag{43}$$

where

$$\begin{aligned} C &= \frac{I_0}{\Delta\omega t_0}\,\frac{\Gamma_e}{\hbar}\,N\,\sigma_0\,\eta\,f_{\mathrm{LM}}, \\ F\,(z,W) &= 1 - h + \frac{3}{4}\,h^2 - \frac{5}{12}\,h^3 + \cdots, \\ G\,(\tau,z,W) &= h - \frac{3}{4}\,h^2(1+2g) \\ &\quad + \frac{5}{12}\,h^3[1 + 2g\,(1+g)\,] - \cdots. \qquad (44) \end{aligned}$$

This expression is valid for all times when $\mu_n d/4W < 1$ and only in the time interval $\tau < 4/\mu_n d$ in the opposite case. In particular, it coincides well with the direct numerical results for $q = 10$ and approximately for $q = 4$. As for the case with $D = 0$ it is described by the formula (39).

It is obvious that the nuclei excited with recoil (the B contribution) are not affected by the coherent (recoilless) radiation and therefore they are decaying spontaneously (dashed line in Fig. 3) for all times. The time dependence and the time-integrated intensity of the B contribution are not dependent on diffusion. The time dependence of the A contribution in the limit of large D is the same as of the B contribution, i.e., $\exp(-\tau)$. The time-integrated intensity of the A contribution approaches a definite limit. The ratio between the time-integrated intensities of the A and B contributions is determined by the Lamb-Mössbauer factor.

The time dependence of the C contribution (Fig. 4 and the dotted line in Fig. 3) is a weakened image of the coherent radiation intensity integrated over the target thickness. While the diffusive mobility is rising, the integrated intensity of this contribution is falling down.

VI. CONCLUSION

The wave equation for the transmission of a plane monochromatic wave (as a component of an arbitrary wave packet of γ radiation) through a medium containing resonant nuclei is formulated for the case of diffusive motion of nuclear particles [Eqs. (1) and (13)]. Under these conditions of motion the nuclear resonant susceptibility of the medium is not dependent on time [Eq. (14)]. However it contains the nuclear temporal response averaged over the time correlation function $F_s(\boldsymbol{k},t)$ [Eq. 16]. On the other hand the effect of diffusive motion can be described with the aid of the spectral function $F_s(\boldsymbol{k},\omega)$ [the Fourier image of $F_s(\boldsymbol{k},t)$], which presents the Doppler shift distribution of the nuclear resonance frequency caused by the diffusive motion of the nuclei. Therefore we think that it is possible to extend the results of the theory to any case where the resonance shape differs from a Lorentzian and where the width is increased, for example, due to inhomogeneous distribution of chemical shifts or hyperfine fields in the sample.

The formulas for the time dependences of the coherent forward and incoherent 4π scattering are obtained for the case where the broadened resonance preserves the Lorentzian shape (in particular, this happens in the case of free diffusion). As for the coherent channel, similar formulas were obtained earlier in Ref. 19. The resonance broadening causes an additional acceleration of the nuclear decay in the coherent channel and a decrease of the time-integrated intensity in this channel, with the initial intensity and the dynamical beat pattern being unchanged. The 4π channel is representative of the spatially incoherent scattering of the radiation. The intensities from individual atoms are summed up in this channel. Being illuminated by the coherent field transmitted through the target [Eq. (27)], the resonant nucleus can be excited both without and with recoil by the prompt part of the coherent wave packet [the first term in (27)]. The nuclei excited with recoil are not coupled with the coherent field any longer and therefore decay spontaneously with the natural decay rate. The intensity in this channel is proportional to $(1 - f_{\mathrm{LM}})$ the smaller the recoilless factor, the more the natural decay is presentive in the incoherent channel. Obviously this channel is not sensitive to the resonance broadening.

The resonant scattering without recoil has a more complicated nature. Here the influence of the delayed part of the coherent wave packet [second term in (27)] is of importance. The response of the nuclei via this channel has a characteristic two-stage time dependence where at the first stage the coherent field causes an accelerated nuclear response. The first stage becomes less important with the increase of the resonance width. In the limit of a very wide resonance the time dependence in this particular channel approaches the natural decay law.

Scattering of the coherent field by the electronic shell reproduces the time dependence of the forward scattering integrated over the target thickness. Its contribution to the total incoherent intensity decreases with resonance broadening.

Thus in the full picture of scattering the balance between the forward and the 4π scattering becomes more and more in favor of the 4π channel when the resonance becomes blurred. One should note that the role of the 4π channel also increases when the recoilless factor is decreasing. The incoherent scattering of synchrotron radiation delivers a good possibility to study variations of these parameters.

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