

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

V. G. Kohn and G. V. Smirnov

Russian Research Centre "Kurchatov Institute," 123182, Moscow, Russia

(Received 12 March 1997)

A general theory of the time dependence of nuclear resonant forward scattering of synchrotron radiation in the presence of diffusive motion of nuclei is further developed. The scattering problem is solved for the two characteristic cases of diffusive motion. The first one is the continuous isotropic localized diffusion of a particle within a cage formed by a drift potential. The second case is the jump anisotropic unlimited diffusion of nuclei on a crystalline lattice. In both cases the frequency dependence of nuclear susceptibility has a complicated shape described by a superposition of Lorentzian functions having different weights and widths. Correspondingly several stages appear in the time evolution of the nuclear forward scattering which are characterized by different decay rates. In the thick absorber case the target can exhibit successively different partial thicknesses in the time evolution of forward scattering. [S0163-1829(98)03709-6]

I. INTRODUCTION

It is well known that Mössbauer spectroscopy provides a unique possibility to explore the dynamics of very slow atomic motions owing to its extremely high-energy resolution. First of all, it is applicable to a diffusive motion of atomic particles which occurs in liquids and solids (for a review see Refs. 1–4). The analysis of Mössbauer spectra affected by diffusion was done by Singwi and Sjölander⁵ in terms of the Van Hove correlation function $G(\mathbf{r}, t)$. This function describes a probability for finding the nucleus in a matter at a position \mathbf{r} at time t , if it was at the origin at time $t=0$. The phase shift of γ rays scattered by the nucleus is related to the shift of the mean position $\mathbf{r}(t)$ of the nucleus. It depends on the time interval between the moments of excitation and deexcitation. The explicit form of the Van Hove function is different in each particular case of diffusive motion. In any case a diffusion manifests itself in the profile of the Mössbauer absorption spectra through the broadening and the change in shape of resonance lines.

Recently a powerful technique for studying nuclear γ resonance was developed with the use of nuclear resonant scattering of synchrotron radiation (SR). This technique is based on measuring the time dependence of intensity reemitted by nuclei after an excitation of the nuclear system by very short pulse of synchrotron radiation. The observed coherent reemission into the forward direction allows us to claim a formation of excitation distributed coherently over the entire nuclear system, called a nuclear exciton, which has an unusual (nonexponential) character of decay. The coherent decay of the nuclear exciton is characterized by a speed-up effect accompanied by quantum and dynamical beats of intensity.^{6,7} Many nuclear and solid-state parameters can be explored by studying the time dependences of the nuclear exciton decay.⁸

A general theory of time-dependent nuclear resonant forward scattering of SR pulse by a system of nuclei moving diffusively has been developed earlier.⁹ It was shown that similarly to the approach developed in Ref. 10, the time de-

pendence of radiation field can be calculated through a representation of SR pulse as a homogeneous coherent superposition of monochromatic waves and a subsequent evaluation of the response of the nuclear ensemble to each monochromatic wave as a scattering amplitude. The latter depends on the diffusive motion of nuclei represented by the double dimensional (momentum and frequency) Fourier image of the Van Hove function. In contrast to the absorption spectra where the diffusion influences only an absorption coefficient, the forward scattering time dependence is influenced by diffusion in a more complicated way. In Ref. 9 only a case of free continuous diffusion was analyzed. Here we develop the theory further to include the continuous localized diffusion and the jump unlimited diffusion. In the next section both the main ideas and results of the general theory⁹ are shortly summarized.

II. GENERAL FORMULAS

A short pulse of synchrotron radiation can be decomposed into a continuous set of coherent monochromatic waves within the frequency interval $\Delta\omega$ centered at the resonance frequency and well exceeding the width of resonance range. With a good accuracy one can consider all monochromatic components to be equivalent in weight. To calculate the forward transmitted wave packet one should integrate all forward transmitted monochromatic components. As it was shown⁹ the result can be written as follows:

$$E(t, z) = E_0(z) \int \frac{d\omega}{2\pi} \exp(-i\omega t) \exp\left(i \frac{K}{2} g^{(n)}(\omega) z\right). \quad (1)$$

Here $E(t, z)$ is a time-dependent electric-field amplitude of synchrotron radiation transmitted through a nuclear target of thickness z . The function $E_0(z)$ has the modulus $(I_0/\Delta\omega)^{1/2} \exp(-\mu_e z/2)$ with $\mu_e = K\chi''$ being the electron absorption coefficient and I_0 being the intensity of SR within the frequency band $\Delta\omega$ as determined by a monochromator system. The wave number $K = 2\pi/\lambda = \omega/c$.

The nuclear part of the susceptibility of the target $g^{(n)}$ is closely related to the scattering amplitude and can be represented as

$$g^{(n)}(\mathbf{k}, \omega) = i \frac{\Gamma_0}{2\hbar} \sum_{ge} B_{ge} \varphi(\mathbf{k}, \omega - \omega_{eg}), \quad (2)$$

where the value

$$B_{ge} = \frac{8\pi f_{\text{LM}}(\mathbf{k})}{\omega^2 V_0 (2I_g + 1) \Gamma_0} |\langle g | \hat{j}(\mathbf{k}) | e \rangle|^2 \quad (3)$$

characterizes the strength of nuclear response at the resonance frequency ω_{eg} . Here the indexes g and e numerate the hyperfine sublevels of the ground and excited states of a nucleus, Γ_0 is the natural width of the excited level, $f_{\text{LM}}(\mathbf{k})$ is the Lamb-Mössbauer factor, I_g is the nuclear spin in the ground state, V_0 is the target volume corresponding to one nucleus, and $\langle g | \hat{j}(\mathbf{k}) | e \rangle$ is the matrix element of the scalar component of the nuclear current density operator along the polarization vector of the incident wave. In particular, the latter parameter equals zero if the transition is forbidden for a given multipolarity of radiation.

The frequency dependence of susceptibility near the resonance is determined entirely by the universal resonance function $\varphi(\mathbf{k}, \omega)$ which takes into account the diffusive motion. It is described by the formula

$$\varphi(\mathbf{k}, \omega) = \int_{-\infty}^t dt' \exp\{i(\omega + i\Gamma_0/2\hbar)(t-t')\} F_s(\mathbf{k}, t-t'), \quad (4)$$

where the correlation function in momentum representation is introduced. Such a representation naturally corresponds to a scattering problem. We term the function $F_s(\mathbf{k}, t)$ as a momentum-time (MT) correlation function. It is tightly related to the space-time (Van Hove) correlation function, $G(\mathbf{r}, t)$, via the Fourier transformation

$$F_s(\mathbf{k}, t) = \int d\mathbf{r} \exp(-i\mathbf{k}\mathbf{r}) G(\mathbf{r}, t). \quad (5)$$

For the convenience of the following calculation, we shall use below a more direct representation for the function $\varphi(\mathbf{k}, \omega)$ which is obtained by changing the variable, namely,

$$\varphi(\mathbf{k}, \omega) = \int_0^{\infty} dt \exp(i\omega t - \Gamma_0 t/2\hbar) F_s(\mathbf{k}, t) \quad (6)$$

A derivation of formulas (1)–(6) is given in the first part of this work.⁹ Here we emphasize once again that we restrict ourselves by the cases when polarization mixing is absent in the coherent forward scattering. For example, this takes place for the $M1$ transition of ^{57}Fe nuclei. On the other hand, we consider the cases where the fast thermal motion of nuclei near a temporal equilibrium site and the slow motion of the equilibrium site itself are factorized and enter in different

ways into the scattering amplitude. The first one enters through the well-known Lamb-Mössbauer factor f_{LM} , while the second one is described by the universal resonance function in a more complicated form. Mathematically this procedure can be done in all cases by simply introducing the Lamb-Mössbauer factor in the explicit form and considering the total dephasing function without the Lamb-Mössbauer factor. We note that the Lamb-Mössbauer factor has different behavior in solids and liquids.

The most frequent application of SR in the nuclear resonance spectroscopy is a measurement of the time dependence of the forward scattering intensity.

$$I_{fs}(t, z) = |E(t, z)|^2. \quad (7)$$

It is of interest to compare the time dependence with the frequency dependence of intensity, which is ordinarily investigated in the Mössbauer absorption spectroscopy. Without a source convolution it is as follows:

$$I_a(\omega, z) = |E(\omega, z)|^2 = |E_0(z)|^2 \exp(-Kz \text{Im}g^{(n)}(\omega)). \quad (8)$$

One can see that the shape of the absorption spectra is directly determined by the real part of the universal resonance function $\varphi(\mathbf{k}, \omega)$. As for the time dependence of the radiation field, it is calculated through the frequency representation, Eqs. (1)–(3), where the entire universal function should be used.

On the other hand, this frequency-dependent function is determined by the time dependence of the two processes shown in Eq. (4), namely, by the decay of individual excited nucleus and by the diffusive motion (represented by the MT correlation function).

The MT correlation function (5) represents the mean value of the phase factor over a whole volume of the target. Being a function of time this averaged phase factor reflects a diffusive motion of nuclei. By its nature the correlation function is very similar to the Lamb-Mössbauer factor which describes a damping of the scattering amplitude owing to the thermal motion. The difference between them is only in the time scales which are characteristic of the motions involved.

The evident property of the Van Hove function $G(\mathbf{r}, 0) = \delta(\mathbf{r})$ gives the property of the MT correlation function $F_s(\mathbf{k}, 0) = 1$. Without a diffusive motion of nuclei in the target, this initial value stays constant. Correspondingly the resonance function takes the ordinary form $\varphi(\mathbf{k}, \omega) = i/(\omega + i\Gamma_0/2)$. However, in a presence of diffusive motion the correlation function can drop down at a time comparable with the lifetime of the nucleus. As a consequence, the coherence time in the forward scattered wave packet becomes shorter, while the spectral width of radiation becomes broader.

It is useful to consider the case of small thickness z of the target where one can expand the exponential function in Eq. (1) in a power series. Then considering only the first term of the expansion, one easily arrives at the following expression for the scattering intensity:

$$I(t,z) = \frac{I_0}{\Delta\omega} \left(\frac{Kz\Gamma_0}{4\hbar} \right)^2 \exp(-\mu_e z - \Gamma_0 t) |F_s(\mathbf{k}, t)|^2 \times \left| \sum_{ge} B_{ge} \exp(-i\omega_{eg} t) \right|^2, \quad t > 0. \quad (9)$$

The time-dependent square modulus of the MT correlation function enters here as a multiplier, i.e., directly influences a decay of the coherent signal. The approximation (9) is usually called a kinematical approximation in which only a scattering of one photon by one nucleus is taken into account.

As it was shown in Eq. (10) the universal resonance function $\varphi(\mathbf{k}, \omega)$ can be found by averaging the resonance factor

$$\varphi(\mathbf{k}, \omega) = \int \frac{d\tilde{\omega}}{2\pi} F_s(\mathbf{k}, \tilde{\omega}) \frac{i}{\omega - \tilde{\omega} + i\Gamma_0/2\hbar}, \quad (10)$$

where $F_s(\mathbf{k}, \omega)$ represents the spectral density of the momentum-time correlation function $F_s(\mathbf{k}, t)$. This expression can be interpreted as an average value of standard scattering amplitude over Doppler shifts of the resonance frequency caused by diffusive motion of nuclei.

III. LOCALIZED DIFFUSION IN GENERAL CASE

In the first part of our work⁹ the case of free diffusion has been analyzed as an example of an application of the general theory. The free diffusion is understood as the unlimited in space, continuous motion of particles in a medium that is described by the diffusion coefficient only. The developed theory can be easily extended to more complicated regimes of diffusive motion. Indeed, the main problem here is to find an explicit form of the Van Hove correlation function. We note that the same problem should be solved in the Mössbauer absorption spectroscopy. Following the first consideration given by Singwi and Sjölander⁵ one can distinguish two limiting cases: a continuous motion of large particles in a medium and a jump motion of nuclei between the sites on the crystal lattice. The first motion can be limited in space under the influence of a drift potential. The second motion generally is unlimited in space, however there are cases where it also can be limited.¹¹⁻¹⁴

Here we consider the localized diffusion which represents the case of spacially restricted diffusive motion of a particle under the influence of a drift potential $U(\mathbf{r})$. It is known (see, for example, Refs. 15,16) that in this case the Van Hove correlation function is a solution of the Fokker-Planck equation

$$\frac{\partial G(\mathbf{r}, t)}{\partial t} = -\hat{L}G(\mathbf{r}, t), \quad \hat{L} = -D\nabla^2 - B(\nabla U(\mathbf{r}))\nabla \quad (11)$$

where \hat{L} is the Fokker-Planck operator describing the Brownian motion in the drift potential $U(\mathbf{r})$ with the diffusion coefficient D and the drift coefficient $B = D/T$. Here T is the absolute temperature in energy units. This equation has a formal solution $G(\mathbf{r}, t) = \exp(-\hat{L}t)G(\mathbf{r}, 0)$ where $G(\mathbf{r}, 0)$ can be taken in the form $G(\mathbf{r}, 0) = \exp(i\mathbf{k}\mathbf{r})w(\mathbf{r})$, where $w(\mathbf{r})$ is the standard Boltzmann distribution function

$$w(\mathbf{r}) = Z^{-1} \exp(-U(\mathbf{r})/T), \quad Z = \int d\mathbf{r} \exp(-U(\mathbf{r})/T). \quad (12)$$

The substitution of this formal solution in Eq. (5) and then in Eq. (4) yields

$$\varphi(\mathbf{k}, \omega) = i \int d\mathbf{r} \exp(-i\mathbf{k}\mathbf{r}) [\omega + i\Gamma_0/2\hbar + i\hat{L}]^{-1} \times \exp(i\mathbf{k}\mathbf{r}) w(\mathbf{r}). \quad (13)$$

It was shown¹⁶ that it is convenient to consider a Hermitian form \hat{H} of the Fokker-Planck operator \hat{L} which is defined by the relation

$$\hat{H} = \exp(U/2T) \hat{L} \exp(-U/2T) = D[-\nabla^2 + V(\mathbf{r})], \quad (14)$$

where

$$V(\mathbf{r}) = (2T)^{-2} (\nabla U(\mathbf{r}))^2 - (2T)^{-1} \nabla^2 U(\mathbf{r}). \quad (15)$$

One can easily see that $\hat{L}w(\mathbf{r}) = \psi_0 \hat{H} \psi_0$ if

$$\psi_0 = Z^{-1/2} \exp(-U(\mathbf{r})/2T), \quad (16)$$

and the resonance function $\varphi(\mathbf{k}, \omega)$ can be found as a quantum-mechanical average of the Hermitian operator representing the resonance interaction

$$\varphi(\mathbf{k}, \omega) = i \langle 0 | \exp(-i\mathbf{k}\mathbf{r}) [\omega + i\Gamma_0/2\hbar + i\hat{H}]^{-1} \times \exp(i\mathbf{k}\mathbf{r}) | 0 \rangle. \quad (17)$$

By employing the total set of eigenfunctions of the Hermitian operator \hat{H} , Eq. (17) can be transformed with the help of the relation

$$\langle 0 | AHB | 0 \rangle = \sum_n \langle 0 | A | n \rangle \langle n | H | n \rangle \langle n | B | 0 \rangle, \quad (18)$$

to the equation

$$\varphi(\mathbf{k}, \omega) = i \sum_n \frac{\langle \psi_0 | \exp(-i\mathbf{k}\mathbf{r}) | \psi_n \rangle \langle \psi_n | \exp(i\mathbf{k}\mathbf{r}) | \psi_0 \rangle}{\omega + i\Gamma_0/2\hbar + iD\varepsilon_n}, \quad (19)$$

where $D\varepsilon_n$ is the eigenvalue of the operator \hat{H} corresponding to the eigenfunction ψ_n . The sum over n denotes the summation over all eigensolutions. When the operator \hat{H} has both the discrete and the continuous parts of spectrum then this sum must be added by the integral.

The last formula allows us to reveal some general properties of localized diffusion. We rewrite it in a more compact form

$$\varphi(\mathbf{k}, \omega) = i \sum_n \frac{A_n(\mathbf{k})}{\omega + i\Gamma_0/2\hbar + iD\varepsilon_n}, \quad (20)$$

with

$$A_n(\mathbf{k}) = \left| \int d\mathbf{r} \exp(-i\mathbf{k}\mathbf{r}) \psi_0^*(\mathbf{r}) \psi_n(\mathbf{r}) \right|^2, \quad \sum_n A_n(\mathbf{k}) = 1. \quad (21)$$

It is easy to verify that $\hat{H}\psi_0=0$. This means that the eigenvalue of the ground state is $\varepsilon_0=0$ independently on the explicit form of the drift potential. The ground state represents the partial state of a nucleus where it does not move diffusively and preserves its average position unchanged during all time. The corresponding resonance has the natural width as for a static nucleus in isolation. The weight of this state is defined by the coefficient A_0 . This coefficient has a property which is essential for the retrieval of the drift potential. According to Eqs. (12), (16), and (21) one can write

$$A_0(\mathbf{k}) = \frac{|f(\mathbf{k})|^2}{|f(0)|^2}, \quad f(\mathbf{k}) = \int d\mathbf{r} \exp(-i\mathbf{k}\mathbf{r} - U(\mathbf{r})/T). \quad (22)$$

Thus the weight of the ground state $A_0(\mathbf{k})$ is directly associated with the drift potential profile $U(\mathbf{r})$ through the Fourier transformation. In the case of nuclear resonance scattering the modulus of \mathbf{k} is fixed, therefore one cannot probe the potential at different magnitudes of \mathbf{k} . However, the measurements at different directions of \mathbf{k} can give information about the symmetry of the potential. In addition, the temperature dependence of A_0 can provide the potential strength. In particular, when A_0 does not depend on T a specific case of localized diffusion is realized—a continuous diffusion inside a strictly restricted volume. This type of diffusion we shall refer to as the bounded diffusion within a cage.

Finally we conclude that in the case of localized diffusion the problem is reduced to finding the total set of eigensolutions of the Hermitian operator \hat{H} with a specific potential obtained by the transformation of a drift potential [see Eq. (15)]. The momentum-time correlation function in this approach takes the form originating from Eq. (6) as follows:

$$F_s(\mathbf{k}, t) = \sum_n A_n(\mathbf{k}) \exp(-D\varepsilon_n t). \quad (23)$$

We note that this expression has a clear physical sense. One can expect different rates of diffusive movement which lead to different speeds of dephasing the coherently scattered radiation (similar to dephasing due to thermal vibrations described by the Lamb-Mössbauer factor). The weights of these movements depend on the propagation vector \mathbf{k} (both a magnitude and a direction) and on the form of the drift potential.

IV. BOUNDED DIFFUSION WITHIN A CAGE

Until now the drift potential was assumed to be a finite one having an arbitrary profile. In this section we regard a particular case where the potential equals zero inside a finite volume, called a cage, and is infinite outside. It can be shown that if the size of the cage tends to infinity then one arrives at the case of free diffusion. Let us consider this limit case in view of the general theory described above.

The potentials $U(\mathbf{r})$ and $V(\mathbf{r})$ are equal to zero in all space (actually in a volume Ω with a size being much larger than the wavelength of radiation). In this case a full set of eigenfunctions can be chosen in the form of plane waves $\psi_{\mathbf{p}}(\mathbf{r}) = \Omega^{-1/2} \exp(i\mathbf{p}\mathbf{r})$ corresponding to eigenvalues $\varepsilon_{\mathbf{p}} = p^2$. The ground state is $\psi_0 = \text{const} = \Omega^{-1/2}$. However, this state is not realized because $A_0 = 0$ for $\mathbf{k} \neq 0$. This can be

easily deduced from Eq. (22) for the potential considered. Moreover, the only one state with $\mathbf{p} = \mathbf{k}$ has a nonzero coefficient $A_{\mathbf{p}} = 1$. Thus, in the limit of an infinitely large cage we arrive once again at the universal resonance function characteristic for a free diffusion⁹

$$\varphi(\mathbf{k}, \omega) = \frac{i}{\omega + i\Gamma_0/2\hbar + iDk^2}. \quad (24)$$

The most simple case of bounded diffusion within a cage is realized when the cage has the form of a rectangular box with dimensions X, Y, Z in the directions x, y, z , correspondingly. This illustrative example might be also of a practical interest in the view of future experiments because it can be realized artificially with the control of parameters such as the diffusion coefficient or the size of the cage.¹⁷ Owing to the fact that the cages are oriented randomly in different parts of a real sample the task is spherically symmetrical. That is why, perhaps, in Ref. 17 the accurate solution of the Fokker-Planck equation with a spherical cavity of radius r as a hole in the Swiss cheese¹⁸ has been considered to analyze the experimental results. Nevertheless, it is useful to consider and analyze in detail the task of the rectangular cavity.

We take the potential $U(\mathbf{r})$ to be equal to zero inside the box and infinity outside of it. The potential $V(\mathbf{r})$ is also equal to zero inside the box but it has a singularity on the box walls. Unlike the similar task of quantum mechanics we have to find now the eigensolutions $\psi_n(\mathbf{r})$ which obey the boundary conditions $(\mathbf{n}\nabla\psi)_B = 0$ where \mathbf{n} is a normal to the box boundaries (see Ref. 16 for details). The eigensolutions for a particle diffusing inside the box with the coordinates $0 < x < X, 0 < y < Y, 0 < z < Z$ are found in the form

$$\psi_{hkl}(x, y, z) = f_h(x)f_k(y)f_l(z), \quad \varepsilon_{hkl} = \pi^2 \left(\frac{h^2}{X^2} + \frac{k^2}{Y^2} + \frac{l^2}{Z^2} \right), \quad (25)$$

where $h, k, l = 0, 1, 2, \dots$. Below we introduce the notation $j = h, k, l, s = x, y, z, S = X, Y, Z$. The solution is factorized into the functions independently describing the motion along each of the three main axes. A particular function is as follows:

$$f_j(s) = 2^{1/2} (C_j S)^{-1/2} \cos(\pi j s / S). \quad (26)$$

Here $C_j = 1$ for all $j > 0$ and $C_j = 2$ for $j = 0$.

One can readily find the weights of different diffusion states in accord with Eq. (21). These also consist of three independent factors

$$A_{hkl}(k_x, k_y, k_z) = a_h(k_x) a_k(k_y) a_l(k_z), \quad (27)$$

with

$$a_j(k_s) = \frac{1}{2C_j} \left[j_0 \left(\frac{k_s S - \pi j}{2} \right) + (-1)^j j_0 \left(\frac{k_s S + \pi j}{2} \right) \right]^2, \quad (28)$$

where $j_0(z) = \text{sinc} z$ is a zero-order spherical Bessel function. It is easy to verify that this formula gives the relation $a_j(0) = \delta_{j0}$. On the other hand, $a_0(k_s) = j_0^2(k_s S/2)$. Therefore the ground state ε_{000} is now really existing.

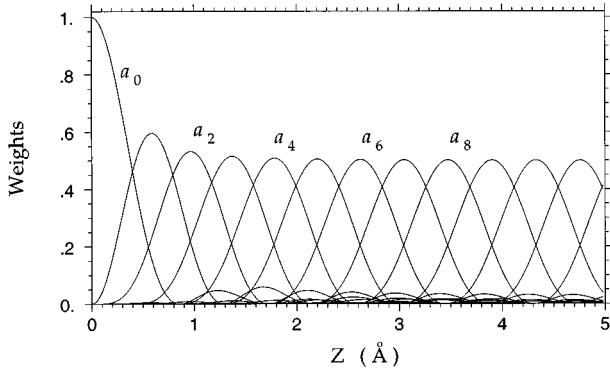


FIG. 1. The weights a_l of the Lorentzian functions contributing into the universal resonance function in the case of bounded diffusion (29) in dependence on the size of the one-dimensional cage Z ; a_0 is the weight of the partial resonance having the natural width Γ_0 .

In the model considered a dependence arises on the orientation of the cage with respect to the γ ray propagation direction. Let us take the orientation for which $k_x = k_y = 0$. In this case the radiation can probe only a diffusive motion along the z direction. Then the general formula (20) transforms to

$$\varphi(k_z, \omega) = \sum_{l=0}^{\infty} \varphi_l(k_z, \omega) = i \sum_{l=0}^{\infty} \frac{a_l(k_z)}{\omega + i q_l \Gamma_0 / 2\hbar}, \quad (29)$$

where the weights $a_l(k_z)$ and the relative widths $q_l(Z)$ of the partial resonances are determined by

$$a_l(k_z) = 4C_l^{-1}(k_z Z)^2 \frac{[1 + (-1)^{l+1} \cos(k_z Z)]}{[(l\pi)^2 - (k_z Z)^2]^2},$$

$$q_l = 1 + 2Dt_0 \frac{(l\pi)^2}{Z^2}, \quad (30)$$

where $t_0 = \hbar/\Gamma_0$ is the natural lifetime of the excited nucleus. The formula for the weights follows directly from Eq. (28). The maximum value of each term in the sum (29) equals $\varphi_l(k_z, 0) = 2t_0 a_l(k_z) q_l^{-1}$, i.e., it is determined by both the weight and the width of each Lorentzian contribution.

Let us analyze the shape of the φ function as a function of the box size Z . The coefficients a_l as the functions of Z are shown in Fig. 1 for the case of the ^{57}Fe nucleus with $k_z = 2\pi/\lambda = 73 \text{ nm}^{-1}$. Each coefficient has a pronounced main maximum and side maxima which are of much lower height. The main maxima for different coefficients are distributed over the Z axis rather regularly. After the initial fast drop down within the interval $0 < Z > \lambda$ the heights of the next maxima are reducing very slowly farther on. It is clearly seen in the figure that at any value of Z there are only a few (one, two, or a maximum three) significant coefficients while all others are very small. Hence only a few Lorentzians contribute essentially to the universal resonance function at any box size. The l indexes of the contributing Lorentzians increase with a rise of the size. The l value which corresponds to the main contribution, equals approximately $l_0 = k_z Z / \pi = 2Z/\lambda$. The relative width of this contribution is

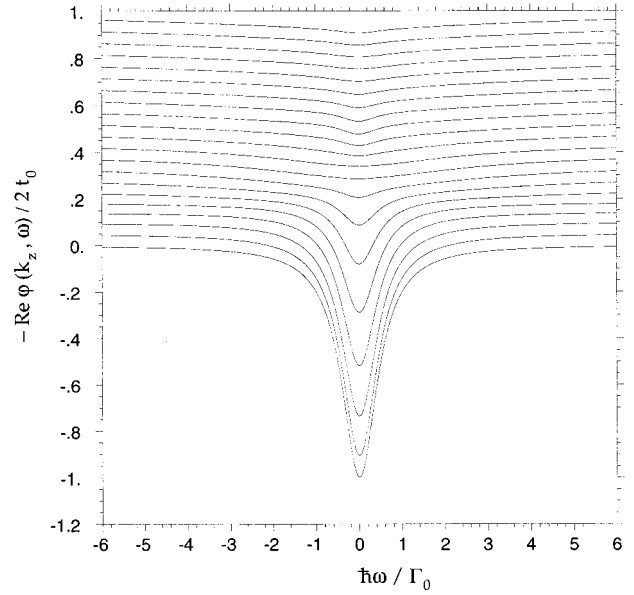


FIG. 2. The shape of the real part of the universal resonance function, describing the absorption ability of the nuclear target, at different sizes of the one-dimensional cage $Z=0, 0.01, \dots, 0.2 \text{ nm}$. Each next curve is shifted up on 0.05 relative to the previous one.

$q_{l_0} = 1 + 2Dk_z^2 t_0$. Thus, we see that the widths of the Lorentzians contributing in the φ function are dependent on Z only very softly when $Z > 2\lambda$.

Figure 1 shows that the behavior similar to that of free diffusion should appear already for not so large a box size, namely, about $Z > 2\lambda$ where the coefficient a_0 becomes close to zero. The existence of a zero term having a natural resonance width makes the situation quite different. Thus one should have a significant change of the resonance shape only within the transition region $0 < Z < 2\lambda$. Studying this transition region is most informative with respect to the potential parameters.

The absorption spectral function $f_a(\omega, Z) = -\text{Re}\varphi(k_z, \omega)/2t_0$ is displayed in Fig. 2 for this transition range of $Z=0, 0.01, \dots, 0.2 \text{ nm}$. For a better view each next curve is shifted up on 0.05 relative to the previous one. The calculation has been made for ^{57}Fe with $\lambda = 0.086 \text{ nm}$ and $D = 10^{-14} \text{ m}^2/\text{s}$. In this case $\Gamma_{fd}/\Gamma_0 \approx 16$ where $\Gamma_{fd} = \Gamma_0 + 2\hbar Dk_z^2$ is the resonance width in the case of free diffusion. As it follows from the calculation, an apparent decrease of the resonance dips of the function $f_a(\omega, Z)$ occurs there accompanied by a significant broadening of the resonance. The resonance shape close to that in the regime of free diffusion is approached already at $Z = \lambda$ where the weight of the unbroadened partial resonance drops to zero (the curve 9). Afterwards the coefficient a_0 increases again at about $Z = 1.4\lambda$ that results in a narrowing of the resonance for this range which is well seen in the figure (the curves 12–15).

We turn now to the time response of the nuclear ensemble. The analysis is the most simple in the case of a thin single line target. Here the intensity is proportional to z^2 , while the time dependence is determined by the following forward scattering function:

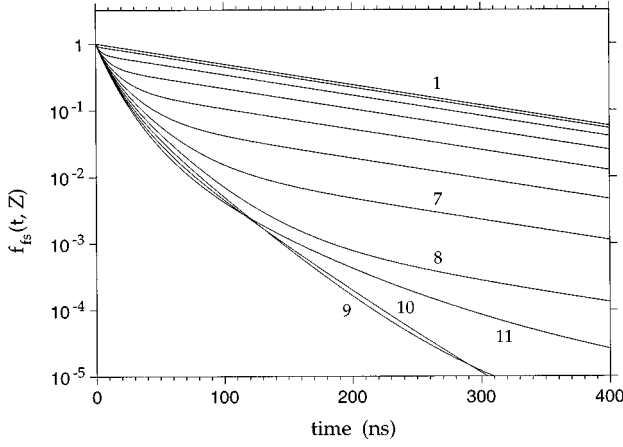


FIG. 3. The time dependence of nuclear forward scattering of synchrotron radiation in the limit of the thin target (31) in the case of bounded diffusion at different sizes of the one-dimensional cage $Z=0, 0.01, \dots, 0.1$ nm.

$$f_{fs}(t, Z) = \exp(-t/t_0) \left| \sum_{l=0}^{\infty} a_l(k_z) \exp(-l^2 \pi^2 D t / Z^2) \right|^2, \quad t > 0. \quad (31)$$

The entire time dependence of the forward scattering intensity is the sum of exponents where each term is characterized by its own decay region $t < t_l$ where $t_l = t_0(1 + 2Dt_0Z^{-2}\pi^2l^2)^{-1}$. So the number of contributing terms decreases when the delay time increases. Finally only the term having the natural decay time becomes dominant. Therefore the contribution of this term given by the coefficient a_0 can be well separated in the time dependence. The function $f_{fs}(t, Z)$ is shown in Fig. 3 for box sizes in the range $Z=0, 0.01, \dots, 0.1$ nm. The uppermost curve corresponds to a rigidly bounded nucleus. It is described by only a zero term with $a_0=1$ and $q_0=1$, i.e., it exhibits the natural decay. With the increase of the box size the diffusion is activated and the next terms related to the broader resonances start to contribute. This results in a faster decay of the scattering intensity observed within the initial time interval. However, until the zero term has a noticeable magnitude (the range $0 < Z < 0.06$ nm in Fig. 1), the natural decay rate is reached. It is manifested by the straight segments of the curves observed at later times in the logarithmic scale (the curves 1–7 in Fig. 3). *In this range of box sizes the coefficient a_0 can be determined directly by extrapolating the straight segments of the curves to zero time.*

The coefficient a_0 drops down sharply with the increase of Z , reaches its minimum and then slightly oscillates (Fig. 1). This behavior is reflected in the nonmonotonous transformation of the time dependences around $Z=0.09$ nm (the curves 8–11 in Fig. 3).

To illustrate the role of bounded diffusion in a scattering from a thicker nuclear target we consider the time response in the case of a single line sample having an effective resonance thickness $\mu_n z = 10$, where $\mu_n = K \sum_{ge} B_{ge}$ [see (3)]. The results of the computer calculation are shown in Fig. 4 for the same range of box sizes. The time dependence contains now the dynamical beats of intensity which are caused

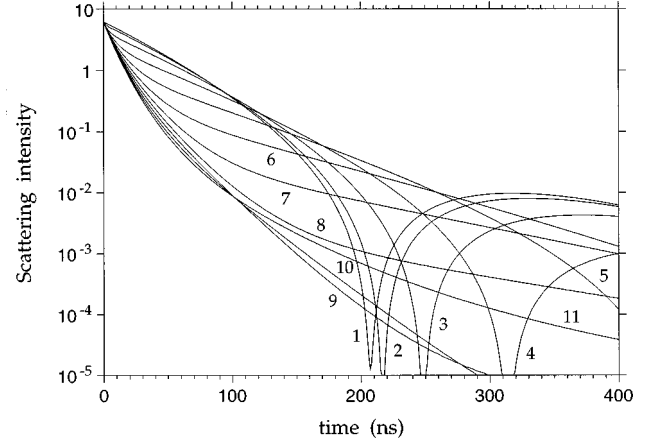


FIG. 4. The time dependence of nuclear forward scattering of synchrotron radiation by the thick target in the case of bounded diffusion at different sizes of the one dimensional cage $Z=0, 0.01, \dots, 0.1$ nm. The effective thickness of target $\mu_n z = 10$.

by multiple scattering of radiation by nuclei in the target. It is of interest that the dynamical beat pattern appears to be highly sensitive to the box size. With increasing the size the beat minima are shifting towards later times. Then they disappear completely in the observation time window (curves 1–5 in Fig. 4). This effect is related to a redistribution of weights of contributing Lorentzians, particularly to the dropping down of the coefficient a_0 which represents the rigidly bound state of the nucleus. The latter leads to decreasing the effective thickness of the target similar to that due to the fall down of the recoilless factor. We shall discuss this effect in more detail in the next section. The time evolution for the intermediate range of Z (curves 6–8 in Fig. 4) is similar to that for a thin target. The nonmonotonous transformation is seen again later on (curves 9–11 in Fig. 4). However, under conditions close to the free diffusion regime, i.e., for the well size large compared to the wavelength of radiation, the dynamical beat pattern specific for the full effective thickness of the target is restored.

The initial slope of all curves seems to be somewhat universal. This slope can be easily obtained analytically using the asymptotic behavior of the φ function. It is known that the asymptotic behavior of the universal resonance function at far tails of resonance allows us to estimate the temporal nuclear response at the initial time. We obtain directly from Eq. (29) in the case of large $|\omega|$ that

$$\lim_{|\omega| \rightarrow \infty} \varphi(k_z, \omega) = \frac{i}{\omega} \left(1 - i \frac{\Gamma_{fd}}{2\hbar\omega} + \dots \right), \quad \Gamma_{fd} = \Gamma_0 + 2\hbar D k_z^2. \quad (32)$$

Here Γ_{fd} just corresponds to the free diffusion case (see above). To derive this result one has to take into account the relations

$$\sum_{l=0}^{\infty} a_l(k_z) = 1, \quad \sum_{l=0}^{\infty} a_l(k_z) \varepsilon_l = k_z^2. \quad (33)$$

The first relation follows from the general theory [see Eq. (21)]. The second one is the consequence of the fact that inside the volume of the cage we have the same equation as

in the case of free diffusion (the potential is absent). Therefore $\varepsilon_l \psi_l(z) = -d^2 \psi_l(z)/dz^2$ and the relation is obtained by inserting this formula into the integral (21) which defines generally the coefficients $a_l(k_z)$ and then integrating by parts.

From Eqs. (1), (2), and (32) we obtain after expanding the exponent in the Taylor series and using the residue theorem

$$\lim_{t \rightarrow 0} I_{fs}(t, z) = |E_0(z)|^2 \left(\frac{\mu_n z}{4t_0} \right)^2 \times \left(1 - t \left[\frac{1}{t_0} + 2Dk_z^2 + \frac{\mu_n z}{4t_0} \right] + \dots \right). \quad (34)$$

In accordance with this formula the initial slope is determined by three physical reasons (represented by the three terms in the straight brackets), namely, by natural decay of the excited state, by diffusive motion of nuclei, and by coherent speedup of decay. The Z dependence of the initial slope is absent. Usually the term related to a diffusion brings the main contribution and this determines the behavior of the curves in Figs. 3, 4. *The time dependence of intensity in the case of limited diffusion cannot be faster than in the case of free diffusion.*

This fact has a clear physical sense. In the limit of small delay time only displacements of nuclei which are smaller than the cage size are essential. Therefore the walls of the cage cannot influence the time behavior. In the opposite limit of very large delay time, the picture of nuclear motion is influenced essentially by the reflections from walls. On average, the coherent nuclear scattering is related to the mean position of a nucleus, as if it is at rest. The strength of this scattering channel is described by the coefficient a_0 which plays the same role as the Lamb-Mössbauer factor which takes into account the thermal motion. Obviously when the mean displacement of the nucleus $Z/2$ becomes comparable with the wavelength of the radiation λ the coefficient a_0 tends to zero due to a dephasing of the scattered waves. When the coefficient a_0 is not too small one can find in the case of the thin target a characteristic intermediate time range (about 80–120 ns in Fig. 3) where the transition from the free diffusion regime to the natural nuclear decay occurs. The time of transition can be estimated as $t_{tr} \approx \ln(1/a_0)/2Dk_z^2 \approx 10 \ln(1/a_0)$ ns in the case considered (^{57}Fe , and $D = 10^{-14}$ m²/s). It depends significantly on the size of cage Z through the coefficient a_0 , on the diffusion coefficient D as well as on the wavelength of the radiation.

The real sample may contain different cages with different sizes and orientation. Therefore the mean value has to be calculated. This procedure is essential in fitting the experimental data. However the general properties of the time spectra considered here will be unchanged.

V. JUMP DIFFUSION

The theory developed for the quasielastic Mössbauer spectroscopy^{5,19} describes a jump diffusion on simple Bravais lattices where all crystal sites are equivalent. The extension of the theory to the case of non-Bravais crystal lattice was done in Refs. 20,21. Here we shall use the results of Ref. 21 where the correlation function $F_s(\mathbf{k}, t)$ has been calcu-

lated in the more general case. This function can be written in a form similar to Eq. (23), namely,

$$F_s(\mathbf{k}, t) = \sum_l a_l(\mathbf{k}) \exp[-\varepsilon_l(\mathbf{k})t/2t_0],$$

$$a_l = \left| \sum_{i=1}^m \sqrt{c_i} \beta_{il} \right|^2, \quad \sum_l a_l = 1, \quad (35)$$

where $t_0 = \hbar/\Gamma_0$ is the nuclear lifetime as above, i is the index of inequivalent sites inside the primitive unit cell of the crystal lattice. This site has the i th local symmetry. The relative l th decay rate $\varepsilon_l(\mathbf{k})$ and the vector $\beta_{il}(\mathbf{k})$ are the eigenvalue and orthonormalized eigenvector of the probability jump matrix, which is a Hermitian one

$$A_{ij}(\mathbf{k}) = \frac{2t_0}{\sqrt{c_i}} \left[\delta_{ij} \sum_q \frac{1}{\tau_{iq}} - \frac{1}{n_{ji} \tau_{ji}} \sum_n \exp(i\mathbf{k} \mathbf{R}_{ij}^{(n)}) \right] \sqrt{c_j}. \quad (36)$$

Here and above, c_i is the probability of the occupation of the i th sublattice, n_{ji} is the number of sites of the i th sublattice which surround the site of the j th sublattice, τ_{ij}^{-1} is the jump rate from the site of symmetry i to any nearest-neighbor site of symmetry j , $\mathbf{R}_{ij}^{(n)}$ is the n th vector distance of the set of distances between nearest-neighbor sites of the i th and j th sublattices. Detailed balance demands that

$$\frac{c_i}{n_{ij} \tau_{ij}} = \frac{c_j}{n_{ji} \tau_{ji}}. \quad (37)$$

If $\tau_{ij} = \tau_{ji}$ then $c_j = \text{const} = 1/m$ and $a_l = m^{-1} |\sum_i \beta_{il}|^2$.

Expression (35) leads immediately to the following expression for the universal resonance function:

$$f(\mathbf{k}, \omega) = i \sum_n \frac{a_l(\mathbf{k})}{\omega + i q_l(\mathbf{k}) \Gamma_0 / 2\hbar}, \quad q_l(\mathbf{k}) = 1 + \varepsilon_l(\mathbf{k}). \quad (38)$$

This formula implies that each resonance becomes a superposition of several resonances having the same position but different widths, i.e., like in the case of bounded diffusion Eqs. (23) and (32).

As an example, we consider in detail the case of jump diffusion in the alloy Fe₃Si which was investigated recently using the nuclear forward scattering of synchrotron radiation.²² The time dependences of the forward scattering along the [113] crystal direction were measured. Single crystals of Fe₃Si have a cubic superstructure consisting of four sublattices. In the entirely ordered crystals the three sublattices are occupied by iron atoms while the fourth one is occupied by Si atoms. The diffusion mechanism of iron in this structure was studied.²⁰ It was proven that in accordance with earlier suggestions iron atoms jump between three iron sublattices only and avoid the fourth silicon sublattice. Therefore the matrix A_{ij} takes the form

$$\hat{A} = \begin{pmatrix} 2\nu & -E\nu & -E^*\nu \\ -E^*\nu & \nu & 0 \\ -E\nu & 0 & \nu \end{pmatrix}, \quad \nu = \frac{2t_0}{\tau}, \quad (39)$$

where

$$E = \cos(k_x d) \cos(k_y d) \cos(k_z d) + i \sin(k_x d) \sin(k_y d) \sin(k_z d) \quad (40)$$

with

$$(k_x, k_y, k_z) = (1, 1, 3)K/\sqrt{11}, \quad d = a/4. \quad (41)$$

Here $K = 2\pi/\lambda = \omega/c$ is the wave number of incident radiation (which fits the nuclear resonance transition in ^{57}Fe in our case), a is a crystal lattice constant of Fe_3Si .

Taking into account the specific values $K = 72.98 \text{ nm}^{-1}$, $a = 0.571 \text{ nm}$ one obtains that $k_x d = 3.1411$, i.e., very close to π . Therefore approximately $E = E^* = -1$ and with definite accuracy the jump matrix (39) has the following analytical eigensolutions

$$\begin{aligned} q_l &= \begin{matrix} 3\nu & \nu & 0, \\ \beta_l &= \frac{1}{\sqrt{6}} \begin{pmatrix} 2 \\ 1 \\ 1 \end{pmatrix} & \frac{1}{\sqrt{2}} \begin{pmatrix} 0 \\ 1 \\ -1 \end{pmatrix} & \frac{1}{\sqrt{3}} \begin{pmatrix} -1 \\ 1 \\ 1 \end{pmatrix}, \\ a_l &= \begin{matrix} 8/9 & 0 & 1/9. \end{matrix} \end{aligned} \quad (42)$$

It follows from Eq. (42) that only two resonance contributions have nonzero weight, one having the natural width and another broadened. Correspondingly the universal resonance function takes the form having only one parameter

$$\varphi(\mathbf{k}, \omega) = \frac{i}{9} \left(\frac{8}{\omega + iq/2t_0} + \frac{1}{\omega + i/2t_0} \right), \quad q = 1 + 6t_0/\tau. \quad (43)$$

In the range of high temperatures where diffusion takes place the hyperfine splitting of the nuclear levels is absent and the time-dependent electric field of the scattered wave is described by the following equation:

$$E(t, z) = E(z) \int \frac{d\omega}{2\pi} \exp\left(-i\omega t - \frac{\mu_n z}{4t_0} \varphi(\mathbf{k}, \omega - \omega_0)\right), \quad (44)$$

where ω_0 is a resonance frequency, z is a crystal plate thickness, and $\mu_n = N\sigma_0\eta f_{\text{LM}}$ is an absorption coefficient at resonance with N being a number of iron atoms in the unit volume, σ_0 being a nuclear cross section at resonance, η being an enrichment by resonant isotope ^{57}Fe and f_{LM} being the Lamb-Mössbauer factor.

We compare the time-dependent forward scattering intensity for different diffusion rates, respectively, different q factors. The time dependences for the two effective thicknesses of target $\mu_n z = 3$ and 21.5 are displayed in Figs. 5, 6. In a thin target limit using approximation (9) one can readily find that the time response is described by the sum of two exponential functions, one of which exhibits the natural decay of the nuclear excitation while another shows the accelerated decay. With an increase of the q factor the acceleration of decay is well seen within an initial time interval in Fig. 5. In the limit $q \gg 1$ the two exponential functions turned out to be well separated in time and the two stages of the decay are clearly observed.

The time dependences for the thicker sample have a more complicated character. The initial stage of decay mostly fol-

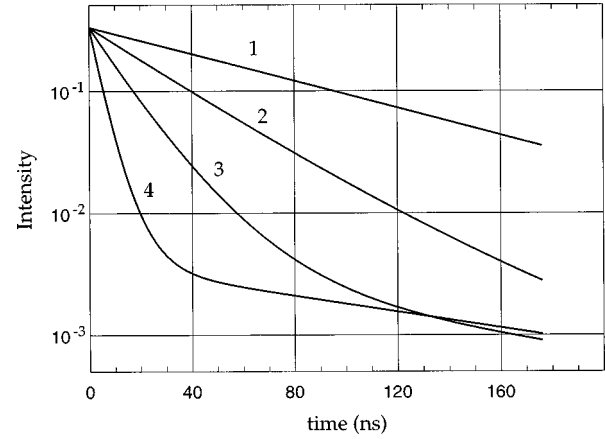


FIG. 5. The time dependence of nuclear forward scattering of synchrotron radiation from Fe_3Si crystal in the direction [113] in the presence of jump diffusion at different q factors (see text). Curves 1–4 correspond to $q = 2.2, 5.1, 11.5,$ and 36 , respectively. The effective thickness of target $\mu_n z = 3$.

lows the same law described above but at a later time the dynamical beats appear within the natural lifetime of nuclear excitation. It is of interest to note that a position of the beat minimum is sensitive to a diffusion rate (like in the case of bounded diffusion where it is sensitive to the cage size, see Fig. 4). The minimum is shifting towards the later times with an increase of the diffusion coefficient. Such a behavior is different from that predicted⁹ for the case of free continuous diffusion where the beat pattern position is unchanged. This difference is tightly related to the difference in form of the resonance universal function. In the case of free diffusion it is a single Lorentzian having variable width. Here and in the case of bounded diffusion it is the sum of several Lorentzians having different weights and widths.

The split of the universal resonance function into several terms leads to the fragmentation of the effective resonance thickness of a target into relevant partial thicknesses which are $a_l \mu_n z$. The contributing Lorentzians turn out to determine essentially the decay of the nuclear exciton within dif-

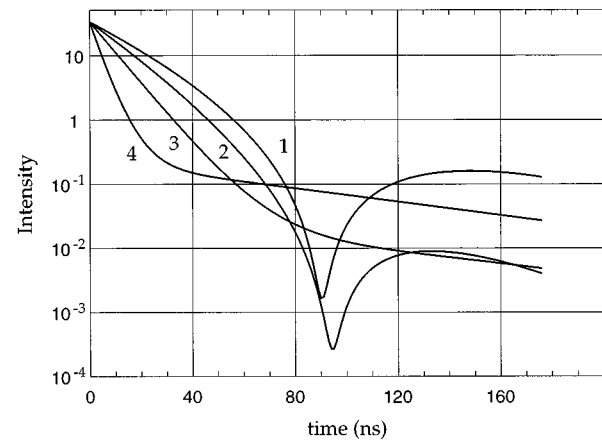


FIG. 6. The time dependence of nuclear forward scattering of synchrotron radiation from the Fe_3Si crystal in the direction [113] in the presence of jump diffusion at different q factors. Curves 1–4 correspond to $q = 2.2, 5.1, 11.5,$ and 36 , respectively. The effective thickness of target $\mu_n z = 21.5$.

ferent time intervals in dependence on the Lorentzian widths. This leads, roughly saying, to the fact that the target exhibits its different partial thicknesses in different parts of the time dependence of the forward scattering. In general, the effect of the thickness split is revealed for any relation between the weights and widths of contributing Lorentzians. Obviously the dynamical beat pattern should be sensitive to this relationship. This is actually the physical reason for the shift of the beat minima with the change of both the potential well size and the diffusion rate.

In our example in the limit of large- q factor $q \gg 1$, where two contributing Lorentzians have essentially different widths, one can distinguish two characteristic stages in the entire time dependence. At an early stage of decay the tails of the resonance are essential and one can consider only the broadened contribution. Making use of the result obtained in Ref. 9 for the case of one broadened resonance one obtains

$$I(t, z) = \frac{I_0}{\Delta \omega} \frac{T_1 t_0}{4t} \exp\left(-\mu_e z - \frac{qt}{2t_0}\right) J_1^2(\sqrt{T_1 t/t_0}),$$

$$t < t_0/q, \quad (45)$$

where $J_1(x)$ is a Bessel function of first order and $T_1 = 8\mu_n z/9$. In this time interval the evolution of the forward scattered intensity is similar to the case of free diffusion, and via parameter q one can determine the diffusion coefficient $D = a^2/32\tau$.

At later times, on the contrary, the region near the center of the resonance is essential in the integral (44). Therefore one can neglect ω in the first term of Eq. (43) and consider approximately the case with one resonance of natural width. As a result the time evolution takes the form

$$I(t, z) = \frac{I_0}{\Delta \omega} \frac{T_2 t_0}{4t} \exp\left(-\left[\mu_e + \frac{8\mu_n}{9q}\right]z - \frac{t}{2t_0}\right)$$

$$\times J_1^2(\sqrt{T_2 t/t_0}), \quad t > t_0/q, \quad (46)$$

where $T_2 = \mu_n z/9$. We note that similar to the case of bound diffusion the broad resonance effectively provides an absorption in addition to the electronic absorption, while the effective thickness of the target turned out to be less in accord with the weight of the resonance of the natural width. Thus one and the same target exhibits, in this case, behavior of the target with the effective thickness T_1 at an early stage and with T_2 at a later stage of decay.

Finally we want to note that the consideration presented above deals with the pure diffusion process, while the relaxation process is assumed to be the same as in the case without diffusion.

VI. CONCLUSION

The response of the nuclear ensemble in the presence of diffusive motion of nuclei is described by the universal resonance function $\varphi(\mathbf{k}, \omega)$ which is related to the Van Hove space-time correlation function $G(\mathbf{r}, t)$. While considering a scattering problem it is natural to use the momentum-time correlation function $F(\mathbf{k}, t)$ which enters directly into the time dependence of nuclear exciton decay in the limit of a

thin target for any regime of diffusion. The spectral density of this function $F(\mathbf{k}, \omega)$ is found by the Fourier transformation of $F(\mathbf{k}, t)$. Actually the universal resonance function $\varphi(\mathbf{k}, \omega)$ represents the averaging of the standard resonance amplitude describing the scattering process by a static nucleus over the spectral density function $F(\mathbf{k}, \omega)$ which reflects the motion of particles in the nuclear ensemble owing to the Doppler effect.

The analytical solution for time dependences of the coherent forward scattering of SR can be obtained only for the case of free diffusion where the universal resonance function has Lorentzian shape. The additional width of the resonance line is simply proportional to the diffusion coefficient in this case. Respectively, an additional exponential factor appears in the time response, the decrement of which contains the diffusion coefficient. This yields an accelerated decay of the coherent signal. As to the dynamical beat structure it does not depend on the resonance broadening.

In contrast to the regime of free diffusion in the case of bounded diffusion inside a potential well, and in the case of jump diffusion between different sites (vacancies in solids) the universal resonance function has a more complicated shape represented in general by the coherent superposition of the Lorentzian functions where the weight and the width of a separate Lorentzian are determined by the specific character of the diffusion process. The main physical parameters affecting the shape of the universal function are the diffusion coefficient, temperature, the drift potential profile, and the jump rate.

Such a shape of the universal function corresponds to a more complicated behavior of the time response. In general, there are several stages of the decay which are characterized by the different decay rates. The initial stage reveals a faster monotonous decay which is more accelerated the larger diffusion coefficient and the size of the potential well are. At later times the decay rate becomes slower and the dynamical beats appear in the case of the thick target. The dynamical beat pattern is transformed drastically (in contrast to the free diffusion regime) depending either on the size of the potential well in the case of bounded diffusion or on the temperature in the case of jump diffusion. When both parameters are rising, the transition to a beat pattern characteristic for a thinner target occurs.

The physical reason for this result is in the split of the universal resonance function into several terms. It leads to a fragmentation of the effective resonance thickness of the target into relevant partial thicknesses. When the contributing Lorentzians have essentially different widths the target exhibits its partial thicknesses in the time dependence within different time intervals. Hence a time-variable thickness is characteristic for the nuclear exciton decay in these cases rather than a unique thickness. In general, the effect of the thickness split is revealed for an arbitrary relation between the weights and widths of contributing Lorentzians. The dynamical beat pattern is sensitive to this relationship. However, under conditions close to the free diffusion regime, i.e., for the well size large compared to the wavelength of the radiation in the case of bounded diffusion, or for high temperatures in the case of jump diffusion, the dynamical beat pattern is restored to the full effective thickness of the target.

The time evolution of the coherent forward scattering of synchrotron radiation is thus transformed not only quantitatively but also qualitatively in contrast to the relevant Mössbauer spectra. This higher sensitivity of measurement is because an interference technique (forward scattering) reveals

complex amplitudes of oscillations of the electromagnetic field, while a spectroscopic method (resonance absorption) exhibits only their strengths (see Ref. 8). This makes forward scattering of synchrotron radiation to be a perspective method for the studies of diffusion.

-
- ¹E. R. Bauminger and I. Nowik, in *Mössbauer Spectroscopy*, edited by D. P. E. Dickson and F. J. Berry (Cambridge University Press, Cambridge, 1986).
- ²A. Abras and J. G. Mullen, *Phys. Rev. A* **6**, 2343 (1972).
- ³G. Vogl, *Hyperfine Interact.* **53**, 197 (1990); in *Mössbauer Spectroscopy Applied to Magnetism and Materials Science*, edited by Gary J. Long and Fernande Grandjean (Plenum, New York, 1996), Vol. 2, p. 85.
- ⁴G. U. Nienhaus and F. Parak, *Hyperfine Interact.* **90**, 243 (1994).
- ⁵K. S. Singvi and A. Sjölander, *Phys. Rev.* **120**, 1093 (1960).
- ⁶J. P. Hannon and G. T. Trammell, in *Resonant Anomalous X-ray Scattering*, edited by G. Materlik, C. J. Sparks, and K. Fischer (Elsevier, Amsterdam, 1994), p. 565.
- ⁷E. Gerdau and U. van Bürck, in *Resonant Anomalous X-ray Scattering* (Ref. 6), p. 589.
- ⁸G. V. Smirnov, *Hyperfine Interact.* **97/98**, 551 (1996); in *X-ray and Inner-Shell Processes*, Proceedings of 17th International Conference, edited by R. L. Johnson, B. F. Sonntag, and H. Schmidt-Bocking, AIP Conf. Proc. No. 389 (AIP, New York, 1997), p. 323.
- ⁹G. V. Smirnov and V. G. Kohn, *Phys. Rev. B* **52**, 3356 (1995).
- ¹⁰Yu. Kagan, A. M. Afanasev, and V. G. Kohn, *J. Phys. C* **12**, 615 (1979).
- ¹¹M. A. Krivoglaz and S. P. Repetskii, *Fiz. Met. Metalloved.* **32**, 1 (1971) [*Phys. Met. Metallogr. (USSR)* **32**, 8991 (1971)].
- ¹²G. Vogl, W. Mansel, and P. H. Dederichs, *Phys. Rev. Lett.* **36**, 1497 (1976).
- ¹³W. Petry, G. Vogl, and W. Mansel, *Phys. Rev. Lett.* **45**, 1862 (1980).
- ¹⁴W. Petry and G. Vogl, *Z. Phys. B* **45**, 207 (1982).
- ¹⁵W. Nadler and K. Schulten, *Phys. Rev. Lett.* **51**, 1712 (1983).
- ¹⁶A. M. Afanasev and V. E. Sedov, *Phys. Status Solidi B* **131**, 299 (1985).
- ¹⁷A. S. Plachinda, V. E. Sedov, V. I. Khromov, I. P. Suzdalev, V. I. Goldanskii, G. U. Nienhaus, and F. Parak, *Phys. Rev. B* **45**, 7716 (1992).
- ¹⁸F. Volino and A. J. Dianoux, *Mol. Phys.* **41**, 271 (1980).
- ¹⁹T. Chudley and R. J. Elliott, *Proc. Phys. Soc. London* **77**, 353 (1961).
- ²⁰B. Sepiol and G. Vogl, *Phys. Rev. Lett.* **71**, 731 (1993).
- ²¹O. G. Randl, B. Sepiol, G. Vogl, R. Feldwisch, and K. Schroeder, *Phys. Rev. B* **49**, 8768 (1994).
- ²²B. Sepiol, A. Meyer, G. Vogl, R. Rufer, A. I. Chumakov, and A. Q. R. Baron, *Phys. Rev. Lett.* **76**, 3220 (1996).