On the theory of nuclear resonant forward scattering of synchrotron radiation

V. G. Kohn* and G. V. Smirnov†
Russian Research Center "Kurchatov Institute," 123182 Moscow, Russia
(Received 6 April 2007; revised manuscript received 23 August 2007; published 28 September 2007)

The problem of time-dependent nuclear forward scattering of synchrotron radiation is solved using the Heitler's quantum theory of radiation. We have shown a way of derivation of the analytical solution that coincides precisely with the formula, earlier evolved by Kagan *et al.* [J. Phys. C **12**, 615 (1979)] within the approach based on the solution of the Maxwell's equations. The work was stimulated by the result presented by Hoy *et al.* [Phys. Rev. B **63**, 184435 (2001)], where the solution of the problem was obtained in the form of a finite series. This solution was derived after a replacement of the solid crystal by a finite set of the "effective" nuclear planes. The mean distance between the planes is much larger than the lattice parameter. Although the solution by Hoy *et al.* is close numerically to the well known and correct solution for large sample thickness containing many effective planes, it is incorrect for small sample thickness because it artificially limits the number of multiple scattering paths in nuclear target of a particular thickness.

DOI: 10.1103/PhysRevB.76.104438 PACS number(s): 76.80.+y, 78.90.+t, 42.25.Bs

I. INTRODUCTION

The discovery of the recoilless absorption and emission of γ quanta by nuclei in crystals, known as Mössbauer effect, led to the development of spectroscopy of an extremely high resolution. The capabilities of the Mössbauer spectroscopy are provided by the exceptional narrowness of the nuclear isomers levels. For example, a level width Γ of the nucleus ^{57}Fe in the first excited state is only 4.66×10^{-9} eV. This causes extremely high sensitivity of Mössbauer spectroscopy to tiny perturbations of the nuclear resonance energy. Due to this sensitivity, it is possible to make resolved hyperfine splitting of nuclear levels, to measure their chemical shifts, to observe the redshift of photon in gravitational field, and to measure with high accuracy the changes in shape and width of nuclear absorption line caused by various relaxation processes.

Besides applications in spectroscopy, Mössbauer effect is a very interesting coherent phenomenon. The coherence was revealed already in the first studies of the time dependence of transmission of γ radiation through the resonant target.[1] Obviously, the time scale of the coherence preservation is determined by the collision time of γ-ray photon with the nucleus target. This time is close to the lifetime of the nuclear excited state, which, e.g., in the case of ^{57}Fe nucleus is $\sim$0.14 μs. During such collision time, more than 10^{11} phased oscillations of the nuclear transition current occur, evidencing the unique scale of the phase memory conservation. The most favorable conditions to study the coherent aspects of nuclear resonant scattering had arisen after the creation of the pulsed sources of synchrotron radiation.[2,3]

The theory of nuclear resonant scattering of γ radiation was developed by different groups within the scope of three formalisms. The first one is based on the solution of the Maxwell's equations where the medium properties are taken into account with the complex space- and time-dependent polarizability. This approach is close to Laue's phenomenological theory of x-ray diffraction in crystals (see, e.g., Ref. 4). However, the polarizability of medium is calculated by the methods of quantum mechanics. Therefore, the developed theory is correct for a description of coherent (i.e., elastic) nuclear resonant scattering. At the same time, this approach is relatively simple and allows one to obtain the solution in the case of complex and spatially heterogeneous problems.

The other two approaches are based on the quantum optics. The second one is founded on the use of the Heitler's quantum theory of radiation (see, e.g., Ref. 5). In this theory, the state of the system is described by defined sets of photons and scatterers. The probability amplitudes are introduced to find a definite number of particles in the system, and the coupled equations for the probability amplitudes are derived. The third approach corresponds to the quantum electrodynamics. It is founded on the solution of the Dyson equations for Green's functions (see, e.g., Ref. 6).

The first theory describes experimental results correctly when many photons are detected. It allows one to calculate easily the intensity of coherent scattering from the systems having complicated geometries of samples or for multicrystalline experimental setups. The quantum optics theories allow one to take into account many delicate effects from first principles, but they are started with rather complicated equations. Their solution is possible only if a large number of simplifying conditions based on *a priori* knowledge of the scattering conditions are accepted.

In the present work, we consider the problem of time-dependent nuclear forward scattering (NFS) of γ radiation. This problem was solved in the frames of the first approach for the case of a radioactive source.[1] Later, the same problem was solved in the frames of the second approach[7] where the same result was obtained, but at the price of much more laborious derivations.

The theoretical description of the NFS of synchrotron radiation (SR) was presented in Ref. 8 in the frames of the first approach. The amplitude of the coherent NFS of the monochromatic electromagnetic wave was found by means of solution of the Maxwell's equations. Then, the frequency spectrum of SR was multiplied by the amplitude of NFS and the inverse Fourier transformation was performed. As a result, the following analytical expression for the time-dependent amplitude of the radiation field scattered by nuclear en-

semble was obtained in the case of unsplit nuclear transition at the frequency ω_j:

$$E(z,t) = -\theta(t)\exp(-i\omega_j t - t/2t_0)\frac{1}{t_0}\left(\frac{\mu_n z t_0}{4t}\right)^{1/2} J_1\left(\left[\frac{\mu_n z t}{t_0}\right]^{1/2}\right), \tag{1}$$

where $\theta(t)$ is the stepwise Heaviside function, J_1 is the Bessel function of first order, $\mu_n = N_0\sigma_0\eta f$ is the linear nuclear absorption coefficient, and z is the thickness of the target. In the expression for the absorption coefficient, N_0 is the number of nuclei per unit volume, σ_0 is the resonance cross section, η is the isotopic enrichment of the target, and f is the Lamb-Mössbauer factor. We note that in the large series of works by Kagan (see review in Ref. 9), both the first and the second approaches were used to solve the various problems.

The first approach allowed one to solve also rather simply the problem of Bragg scattering from a crystal,[8] and not only from a free target but from the target subjected to ultrasound,[10,11] as well as from the target having the hyperfine splitting of nuclear transition.[12] For the case of NFS, the theory was generalized to account for the atomic diffusion in the sample.[13–15] In addition, the possibility of the fast reorientation of spins by magnetic field during scattering process was taken into account.[16,17] In Ref. 17, a general solution applicable actually to any type of time-dependent interactions was derived. In all cases, where the theory was compared with the experiment, the agreement between the theoretical and experimental results was lying within the experimental error.

The third approach was also used in a large number of works, beginning from the pioneer works by Hannon and Trammell (see review in Ref. 18). This way, the quantum beats in the time dependences related to the hyperfine splitting of resonance level were considered.[19] In spite of the difference between the approaches in the two theories mentioned above, all results obtained in both of them for the time-dependent NFS were coincident.

The second approach was applied to describe the process of the NFS of SR rather recently.[20] The initial set of the coupled equations was written following the procedure employed first for the case of Mössbauer source in Ref. 7. However, while searching the solution of the coupled equations, the authors had applied a simplified model of the target, namely, they presented the target by a finite series of the "effective" nuclear planes, where only the scattering can occur. One should say that *a priori*, the model was not well justified from the physical point of view and was introduced with the aim of finding the analytical solution of the coupled equations. Namely, they obtained expression for the time-dependent probability amplitude to find a γ-ray photon scattered in the forward direction in the form

$$\psi(N,t) = -CN\exp(-i\omega_j t - \Gamma t/2\hbar) \times\left[1 + \frac{1}{N}\sum_{n=1}^{N-1}\left(\frac{-\gamma_R}{2\hbar}\right)^n\binom{N}{n+1}\frac{t^n}{n!}\right], \tag{2}$$

where C is normalization factor and γ_R is the radiative width of nuclear transition. As seen, the solution is found through the phenomenological parameter N, which cannot be determined without an additional information. The parameter N is proportional to the effective resonance thickness of the target. The authors suggest finding the relation between them from the fitting of the experimental data with the theory or by comparison of the first term in the sum [Eq. (2)] with the scattering theory in the first Born approximation.

Thus, we are in front of the two solutions of the same problem. Solution (1) has been obtained by the first approach and confirmed by the third approach. Solution (2) was obtained by the second approach. This raises the question of whether it is possible to obtain solution (1) also by the second approach. The answer is yes. It is just the aim of this work to show how solution (1) can be derived by means of the quantum theory. In addition, we will discuss why solution (2) can lead to the problems. In the next section, we present shortly the basic equations of the quantum theory of radiation. In Sec. III, we present our solution of the equations which leads to Eq. (1). The discussion of the obtained solution is held in Sec. IV.

II. BASIC EQUATIONS

In this section, we present the basic equations of the quantum theory of radiation that were also employed in Refs. 7 and 20. The system in the primary state is represented by a set of the resonant nuclei and the photons of SR, described by the plane waves. In the case of our interest, all nuclei have only one excited state, characterized by the frequency $\omega_j = E_j/\hbar$. The photons excite the nuclei, which, in turn, give birth to the secondary photons and to the conversion electrons, also described by the plane waves. The general state of the system at the time t can be represented in the form

$$\Psi(t) = \Psi_{SR}(t) + \Psi_{nucl}(t) + \Psi_{scat}(t) + \Psi_{ce}(t), \tag{3}$$

where the different terms in the sum describe the states with the particular particles created,

$$\Psi_{SR}(t) = \sum_{\mathbf{k}} a_{\mathbf{k}}(t)\exp(-i\omega_{\mathbf{k}}t)|\mathbf{k};0;0;0\rangle, \tag{4}$$

$$\Psi_{nucl}(t) = \sum_{m} b_m(t)\exp(-i\omega_j t)|0;m;0;0\rangle, \tag{5}$$

$$\Psi_{scat}(t) = \sum_{\mathbf{k}} c_{\mathbf{k}}(t)\exp(-i\omega_{\mathbf{k}}t)|0;0;\mathbf{k};0\rangle, \tag{6}$$

$$\Psi_{ce}(t) = \sum_{\mathbf{p}} d_{m,\mathbf{p}}(t)\exp(-i\omega_{\mathbf{p}}t)|0;0;0;m,\mathbf{p}\rangle. \tag{7}$$

Here, the wave function in the occupation number representation $|a;b;c;d\rangle$ describes the state in which definite numbers of the particles of every sort are present. The quantities $a_{\mathbf{k}}(t)$, $b_m(t)$, $c_{\mathbf{k}}(t)$, and $d_{m,\mathbf{p}}(t)$ represent the probability amplitudes for the creation of the relevant particles, namely, of the photon of SR, and having momentum $\hbar\mathbf{k}$, of the nucleus in the crystal with index m, of the secondary photon scattered

by nuclear system and having momentum $\hbar\mathbf{k}$, and of the conversion electron emitted by the mth nucleus and having momentum $\hbar\mathbf{p}$.

The probability amplitudes are found from the solution of the Schrödinger equations. The Hamiltonian of the system is conveniently presented in the form $H=H_0+\hbar V$, where H_0 describes the system of noninteracting particles, and $\hbar V$ presents the interaction. The set of the coupled equations is most simply expressed in terms of the frequency spectrum of the relevant amplitudes. We write down the decomposition over the frequency spectrum

$$c_{\mathbf{k}}(t)\exp(-i\omega_{\mathbf{k}}t)=\frac{i}{2\pi}\int_{-\infty}^{\infty}d\omega C_{\mathbf{k}}(\omega)\exp(-i\omega t) \tag{8}$$

and analogously for the other amplitudes. The equations for the spectral density amplitudes can be written in the form

$$\begin{aligned}(\omega-\omega_j+i\varepsilon)B_m(\omega)=&\sum_{\mathbf{k}}A_{\mathbf{k}}(\omega)V_{\mathbf{k},j}\exp(i\mathbf{k}\mathbf{r}_m)\\&+\sum_{\mathbf{k}}C_{\mathbf{k}}(\omega)V_{\mathbf{k},j}\exp(i\mathbf{k}\mathbf{r}_m)\\&+\sum_{\mathbf{p}}D_{m,\mathbf{p}}(\omega)V_{\mathbf{p}}\exp(i\mathbf{p}\mathbf{r}_m),\end{aligned} \tag{9}$$

$$(\omega-\omega_{\mathbf{k}}+i\varepsilon)C_{\mathbf{k}}(\omega)=\sum_m B_m(\omega)V^*_{\mathbf{k},j}\exp(-i\mathbf{k}\mathbf{r}_m), \tag{10}$$

$$(\omega-\omega_{\mathbf{p}}+i\varepsilon)D_{m,\mathbf{p}}(\omega)=B_m(\omega)V^*_{\mathbf{p}}\exp(-i\mathbf{p}\mathbf{r}_m). \tag{11}$$

Here, the pole is shifted to the lower part of the complex plane in order to provide the condition for all time-dependent amplitudes to be equal zero at $t<0$. The matrix elements of the interaction Hamiltonian describe the probabilities of different transitions in the nuclear system.

The set of equations can be partially solved by substituting Eqs. (10) and (11) into Eq. (9) and by isolating the processes at a single nucleus. As a result, the equation for the amplitude of nuclear excitation is obtained in the form

$$\begin{aligned}(\omega-\omega_j+i\Gamma/2\hbar)B_m(\omega)=&\sum_{\mathbf{k}}A_{\mathbf{k}}(\omega)V_{\mathbf{k},j}\exp(i\mathbf{k}\mathbf{r}_m)\\&+\sum_{m'\neq m}B_{m'}(\omega)\sum_{\mathbf{k}}|V_{\mathbf{k},j}|^2\\&\times\frac{\exp(i\mathbf{k}[\mathbf{r}_m-\mathbf{r}_{m'}])}{(\omega-\omega_{\mathbf{k}}+i\varepsilon)}.\end{aligned} \tag{12}$$

Here, $\Gamma=\gamma_c+\gamma_R$ is the entire width of the nuclear energy level, formed by the conversion width,

$$\gamma_c=\frac{\hbar V}{4\pi^2}\int p^2dpd\Omega|V_{\mathbf{p}}|^2\delta(\omega-\omega_{\mathbf{p}}), \tag{13}$$

and by the radiative width,

$$\gamma_R=\frac{\hbar V}{4\pi^2}\int k^2dkd\Omega|V_{\mathbf{k},j}|^2\delta(\omega-\omega_{\mathbf{k}}), \tag{14}$$

where V is the volume of the whole system. While evolving Eqs. (13) and (14), the standard transition from summation to integration was used. We note that in Eq. (12), the frequency of nuclear transition is also renormalized (see Ref. 21), but for simplicity of presentation, we have left the old symbol.

The synchrotron radiation is propagating in the strictly defined direction, which we will identify with the z axis of the coordinate system. In the disordered crystalline systems, only the forward scattering (in which the phases of all scattered waves coincide) is coherent. Therefore, regarding the forward scattering, we leave in the sum of Eq. (12) only the terms related to $\mathbf{k}$, which are directed along the z axis. Only the projections of nuclear coordinates on the z axis are then essential. We also take into account that the synchrotron radiation is monochromatized before it interacts with the nuclear target. Since the potential of interaction weakly depends on k within the width of the frequency spectrum $\Delta\omega_p$ of the monochromatic radiation and, hence, the amplitude $A_{\mathbf{k}}(\omega)$ can be approximated with high accuracy by a constant, Eq. (12) is transformed to the form

$$\begin{aligned}\left(\omega-\omega_j+\frac{i\Gamma}{2\hbar}\right)B_m(\omega)=&F_0\exp(ik_0z)+\sum_{m'\neq m}B_{m'}(\omega)\sum_k|V_{k,j}|^2\\&\times\frac{\exp(ik[z_m-z_{m'}])}{(\omega-\omega_k+i\varepsilon)},\end{aligned} \tag{15}$$

where k_0 is the circular wave number, lying at the center of the considered frequency interval, and

$$F_0=A\frac{L\Delta\omega_pV_{k_0,j}}{2\pi c}. \tag{16}$$

Here, L is the size of the whole system along the z axis (instead of V). Passing from the sum to integral in the second term of the right hand part of Eq. (15) and calculating the integral by the residue theorem, we finally obtain

$$\begin{aligned}\left(\omega-\omega_j+\frac{i\Gamma}{2\hbar}\right)B_m(\omega)=&F_0\exp(ik_0z)-i\frac{\gamma_R}{2\hbar}\sum_{m'<m}B_{m'}(\omega)\\&\times\exp(ik[z_m-z_{m'}]).\end{aligned} \tag{17}$$

Here the summation over m' is performed for those nuclei, for which $z_{m'}$ is less than z_m, $k=\omega/c$, and γ_R is the radiative width of the level having, in the regarded approximation, the form

$$\gamma_R=\hbar\frac{2L}{c}|V_{k_0,j}|^2. \tag{18}$$

Equation (17) was obtained in Ref. 20. To solve this equation, the authors had applied a simple model of the effective planes. In this model, the real scattering target was represented by a finite number of planes, fixed perpendicular to the beam direction, where only a photon can be scattered. All resonant nuclei were located on those planes. The number of planes for a particular thickness of target was equal to N, and the average distance between these planes essentially exceeded the interatomic distances. The solution for such a model led to formula (2), having the form of sum with a

finite number of terms. In the next section, we perform another solution which will result exactly in the formula, Eq. (1), obtained earlier.

III. SOLUTION OF THE EQUATIONS

First of all, we note that the scattering amplitude for the NFS is small. This inevitably leads us to a conclusion that photons can move through a large distance in the target without scattering. Therefore, we can neglect the discreteness of matter at the atomic scale with high accuracy. The same approximation is applied in the theory of x-ray diffraction. On the other hand, even at a low concentration of nuclei, an excited nucleus can occupy a position in target having any coordinate. One can speak only about the average distance between the excited nuclei a, which is definitely much more than interatomic distances. That is why we can pass to the continuous approximation and substitute the sum over m in Eq. (17) by integral over z with the weight dz/a. We note that just the model of nuclear continuum was used in Ref. 7, where the quantum theory of radiation was employed for the case of the Mössbauer source. In this model, Eq. (17) is transformed to the integral form

$$B(z,\omega) = F(\omega)e^{ik_0 z} + G(\omega)e^{ikz}\int_0^z dz' B(z',\omega)e^{-ikz'}, \quad (19)$$

where

$$F(\omega) = \frac{F_0}{(\omega - \omega_j + i\Gamma/2\hbar)}, \quad G(\omega) = \frac{1}{a}\frac{(-i\gamma_R/2\hbar)}{(\omega - \omega_j + i\Gamma/2\hbar)}. \quad (20)$$

Here, $B(z,\omega)$ has a sense of the excitation amplitude for the nuclei with coordinate z. The latter is counted off from the front surface of the target.

Further, we take into account that k_0 actually corresponds to the resonant frequency, i.e., it is equal to ω_j/c, and $k = \omega/c$. The frequencies near the resonance are of our interest. Taking into account very small width of the nuclear resonance, the difference between k and k_0 in the phase factors can be neglected without loss of numerical accuracy. In this approximation, the solution has the form

$$B(z,\omega) = F(\omega)\exp(ik_0 z + G(\omega)z). \quad (21)$$

Next, we must calculate the coefficients $C_k(\omega)$ in accord with Eq. (10). In the considered approximation, we have

$$C_k(\omega) = H_k(\omega)\int_0^z dz' B(z',\omega)\exp(-ikz'),$$

$$H_k(\omega) = \frac{V_{k_0}^*}{a(\omega - \omega_k + i\varepsilon)}. \quad (22)$$

According to Eq. (8), the amplitudes for emission of photon in the time domain are

$$c_k(t) = \frac{i}{2\pi}\int d\omega C_k(\omega)\exp(i(\omega_k - \omega)t). \quad (23)$$

Finally, according to Eq. (8), the general amplitude for emission of the secondary photons is

$$\Psi_{scat}(t) = \frac{L}{2\pi c}\int d\omega_k c_k(t)\exp(-i\omega_k t). \quad (24)$$

We substitute Eq. (21) in Eq. (22) and calculate the integral in the same approximation, as earlier, i.e., replacing k by k_0 in the phase factors. As a result, we obtain

$$C_k(\omega) = \frac{aU}{V_{k_0,j}^*}H_k(\omega)[\exp(G(\omega)z) - 1]. \quad (25)$$

where U is the normalization factor,

$$U = \frac{V_{k_0}^*}{a}\frac{F(\omega)}{G(\omega)} = i\hbar A\frac{L\Delta\omega_p|V_{k_0,j}|^2}{\pi c\gamma_R} = iA\frac{\Delta\omega_p}{2\pi}. \quad (26)$$

Substituting now Eq. (25) in Eq. (23) and calculating the integral by its residue in $H_k(\omega)$, we easily arrive at

$$c_k(t) = U[\exp(G(\omega_k)z) - 1]. \quad (27)$$

Now, substituting Eq. (27) in Eq. (24) and replacing the variable, we finally obtain the solution in the form

$$\Psi_{scat}(t) = \frac{LU}{2\pi c}\int d\omega \exp(-i\omega t)[\exp(G(\omega)z) - 1]. \quad (28)$$

The obtained integral coincide exactly with that considered in Ref. 8; so, we can write down the solution immediately (see derivation in Ref. 8) as

$$\Psi_{scat}(t) = -\frac{LU}{c}\theta(t)\exp\left(-i\omega_j t - \frac{\tau}{2}\right)\frac{1}{t_0}\left(\frac{\xi}{4\tau}\right)J_1([\xi\tau]^{1/2}), \quad (29)$$

where

$$\tau = \frac{t}{t_0}, \quad t_0 = \frac{\hbar}{\Gamma}, \quad \xi = \frac{2\gamma_R}{a\Gamma}z = \mu_n z, \quad \mu_n = N_0\sigma_0\eta f. \quad (30)$$

Here, we introduced the parameters used in Eq. (1). From comparison of the two obtained results, we arrive at the expression for parameter a,

$$a = \frac{2}{\mu_n}\frac{\gamma_R}{\Gamma} = \frac{2}{N_0\sigma\eta f}, \quad (31)$$

where $\sigma = \sigma_0\Gamma/\gamma_R$ is the total cross section. It is of interest to note that parameter N from the theory of Ref. 20 equals

$$N = \frac{\mu_n z\Gamma}{2\gamma_R} = \frac{z}{a}. \quad (32)$$

Thus, the relation between N and a is rather simple.

IV. DISCUSSION

We have shown that the analytical solution (1) for the time dependence of the NFS of synchrotron radiation pulse,

well known for decades, can also be derived within the second approach based on the Heitler's quantum theory of radiation. In our derivation, we apply standard approximations in which we neglect a discreteness of matter at the atomic scale but allow nucleus to have any coordinate in the target. Formula (2) by Hoy *et al.*[20] was obtained for quite different model of nuclear arrangement, namely, where the nuclei are grouped in the "effective planes" (EPs). As a consequence, they cannot have an arbitrary coordinate in the target. The number of planes N is related to the target thickness. There is a serious physical problem with the parameter N. Because of being integer, it cannot be applied to the target of an arbitrary thickness z. The authors proposed determining the value of N from the experiment or from the comparison with the other theories.

Both variants are unacceptable. In the first variant, the theory becomes incomplete whereas the theory of x-ray nuclear forward scattering and result (1) for NFS are complete. In the second variant, as it is shown above, we have $z=Na$ and only discrete set of z values can be considered. It is not a severe problem if $z \gg a$ and $N \gg 1$. We can show analytically that for large N, both formulas (1) and (2) gives the same numerical value, and a difference between them goes to zero for infinite N. Physically, it is obvious because the discreteness of the EP model becomes not so significant for large N. This fact was claimed in Ref. 20 and earlier in Ref. 22 on the basis of numerical calculations. However, for small target thickness, the EP model by Hoy *et al.* is unclear.

Indeed, let us consider the case of one effective plane $N=1$. This case is realized if $\mu_n z < 2/(1+\alpha)$, where α is the conversion coefficient for the nuclear transition. In the case of ^{57}Fe nuclei, $\alpha=8.19$, and we come to the condition $z < 0.22/\mu_n$. According to the theory of Hoy *et al.* for nuclear target, having the thickness within this interval, the response is purely exponential, i.e., the same as for one nucleus. It is an effect of the model where all nuclei have the common coordinate that of the effective plane. On the other hand, it is clear from the physical point of view that the multiple scattering is possible for target of any thickness because the process is spontaneous and we deal only with its probability. In reality, the nuclei can have arbitrary coordinate in the target, and the second nucleus at the coordinate z_2 can scatter the photon which was scattered previously by the first nucleus at the coordinate z_1 with $z_1 < z_2 < 0.22/\mu_n$. The probability of this process will be small but not zero. Equation (1) allows these processes. That is why the old solution, known since 1979, is correct, whereas the new solution, proposed in 2001, is only approximation to it.

The authors of Ref. 20 insist on the exclusive merits of their presentation. In particular, they show that the obtained solution allows one to throw light on the nature of the dynamics of interaction of a quantum with the nuclear system in the direction of its propagation through the target and to show in what way the set of the different scattering paths specifies the observed time dependence of the scattering intensity. As known, the characteristic features of this dependence is a speedup scattering of a quantum at the first stage, which transforms at the next stages to the aperiodic beating of the scattering intensity. Let us consider this question in more detail.

It is seen from expression (2) that each next term of the sum changes its sign. The sign changes each time when the number of scattering planes increases by 1. As well known, the change of sign is caused by the phase shift $-\pi$ of the plane wave scattered coherently by a single nucleus. One fraction of the phase shift $-\pi/2$ arises due to scattering at resonance. The other fraction is the result of interference of the wavelets scattered by centers located on a plane (Fresnel interference). Thus, the partial waves corresponding to the onefold and twofold scattering are in the opposite phase. Hence, the sum over the two paths decays faster than that determined by the exponential law. The speedup decay just results from here. If the multifold nuclear resonant scattering can occur, the sum over all possible paths results in the appearance of nodes and antinodes in the scattered wave. Such a modulation of the wave amplitude provides the beating of the forward scattered intensity.

It should be noted, however, that the mentioned physical interpretation is not a privilege of expression (2). To show this, we perform the series expansion of formula (1). We write down this formula in the form of power series using the well known expansion of the Bessel function

$$J_1(z) = \frac{z}{2}\sum_{n=0}^{\infty}(-1)^n \frac{(z/2)^{2n}}{n!(n+1)!}. \tag{33}$$

The immediate calculation results in

$$E(z,t) = -E_{\omega_j}(z)\frac{\mu_n z}{4t_0}\exp\left(-i\omega_j t - \frac{t}{2t_0}\right) \times\left[1+\sum_{n=1}^{\infty}\frac{(-1)^n}{n!(n+1)!}\left(\frac{\mu_n z t}{4t_0}\right)^n\right]. \tag{34}$$

Comparison of formulas (2) and (34) reveals close similarity of their structures. The exponential time dependence in front of the brackets is the same in both cases. Besides that, the sum in both cases contains the sign-changing terms. Moreover, in the limit $N\to\infty$, even coefficients becomes numerically identical. Thus, one can say that each next $n+1$ term in expansion (34) corresponds to the contribution of the path, where scattering happened sequentially $n+1$ times. The infinite number of paths of different multiplicities contributes to the total amplitude in expansion (34), but the contribution of the multifold scattering drops down with rising n.

Thus, the physical picture of scattering over paths results from this solution as well. However, there is a specificity of solution (34): it contains the contributions of the unlimited number of scattering paths. Having such a structure, expansion (34) and its limit, formula (1), describe the time-dependent scattering from the nuclear target of any thickness.

Finally, we consider the question concerning the real multiple scattering of a photon by a nuclear target. We have shown that the terms of the expansion of the analytical solution, obtained in the theory,[8] also represent the contributions of the partial scattering paths related to the different multiplicities of scattering. Obviously, these are the virtual processes, and only their superposition can have a physical sense. The real multiple scattering is revealed by the space-

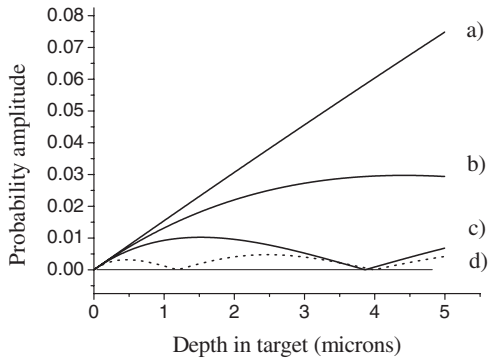

FIG. 1. The distributions of the probability amplitude of finding a photon inside the target at different delay times: (a) 1 ns, (b) 20 ns, (c) 60 ns, and (d) 200 ns. The calculations are performed in accord with formula (1), where the exponential factor is omitted.

time structure of the total amplitude of the scattered radiation (1), which is contributed by all paths of the virtual multiple scattering. The function $J_1(\sqrt{u})/\sqrt{u}$ determines the modulation of the photon wave packet in space and in time. It has zeros at values of the dimensionless argument $u=\mu_n z t/t_0 = 14.8, 49.2, \ldots$, i.e., the packet contains nodes and antinodes following each other. Such a pattern can be imagined as traveling in space train, consisting of the bright and dark zones. Figure 1 displays the amplitudes of the wave packet inside the target at different times after the arrival of the SR pulse (the value $\mu_n = 8.8\ \mu\text{m}^{-1}$ was used for calculations). At the first instant (1 ns), the amplitude is growing nearly linearly with the thickness directly reflecting the coherent enhancement, which takes place in the scattering process. Later on, the amplitude of finding a photon is decreasing since the probability for it to be absorbed by a nucleus is rising. Already at 60 ns, the probability of finding a photon at the depth near 4 μm drops down to zero because it is definitely absorbed by a nucleus at this depth. The amplitude at 200 ns reveals the two events of the photon absorption depths of 1.2 and 4 μm.

A definite space-time distribution of the nuclear excitation amplitude corresponds to such structure of the field inside the target. One can show (see, e.g., Fig. 7 in Ref. 23) that antinodes of the excitation amplitude correspond to nodes of the field and vice versa. This picture reflects the dynamics of "pumping" of the electromagnetic energy from the radiation field to the nuclear system and backward while γ quantum propagates in the target. The space-time modulation of both the nuclear excitations and the propagating field reveals sequential processes of absorption and re-emission of radiation by nuclei. Correspondingly, the number of the intensity beats of radiation propagated through the target is related to the number of scattering events in the target and, therefore, exhibits the real multiplicity of scattering.

In conclusion, we have shown that an adequate solution of the problem of the nuclear forward scattering of synchrotron radiation within the scope of the Heitler's quantum theory of radiation leads to the well known and correct formula, which was obtained 30 years ago.[8] The solution obtained by Hoy *et al.*[20] is incorrect for small sample thickness because it artificially limits the number of multiple scattering paths in nuclear target of a particular thickness.

ACKNOWLEDGMENTS

The work of V.G.K. was supported by RFBR Grant No. 07-02-00067a and RS Grant No. 6869.2006.2.

*URL: http://kohnvict.narod.ru
†g.smirnov@gmx.net

[1] F. J. Lynch, R. E. Holland, and M. Hamermesh, Phys. Rev. **120**, 513 (1960).
[2] *Nuclear Resonant Scattering of Synchrotron Radiation*, edited by E. Gerdau and H. de Waard, (Baltzer, Bussum, 2000), vols. 123-125 of Hyperfine Interact.
[3] G. V. Smirnov, Bull. Russ. Acad. Sci. Phys. **67**, 984 (2003).
[4] A. Autier, *Dynamical Theory of X-ray Diffraction*, 1st ed. (Oxford University Press, New York, 2001).
[5] W. Heitler, *The Quantum Theory of Radiation*, 3rd ed. (Oxford University Press, New York, 1957).
[6] A. I. Akhiezer and V. B. Berestetskij, *Quantum Electrodynamics* (Interscience, New York 1965); *Kvantovaja Elektrodinamika*, (Nauka, Moscow, 1969).
[7] S. M. Harris, Phys. Rev. **124**, 1178 (1961).
[8] Yu. Kagan, A. M. Afanasev, and V. G. Kohn, J. Phys. C **12**, 615 (1979).
[9] Yu. Kagan, Hyperfine Interact. **123/124**, 83 (1999).
[10] V. G. Kohn and Yu. V. Shvyd'ko, J. Phys.: Condens. Matter **7**, 7589 (1995).
[11] G. V. Smirnov and W. Potzel, Hyperfine Interact. **123/124**, 633 (1999).
[12] Yu. V. Shvyd'ko, A. I. Chumakov, G. V. Smirnov, V. G. Kohn, T. Hertrich, U. van Burck, E. Gerdau, H. D. Ruter, and J. Metge, Europhys. Lett. **22**, 305 (1993).
[13] G. V. Smirnov and V. G. Kohn, Phys. Rev. B **52**, 3356 (1995).
[14] V. G. Kohn and G. V. Smirnov, Phys. Rev. B **57**, 5788 (1998).
[15] G. V. Smirnov, V. G. Kohn, and W. Petry, Phys. Rev. B **63**, 144303 (2001).
[16] Yu. V. Shvyd'ko, T. Hertrich, U. van Burck, E. Gerdau, O. Leupold, J. Metge, H. D. Ruter, S. Schwendy, G. V. Smirnov, W. Potzel, andP. Schindelmann, Phys. Rev. Lett. **77**, 3232 (1997).
[17] Yu. V. Shvyd'ko, Phys. Rev. B **59**, 9132 (1999).
[18] J. P. Hannon and G. T. Trammell, Hyperfine Interact. **123/124**, 127 (1999).
[19] G. T. Trammell and J. P. Hannon, Phys. Rev. B **18**, 165 (1978); **19**, 3835 (1979).
[20] G. R. Hoy, J. Odeurs, and R. Coussement, Phys. Rev. B **63**, 184435 (2001).
[21] Yu. Kagan and A. M. Afanas'ev, Zh. Eksp. Teor. Fiz. **50**, 271 (1966) [Sov. Phys. JETP **23**, 178 (1966)].
[22] G. R. Hoy, J. Phys.: Condens. Matter **9**, 8749 (1997).
[23] G. V. Smirnov, Hyperfine Interact. **123/124**, 31 (1999).