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Nuclear resonant forward scattering of x rays: Time and space picture

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The problem of forward resonant scattering of x rays by an ensemble of nuclei is being solved directly in time and space. The wave equation describing the propagation of the radiation through the nuclear ensemble is derived. It is a first-order integrodifferential equation. Its kernel is a double time function $K(t,\tilde{t})$ that represents a coherent single scattering response of the nuclear system at time t to excitation at $\tilde{t}$. The explicit form of the kernel is defined by the character of interactions, the nuclei experience with the environment and by the character of their spatial motion. A general procedure of the solution of the wave equation is introduced that is independent of the type of the kernel. Examples for various kernels are presented and discussed for some particular cases: collective or diffusive motion of nuclei in space, thermal lattice vibrations, time-independent hyperfine interactions, and time-dependent hyperfine interactions due to atomic spin fluctuations or external magnetic-field switching. [S0163-1829(99)09113-4]

I. INTRODUCTION

Time spectroscopy of nuclear forward scattering (NFS) of synchrotron radiation is an experimental technique complementary to Mössbauer absorption spectroscopy in the energy domain providing similar, additional or even unique information (for a review see, e.g., Refs. 1 and 2). Its uniqueness arises from the fact that NFS is spatially coherent. Interference effects in scattering play a significant role and enable to reveal information inaccessible to absorption spectroscopy.

The first theory of the time dependence of NFS of synchrotron radiation was developed by Kagan, Afanas'ev, and Kohn.[3] Fourier transformation of the frequency dependence of the transmission amplitude through a single resonance absorber was used to calculate the time spectrum of NFS. Important features that arise in thick samples—speed-up of the nuclear decay and dynamic beat—were considered. Trammell and Hannon[4] pointed to a simple but important effect that arises in time spectra from samples possessing multiple nuclear resonances—quantum beat. Numerous subsequent theoretical papers[5–8] have used Fourier transformation from the frequency to the time domain to consider more complicated cases of interactions of nuclei with their environment. Fourier transformation techniques can be used provided the frequency spectrum of transmission through the given sample is known.

An approach, which does not require the initial knowledge of the transmission frequency spectrum, is based on the solution of the scattering problem directly in time and space. This approach was used to handle NFS in particular cases of both time-independent[9] and time-dependent[10–14] interactions.

In the present paper this approach is generalized to arbitrary type of interactions, the nuclei experience with their environment, and arbitrary character of their spatial motion. A wave equation is derived for the propagation of x rays directly in time and space through the nuclear resonant medium, Secs. II and IV. The equation is a first-order integrodifferential wave equation. Its kernel is the double time nuclear self-correlation function $K(t,\tilde{t})$ defined by the type of interactions the nuclei experience with the environment and their spatial motion, Sec. III. $K(t,\tilde{t})$ describes a coherent single scattering response in the forward direction of the nuclear system at time t to excitation at $\tilde{t}$. A general procedure is introduced of solving the wave equation independent of the type of the kernel, Sec. IV B. Examples of calculations are given of the nuclear self-correlation functions and time spectra of NFS in single scattering approximation for the following particular cases: collective motion of the nuclear ensemble in space, Sec. V A 1; diffusive nuclear motion, Sec. V A 2; thermal lattice vibrations, Sec. V A 3; time-independent hyperfine interactions, Sec. V B; time-dependent hyperfine interactions due to atomic spin fluctuations, Sec. V C; or due to external magnetic field switching, Sec. V D.

II. FORMULATION OF THE PROBLEM

We will evaluate the time dependence of coherent scattering of x rays from an ensemble of resonant nuclei in the direction of the incident beam—nuclear forward scattering. The nuclei are supposed to be moving in space, or (and) located in an environment with hyperfine interactions which, generally speaking, are time dependent. The motion in space may be a result of an external force, or thermal lattice vibrations, or diffusion. Nuclear spin relaxation in a fluctuating environment or time-dependent external perturbations of hyperfine fields can be mentioned as examples of time-dependent hyperfine interactions. The purpose of the paper is to derive directly in time and space the wave equation of propagation of radiation in such media and to develop a general procedure of its solution.

The electric component $\boldsymbol{e}(\boldsymbol{r},t)$ of the radiation field propagating in the resonant medium we calculate by using Maxwell's wave equation

$$\nabla^2\boldsymbol{e}-\mathrm{grad\,div}\,\boldsymbol{e}-\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\boldsymbol{e}=\frac{4\pi}{c^2}\frac{\partial}{\partial t}\boldsymbol{i} \qquad (1)$$

with $\boldsymbol{i}(\boldsymbol{r},t)$ being the macroscopic current density induced by the radiation in the system of resonant nuclei. The sample is assumed to be a plate of thickness L. The reference system is attached to its entrance surface with the z axis directed perpendicular and inwards to the plate. The incident radiation is represented as a plane wave modulated in time,

$$\boldsymbol{e}_{\text{in}}(\boldsymbol{r},t)=\boldsymbol{\mathcal{E}}(t)\,e^{i(\tilde{\boldsymbol{k}}\boldsymbol{r}-\tilde{\omega}t)},\tag{2}$$

with carrier frequency $\tilde{\omega}$ and wave vector $\tilde{\boldsymbol{k}}\,(\tilde{k}=\tilde{\omega}/c)$. The carrier frequency is assumed to be close to the nuclear resonance frequency $E_0/\hbar$.

The solution of Eq. (1) for the field propagating in the sample in the primary beam direction is sought in the form

$$\boldsymbol{e}(\boldsymbol{r},t)=\boldsymbol{E}(z,t)e^{i(\tilde{\boldsymbol{k}}\boldsymbol{r}-\tilde{\omega}t)}\tag{3}$$

with $\boldsymbol{E}(z,t)$ being an envelope that is varying slowly in time and space compared to the exponent. The envelope contains only the spatial coordinate z, since refraction and absorption in the sample occur along the normal to the sample surface, i.e., along z. The induced current density is sought in the same form

$$\boldsymbol{i}(\boldsymbol{r},t)=\boldsymbol{I}(z,t)e^{i(\tilde{\boldsymbol{k}}\boldsymbol{r}-\tilde{\omega}t)}.\tag{4}$$

The presentation by Eqs. (3) and (4) is known as the slowly varying envelope approximation. In this case the second-order Maxwell's wave equation reduces to the first-order differential equation for the envelopes $\boldsymbol{E}(z,t)$ and $\boldsymbol{I}(z,t)$:

$$\sin\Theta\,\frac{\partial}{\partial z}\boldsymbol{E}(z,t)=-\frac{2\pi}{c}\boldsymbol{I}(z,t)\tag{5}$$

with Θ being the angle of incidence ($\sin\Theta=\tilde{k}_z/\tilde{k}$). By using Eq. (2) the boundary conditions for Eq. (5) read as follows:

$$\boldsymbol{E}(0,t)=\boldsymbol{\mathcal{E}}(t).\tag{6}$$

Wave equations for the slowly varying envelopes of the radiation field and induced current density were first introduced in optics (see, e.g., Ref. 15) and in x-ray diffraction in crystals.[16] Recently it was also used for the analysis of nuclear resonance scattering problems.[10–12]

The time spectrum of nuclear forward scattering is the time dependence of the radiation intensity emerging from the sample in the primary beam direction:

$$S(t)\propto|\boldsymbol{E}(L,t)|^2=\sum_s |E^s(L,t)|^2.\tag{7}$$

Here the superscript s corresponds to any of two orthogonal polarization components of the radiation, given by the polarization vectors $\boldsymbol{e}^s$.

III. NUCLEAR CURRENTS AND SELF-CORRELATION FUNCTION

A. Current density of the nuclear ensemble

In each particular case the right-hand side of Eq. (5), i.e., the current density induced by the radiation in the ensemble of resonant nuclei, should be specified. The current density of the ensemble is the sum of the current densities $\boldsymbol{i}_\alpha(\boldsymbol{r}-\boldsymbol{r}_\alpha(t),t)$ of individual nuclei numbered here by α. The position vector $\boldsymbol{r}_\alpha(t)=\boldsymbol{R}_\alpha+\boldsymbol{u}_\alpha(t)$ of the nucleus α is presented as a sum of its equilibrium position $\boldsymbol{R}_\alpha$ and its displacement $\boldsymbol{u}_\alpha(t)$ from the equilibrium due to either thermal lattice vibrations, or (and) diffusion, or (and) vibrations induced by an external force, etc. For convenience of further calculations the presentation of the nuclear current density in terms of momentum $\boldsymbol{k}$ is used: $\boldsymbol{i}_\alpha(\boldsymbol{k},t)$. The coherent current density of the nuclear ensemble is then given by

$$\boldsymbol{i}(\boldsymbol{r},t)=\sum_\alpha \int \frac{d\boldsymbol{k}}{(2\pi)^3}\langle \boldsymbol{i}_\alpha(\boldsymbol{k},t)\,e^{i\boldsymbol{k}(\boldsymbol{r}-\boldsymbol{r}_\alpha)}\rangle.\tag{8}$$

$\langle\cdots\rangle$ means statistical average over the initial state of the crystal, assuming its thermal equilibrium, and also averaging over all other stochastic degrees of freedom. The coherent part of the current density of an individual nucleus α is calculated by using its density matrix:

$$\boldsymbol{i}_\alpha(\boldsymbol{k},t)=\text{Tr}\{\hat{\boldsymbol{j}}(\boldsymbol{k})\,\hat{\rho}_\alpha(t)\}.\tag{9}$$

Here $\hat{\rho}_\alpha(t)$ is the density matrix operator of the nucleus, which is defined by the Liouville–von Neumann equation

$$i\hbar\,\frac{\partial\hat{\rho}_\alpha(t)}{\partial t}=[\hat{\mathcal{H}}_\alpha(t)+\hat{\mathcal{H}}^{\gamma}_\alpha(t)\,,\,\hat{\rho}_\alpha(t)].\tag{10}$$

$\hat{\mathcal{H}}_\alpha(t)$ in Eq. (10) is the Hamiltonian of a nucleus α with the following components:

$$\hat{\mathcal{H}}_\alpha(t)=E_\lambda-i\frac{\Gamma_0}{2}\,\delta_{\lambda,e}+\hat{\mathcal{H}}^{\text{hf}}_{\alpha_\lambda}(t).\tag{11}$$

E_λ is the energy in the ground ($\lambda=g$) or in the excited ($\lambda=e$) state defined by internal nuclear interactions. Each nucleus is also characterized by the energy $E_0=E_e-E_g$ of the nuclear transition $e\Rightarrow g$, by the full natural energy width Γ_0 of the nuclear excited state, by its spin $\boldsymbol{J}_\lambda$, magnetic moment $\boldsymbol{\mu}_\lambda$, quadrupole moment Q, etc. The $\hat{\mathcal{H}}^{\text{hf}}_{\alpha_\lambda}(t)$ represents hyperfine interactions which are generally speaking time dependent. Its actual type will be specified later.

The $\hat{\mathcal{H}}^{\gamma}_\alpha(t)$ in Eq. (10) is the Hamiltonian representing the interaction of the nucleus with the radiation field. It is given by the standard expression $\hat{\mathcal{H}}^{\gamma}_\alpha(t)=-c^{-1}\int d\boldsymbol{r}\,\hat{\boldsymbol{j}}(\boldsymbol{r}-\boldsymbol{r}_\alpha)\,\boldsymbol{A}(\boldsymbol{r},t)$ (Ref. 17) with $\boldsymbol{A}(\boldsymbol{r},t)$ being the vector potential and with $\hat{\boldsymbol{j}}(\boldsymbol{r}-\boldsymbol{r}_\alpha)$ here and $\hat{\boldsymbol{j}}(\boldsymbol{k})$ in Eq. (9) being the Schrödinger picture nuclear current-density operator in the real and reciprocal space, respectively. By using the Coulomb gauge with zero scalar potential, the representation of the field $\boldsymbol{e}(\boldsymbol{r},t)$ by Eq. (3) and applying the slowly varying envelope approximation we obtain for the interaction Hamiltonian[10,11]

$$\hat{\mathcal{H}}^{\gamma}_\alpha(t)=i\tilde{\omega}^{-1}\hat{\boldsymbol{j}}(-\tilde{\boldsymbol{k}})\,\boldsymbol{E}(z_\alpha,t)\,e^{i(\tilde{\boldsymbol{k}}\boldsymbol{r}_\alpha-\tilde{\omega}t)}.\tag{12}$$

We assume that the interaction (12) of the nuclei with the radiation field is weak compared to the other interactions given by Eq. (11). This allows us to use perturbation theory and to obtain in its second order (resonant scattering is a second-order scattering process) the following expression for the current density,[18]

$$\boldsymbol{i}_\alpha(\boldsymbol{k},t)=\frac{1}{i\hbar}\mathrm{Tr}\left\{\check{\boldsymbol{j}}_\alpha(\boldsymbol{k},t)\int_{-\infty}^{t}[\check{\mathcal{H}}_\alpha^{\gamma}(\tilde{t}),\check{\rho}_\alpha(-\infty)]\,d\tilde{t}\right\}. \tag{13}$$

Here the symbols with the inverted caret ˇ denote interaction picture operators defined according to the rule

$$\check{\Lambda}_\alpha(t)=\hat{U}_\alpha^{-1}(t,-\infty)\,\hat{\Lambda}_\alpha(t)\,\hat{U}_\alpha(t,-\infty), \tag{14}$$

with

$$\hat{U}_\alpha(t_2,t_1)=\hat{T}\exp\left\{-\frac{i}{\hbar}\int_{t_1}^{t_2}\hat{\mathcal{H}}_\alpha(t)\,dt\right\} \tag{15}$$

being the evolution operator of a nucleus α in the absence of the radiation field, and $\hat{T}$ being the time-ordering operator.

The trace in Eq. (13) is calculated by using a full set of nuclear state vectors $|\alpha_\lambda\rangle$ that in the present paper are chosen as eigenstate vectors of the nuclear Hamiltonian (11) at $t=-\infty:\hat{\mathcal{H}}_\alpha(-\infty)$. It is assumed that initially at $t=-\infty$ all the sublevels in the ground state are populated equally, i.e., $\langle\alpha_g|\hat{\rho}_\alpha(-\infty)|\alpha_g\rangle=1/(2I_g+1)$, while those in the excited state are not populated at all, i.e., $\langle\alpha_e|\hat{\rho}_\alpha(-\infty)|\alpha_e\rangle=0$. It is also taken into account that only matrix elements of the operator $\hat{\boldsymbol{j}}$ between the ground and excited nuclear states have significant values. After transformations we obtain the following general expression for the s-polarization component of the current density of an individual nucleus

$$i_\alpha^{s}(\boldsymbol{k},t)=\frac{1}{\hbar\tilde{\omega}(2I_g+1)}\sum_{\alpha_g,\alpha_e,\tilde{s}}j_{\alpha_g\alpha_e}^{s}(\boldsymbol{k},t)\times\int_{-\infty}^{t}j_{\alpha_e\alpha_g}^{\tilde{s}}(-\tilde{\boldsymbol{k}},\tilde{t})\,E^{\tilde{s}}(z_\alpha,\tilde{t})e^{i(\tilde{\boldsymbol{k}}\boldsymbol{r}_\alpha-\tilde{\omega}\tilde{t})}\,d\tilde{t}. \tag{16}$$

The two matrix elements $j_{\alpha_g\alpha_e}^{s}(\boldsymbol{k},t)$ and $j_{\alpha_e\alpha_g}^{\tilde{s}}(-\tilde{\boldsymbol{k}},\tilde{t})$ in Eq. (16) represent two stages of the nuclear resonant scattering process via one of the possible nuclear transitions $\langle\alpha_g|\Leftrightarrow|\alpha_e\rangle$. The matrix element $j_{\alpha_e\alpha_g}^{\tilde{s}}(-\tilde{\boldsymbol{k}},\tilde{t})$ describes excitation of the nucleus at time instant $\tilde{t}$ with absorption of the radiation field in the polarization state $\tilde{s}$ and with the wave vector $\tilde{\boldsymbol{k}}$, while $j_{\alpha_g\alpha_e}^{s}(\boldsymbol{k},t)$ describes deexcitation of the nucleus at time instant t with the emission of the radiation in the polarization state s and with wave vector $\boldsymbol{k}$. The matrix elements $j_{\alpha_g\alpha_e}^{s}(\boldsymbol{k},t)$ and $j_{\alpha_e\alpha_g}^{\tilde{s}}(-\tilde{\boldsymbol{k}},\tilde{t})$ are calculated according to Eqs. (14) and (15), in particular:

$$j_{\alpha_g\alpha_e}^{s}(\boldsymbol{k},t)=\sum_{\tilde{\alpha}_g\tilde{\alpha}_e}U_{\alpha_g\tilde{\alpha}_g}^{-1}(t,-\infty)\,j_{\tilde{\alpha}_g\tilde{\alpha}_e}^{s}(\boldsymbol{k})\,U_{\tilde{\alpha}_e\alpha_e}(t,-\infty). \tag{17}$$

For the calculation of the nuclear current-density matrix elements $j_{\alpha_g\alpha_e}^{s}(\boldsymbol{k})$ we refer to textbooks[17,19] and papers.[20–22,5]

Inserting Eq. (16) into Eq. (8) we obtain the following expression for the s component of the coherent current-density induced in the nuclear ensemble:

$$i^{s}(\boldsymbol{r},t)=\int\frac{d\boldsymbol{k}}{(2\pi)^3}e^{i(\boldsymbol{k}\boldsymbol{r}-\tilde{\omega}t)}\sum_{\alpha,\tilde{s}}e^{i(\tilde{\boldsymbol{k}}-\boldsymbol{k})\boldsymbol{R}_\alpha}\times\int_{-\infty}^{t}F_\alpha^{s\tilde{s}}(\boldsymbol{k},\tilde{\boldsymbol{k}},t,\tilde{t})E^{\tilde{s}}(z_\alpha,\tilde{t})d\tilde{t}, \tag{18}$$

where $F_\alpha^{s\tilde{s}}(\boldsymbol{k},\tilde{\boldsymbol{k}},t,\tilde{t})$ is defined as

$$F_\alpha^{s\tilde{s}}(\boldsymbol{k},\tilde{\boldsymbol{k}},t,\tilde{t})=\frac{1}{\hbar\tilde{\omega}(2I_g+1)}\times\langle e^{-i\boldsymbol{k}\boldsymbol{u}_\alpha(t)}e^{i\tilde{\boldsymbol{k}}\boldsymbol{u}_\alpha(\tilde{t})}\rangle e^{i\tilde{\omega}(t-\tilde{t})}\times\sum_{\alpha_g,\alpha_e}\langle j_{\alpha_g\alpha_e}^{s}(\boldsymbol{k},t)j_{\alpha_e\alpha_g}^{\tilde{s}}(-\tilde{\boldsymbol{k}},\tilde{t})\rangle. \tag{19}$$

It is clear from Eq. (18) as well as from Eqs. (13) and (16) that $\tilde{t}\leqslant t$, which has the evident meaning that the time of excitation $\tilde{t}$ always precedes the time of deexcitation t.

B. Self-correlation function

Let us combine into groups the terms in the sum Σ_α of Eq. (18) with the same values of $F_\alpha^{s\tilde{s}}(\boldsymbol{k},\tilde{\boldsymbol{k}},t,\tilde{t})$. These groups we will tag by index β. Physically this procedure means that we combine into groups resonant nuclei α with the same interactions with the environment and the same spatial motion. Groups of nuclei in equivalent sites of the crystal unit cell, or nuclei experiencing the same type of fluctuations of their atomic spins, or the same type of diffusion in a crystal lattice, etc., are examples of such groups. Thus in the sum over the whole nuclear ensemble in Eq. (18) we single out the sums Σ'_α over all nuclei within each group β. We assume that the number of nuclei within each group is macroscopic and make use of the relation

$$\sum_\alpha{}'\exp[i(\tilde{\boldsymbol{k}}-\boldsymbol{k})\boldsymbol{R}_\alpha]=(2\pi)^3N_0w_\beta\delta(\tilde{\boldsymbol{k}}-\boldsymbol{k}), \tag{20}$$

where N_0 is the number of all resonant nuclei per unit volume and w_β is the relative weight of the group β. We also assume that the sample is homogeneous in the (x,y) plane so that no effects of nuclear resonant small-angle scattering occur.[23] As a result we finally obtain the expression for the coherent part of the macroscopic nuclear current density. It has the form of Eq. (4) with the s component of the envelope of the macroscopic nuclear current density given by

$$I^{s}(z,t)=\frac{c}{2\pi}\left\{\frac{\sigma_R N_0\Gamma_0}{4\hbar}\right\}\int_{-\infty}^{t}\sum_{\tilde{s}}K^{s\tilde{s}}(t,\tilde{t})E^{\tilde{s}}(z,\tilde{t})d\tilde{t}. \tag{21}$$

The double-time function $K^{s\tilde{s}}(t,\tilde{t})$ is the self-correlation function of the nuclear ensemble, which is defined as

$$K^{s\tilde{s}}(t,\tilde{t})=\sum_\beta L_\beta^{s\tilde{s}}(t,\tilde{t})M_\beta(t,\tilde{t}), \tag{22}$$

$$L^{s\tilde{s}}_{\beta}(t,\tilde{t})=e^{i\tilde{\omega}(t-\tilde{t})}X_{\beta}\sum_{\beta_g,\beta_e}\langle j^{s}_{\beta_g\beta_e}(\tilde{\boldsymbol{k}},t)j^{\tilde{s}}_{\beta_e\beta_g}(-\tilde{\boldsymbol{k}},\tilde{t})\rangle, \tag{23}$$

$$M_{\beta}(t,\tilde{t})=\langle e^{-i\tilde{\boldsymbol{k}}\boldsymbol{u}_{\beta}(t)}e^{i\tilde{\boldsymbol{k}}\boldsymbol{u}_{\beta}(\tilde{t})}\rangle, \tag{24}$$

$$X_{\beta}=\frac{4\tilde{k}}{4c^2(2I_e+1)\Gamma_{\gamma}}w_{\beta}. \tag{25}$$

The factor

$$\sigma_R=\frac{4\pi}{\tilde{k}^2}\frac{2I_e+1}{2(2I_g+1)}\frac{\Gamma_{\gamma}}{\Gamma_0} \tag{26}$$

in Eq. (21) is the cross section of the nuclear resonant absorption. Γ_{γ} in Eq. (25) is the full radiative width of the nuclear transition $e\Rightarrow g$.

The function $L^{s\tilde{s}}_{\beta}(t,\tilde{t})$ in Eq. (23) can be presented in an alternative way:

$$L^{s\tilde{s}}_{\beta}(t,\tilde{t})=e^{i\tilde{\omega}(t-\tilde{t})}X_{\beta}\times\sum_{\beta_g\beta_e}\langle[U^{-1}_{\beta}(t,\tilde{t})j^{s}(\tilde{\boldsymbol{k}})U_{\beta}(t,\tilde{t})]_{\beta_g\beta_e}\rangle j^{\tilde{s}}_{\beta_e\beta_g}(-\tilde{\boldsymbol{k}}). \tag{27}$$

For this we have used Eqs. (17) and the composition law $\hat{U}(t_3,t_1)=\hat{U}(t_3,t_2)\hat{U}(t_2,t_1)$ valid for the evolution operators.[24]

The self-correlation function $K^{s\tilde{s}}(t,\tilde{t})$ has the property

$$K^{s\tilde{s}}(t,t)=\delta^{s\tilde{s}}, \tag{28}$$

which can be easily verified by taking into account that according to Eq. (24) $M_{\beta}(t,t)=1$ independent of how the nuclei are moving, that according to Eq. (15) $U_{\beta}(t,t)=1$ independent of the type of hyperfine interactions, and by the relation

$$\sum_{\beta_g,\beta_e}j^{s}_{\beta_g\beta_e}(\tilde{\boldsymbol{k}})\,j^{\tilde{s}}_{\beta_e\beta_g}(-\tilde{\boldsymbol{k}})=\delta^{s\tilde{s}}\frac{\Gamma_{\gamma}(2I_e+1)c^2}{4\tilde{k}}. \tag{29}$$

The latter can be proved by using the optical theorem.

The function $M_{\beta}(t,\tilde{t})$ in Eqs. (22) and (24) represents spatial motion of the nuclei belonging to a group β, while $L^{s\tilde{s}}_{\beta}(t,\tilde{t})$ in Eqs. (22) and (23) represents nuclear spin motion and other internal nuclear degrees of freedom.

The function $M_{\beta}(t,\tilde{t})$ is a kind of a self-correlation function introduced by Van Hove[25] in connection with neutron scattering. It was later used by Singwi and Sjölander[26] to build the theory of resonance absorption of nuclear γ rays for an arbitrary system of interacting particles.

The function $L^{s\tilde{s}}_{\beta}(t,\tilde{t})$ is similar to the self-correlation function used by Afanas'ev and Kagan[27] and Blume and Tjon[28] to evaluate the Mössbauer line shape in the presence of time-dependent hyperfine fields.

The self-correlation functions typically used have a $(t-\tilde{t})$ dependence. The function defined by Eqs. (22)–(24) contains both time variables separated. As shown in Sec. IV F the self-correlation functions $K(t,\tilde{t})$ that are reduced to $K_1(t-\tilde{t})$ describe coherent *elastic* scattering, while the self-correlation function $K(t,\tilde{t})$ with separated time variables describes coherent *inelastic* scattering.

The coherent elastic scattering arises under conditions of time-independent hyperfine interactions, as discussed in Sec. V B. It may also arise under conditions of time-dependent hyperfine interactions or spatial motion. However, this happens only, if these variations in time are stochastic, i.e., in no way correlate with the instant of photon absorption $\tilde{t}$ or emission t. Averaging over these stochastic variations results in the $(t-\tilde{t})$ dependence of the self-correlation function. Atomic spin fluctuations discussed in Sec. V C and diffusive motion of nuclei in space (Sec. V A) are such examples.

Nonstochastic variations in time usually cause the coherent inelastic scattering. It arises if, e.g., collective synchronized motion of nuclei in space takes place, Sec. V A 1, or synchronized time-dependent hyperfine interactions, Sec. V D, are induced.

IV. WAVE EQUATION AND ITS SOLUTION

A. Nuclear forward scattering wave equation

By using Eqs. (5) and (21) we obtain the wave equation for propagation of x rays in the nuclear resonant medium:

$$\frac{\partial E^{s}(\xi,\tau)}{\partial\xi}=-\int_{-\infty}^{\tau}\sum_{\tilde{s}}K^{s\tilde{s}}(\tau,\tilde{\tau})E^{\tilde{s}}(\xi,\tilde{\tau})\,d\tilde{\tau}. \tag{30}$$

Here dimensionless space and time variables

$$\xi=\frac{1}{4}\sigma_R N_0\frac{z}{\sin\Theta},\qquad \tau=\frac{t\Gamma_0}{\hbar} \tag{31}$$

are used. The dimensionless space variable ξ scales with the so-called effective resonant thickness $T_R=\sigma_R N_0 L$. The boundary condition for the wave equation (30) is given by Eq. (6).

The wave equation (30) can be represented also in short as follows:

$$\frac{\partial\boldsymbol{E}(\xi,\tau)}{\partial\xi}=-\hat{D}(\tau,\tilde{\tau})\boldsymbol{E}(\xi,\tilde{\tau}), \tag{32}$$

$$\hat{D}(\tau,\tilde{\tau})=\int_{-\infty}^{\tau}\hat{K}(\tau,\tilde{\tau})d\tilde{\tau}. \tag{33}$$

Equation (32) formally is similar to the Schrödinger equation. For this one has to imagine that ξ plays the role of the time variable in the Schrödinger equation and $-i\hat{D}$ plays the role of the ''time''-, i.e., ξ-independent Hamiltonian. Different techniques of solving the Schrödinger equation exist. To solve Eq. (32) we use a method similar to that of quantum-mechanical perturbation theory.

B. General solution

The general solution of the wave equation (30) can be given as a power series of ξ:

$$\boldsymbol{E}(\xi,\tau)=\sum_{p=0}^{\infty}\frac{(-\xi)^p}{p!}\boldsymbol{E}^{(p)}(\tau). \tag{34}$$

The first term $\boldsymbol{E}^{(0)}(\tau)$ is taken to be independent of the space variable ξ and defined by the boundary condition:

$$\boldsymbol{E}^{(0)}(\tau)=\boldsymbol{\mathcal{E}}(\tau). \tag{35}$$

The other terms, $\boldsymbol{E}^{(p)}(\tau)$, can be obtained after substitution of Eq. (34) into Eq. (32). This results in the recursion relation

$$\boldsymbol{E}^{(p)}(\tau)=\hat{D}(\tau,\tilde{\tau})\,\boldsymbol{E}^{(p-1)}(\tilde{\tau}) \tag{36}$$

with the help of which and of Eqs. (33) and (35) all the amplitudes $\boldsymbol{E}^{(p)}(\tau)$ in Eq. (34) can be calculated. The $\boldsymbol{E}^{(p)}(\tau)$ will be referred to as multiple scattering amplitudes of the order p.

By using Eqs. (34)–(36) the solution of the wave equation can be also represented in the following compact form:

$$\boldsymbol{E}(\xi,\tau)=\exp[-\xi\hat{D}(\tau,\tilde{\tau})]\boldsymbol{\mathcal{E}}(\tilde{\tau}). \tag{37}$$

It is remarkable that the solution of the wave equation, given by Eqs. (34)–(36), is general and is independent of the explicit form of the nuclear interactions which are hidden in the kernel $K^{s\tilde{s}}(\tau,\tilde{\tau})$. Equations (34)–(36) may be used conveniently for numerical calculations of the NFS time spectra. The kernel $K^{s\tilde{s}}(\tau,\tilde{\tau})$ is calculated once by using Eqs. (22)–(27). Then to calculate the time response given by Eq. (34) one applies the general procedure based on the recursion relations (36). Such a procedure was implemented in the code MOTIF.[29]

In a few particular cases one can obtain analytical solutions.

C. Single scattering approximation

One of such cases is the single scattering approximation, which is valid for thin samples or for short time intervals. Even though the solution in this approximation may not be exact for a real sample, nevertheless it gives a good basis for the analysis of physical problems.

The single scattering approximation is obtained by retaining only the zeroth and first terms ($p=0,1$) in the general solution (34). By using Eqs. (33)–(36) the radiation field in this approximation reads

$$E^{s}(\xi,\tau)=\mathcal{E}^{s}(\tau)-\xi\int_{-\infty}^{\tau}\sum_{\tilde{s}}K^{s\tilde{s}}(\tau,\tilde{\tau})\mathcal{E}^{\tilde{s}}(\tilde{\tau})d\tilde{\tau}. \tag{38}$$

If the incident radiation pulse is very short and can be approximated by

$$\boldsymbol{\mathcal{E}}(\tau)=\boldsymbol{e}^{0}\mathcal{E}\delta(\tau), \tag{39}$$

which corresponds to the conditions of experiments with pulsed synchrotron radiation, then Eq. (38) simplifies to

$$E^{s}(\xi,\tau)=\delta(\tau)\delta^{s0}-\xi K^{s0}(\tau,0). \tag{40}$$

Here $\delta(\tau)$ is the δ function, δ^{s0} is the Kronecker symbol, and $\boldsymbol{e}^{0}$ is the polarization vector of the incident radiation. We assume in Eq. (40) and everywhere in the following that $\mathcal{E}=1$.

The nuclear response is given by the second term of Eq. (40). The single scattering solution (40) provides a clear physical interpretation of the kernel $K^{s\tilde{s}}(\tau,\tilde{\tau})$. The kernel gives the coherent single scattering response in the forward direction of the nuclear system at time τ to the excitation at $\tilde{\tau}$. The superscripts $\tilde{s}$ and s represent the states of polarization of the incoming and scattered radiation.

D. Immediate response

At the time instant $t=0+$ (i.e., immediately after the excitation) the solution (40) in the single scattering approximation is exactly valid. It is valid exactly for a nuclear ensemble experiencing any spatial motion and any hyperfine interactions. It is also valid for samples of any thickness. It is valid exactly, since no multiple scattering is possible within such a short time.

By using Eqs. (40),(28), and (31) we obtain for the response in the forward direction immediately after excitation:

$$E^{s}(L,0+)=-\frac{\sigma_R N_0\,L}{4\sin\Theta}\,\delta^{s0}. \tag{41}$$

The immediate response of the nuclear system to instantaneous excitation given by Eq. (41) is independent of the character of nuclear motions and hyperfine interactions. It is also independent of whether the nuclei belong to atoms in a solid, or in a liquid, or in a gas. This fact has important consequences for the possibility of studying thermal excitatious which are discussed in Sec. V A 3 in more details.

E. Single resonance

An exact analytical solution is also obtainable in the case of a single nuclear resonance. A single resonance occurs, e.g., in the absence of hyperfine interactions: $\hat{\mathcal{H}}^{\mathrm{hf}}_{\beta_\lambda}=0$. In this case the evolution operator (15) is a c value: $U_{\beta}(t,\tilde{t})=\exp\{-(i/\hbar)[E_{\lambda}-i(\Gamma_0/2)\,\delta_{\lambda,e}](t-\tilde{t})\}$, independent of the group number β. For simplicity we assume that the motional part $M_{\beta}(t,\tilde{t})=1$. By using these facts together with Eqs. (22)–(27) we obtain that the single resonance correlation function is elastic: $K^{s\tilde{s}}(\tau,\tilde{\tau})=K_{1}^{s\tilde{s}}(\tau-\tilde{\tau})$, and is given by

$$K_{1}^{s\tilde{s}}(\tau)=\delta^{s\tilde{s}}\,\psi(\tau), \tag{42}$$

where

$$\psi(\tau)=\exp\left[i\left(\frac{\hbar\tilde{\omega}-E_0}{\Gamma_0}+\frac{i}{2}\right)\tau\right]\theta(\tau), \tag{43}$$

and $\theta(\tau)$ is the unit step function, nonzero if $\tau\geqslant 0$.

There is no polarization mixing and polarization dependence under these conditions. Therefore we omit the polarization index. We assume again that the incident radiation pulse is very short and can be represented by Eq. (39). As a result by using Eqs. (33)–(36) and the self-correlation function (42) all multiple scattering amplitudes can be calculated:

$E^{(p)}(\xi,\tau)=\psi(\tau)\xi^{p}\tau^{p-1}/(p-1)!$ and the analytical solution for the nuclear response to the prompt excitation is readily obtained:

$$E(\xi,\tau)=\delta(\tau)-\psi(\tau)\xi\sum_{p=1}^{\infty}\frac{(-\xi\tau)^{p-1}}{(p-1)!p!} \tag{44}$$

$$=\delta(\tau)-\psi(\tau)\xi\frac{J_1(2\sqrt{\xi\tau})}{\sqrt{\xi\tau}}. \tag{45}$$

The solution (45) is in agreement with the result obtained earlier in Ref. 3 by using Fourier transformation of the transmission frequency amplitude through the single resonance absorber. It is also in agreement with the solution obtained for the problem of the radiation pulse propagation in a single resonance optical medium.[30,31]

The terms in the sum of Eq. (44) are the multiple scattering amplitudes of the order p. If the condition $\xi\tau\ll 1$ is fulfilled (i.e., the sample is thin enough and the time of observation τ is not too far away from the excitation time $\tau=0$) the solution (44) can be restricted to the term with $p=1$. This actually represents the single scattering approximation discussed in Sec. IV C. Under these conditions the time response is simply proportional to $\xi\psi(\tau)$ and has the usual de excitation factor $\exp(-\Gamma_0 t/2)$ characteristic for the natural decay of an isolated nucleus.

If the sample is not thin ($\xi\gg 1$) or the instant of observation τ is far away from $\tau=0$, then other terms in the sum of Eq. (44), which represent multiple scattering, start to play a significant role. As a result, according to Eq. (45), the response acquires the modulation $J_1(2\sqrt{\xi\tau})/\sqrt{\xi\tau}$, which alters the natural decay by additional damping and oscillations. The latter is often referred to as dynamical beat and was observed both in the experiments with radioactive sources[32,33,11] and with synchrotron radiation.[34] The faster damping is often referred to as coherent speed up of the decay in NFS.[3,35,36]

How many scattering events in a sample with thickness parameter ξ experiences a photon arriving in the detector at time τ? In other words, how many terms in the sum of Eq. (44) are significant for the given values of ξ and τ? To estimate this value, which we denote as $p_{\xi\tau}$, let us assume that $p_{\xi\tau}\gg 1$ and use Stirling's formula $p!=\sqrt{2\pi}\,p^{p+0.5}e^{-p}$ for factorials in the denominator of Eq. (44). Under these assumptions the term with the number p in the sum of Eq. (44) can be approximated by $(\xi\tau e^{2}/p^{2})^{p-1}/\sqrt{2\pi}$. Thus the required number of scattering events can be estimated to be $p_{\xi\tau}\geqslant e\sqrt{\xi\tau}$.

By using the solution (45) and the wave equation (5) one can obtain also the following expression for the coherent nuclear current density induced in the sample by the instantaneous radiation pulse:

$$I(\xi,\tau)=\frac{c}{8\pi}N_0\sigma_R J_0(2\sqrt{\xi\tau})\psi(\tau). \tag{46}$$

It shows that directly after the excitation at $\tau=0$ there exists a homogeneous distribution of the nuclear currents over the whole sample: $I(\xi,0+)=$ const. Later the nuclear currents acquire an inhomogeneous spatial distribution which varies with time. According to Eq. (5) the nuclear ensemble radiates in forward direction the field $E(L,\tau)=-(2\pi/c\sin\Theta)\int_0^L I(z,\tau)dz$. At those instants, when the net coherent current density $\int_0^L I(z,\tau)dz=0$, i.e., when the currents interfere destructively, the nuclei do not radiate coherently in forward direction. These instants correspond to the minima of the dynamical beat.

F. Particular solutions in the frequency domain

An alternative procedure of the solution to that given by Eqs. (34)–(36) is to transform the wave equation (32) into the frequency domain, to solve it there, and then to Fourier-transform the solution back into the time domain. The transformation of the wave equation into the frequency domain results in

$$\frac{\partial \boldsymbol{E}(\xi,\varepsilon)}{\partial\xi}=-\int_{-\infty}^{+\infty}\frac{d\tilde{\varepsilon}}{2\pi}\hat{K}(\varepsilon,\tilde{\varepsilon})\boldsymbol{E}(\xi,\tilde{\varepsilon}), \tag{47}$$

where

$$\hat{K}(\varepsilon,\tilde{\varepsilon})=\int\int e^{i\varepsilon\tau}e^{-i\tilde{\varepsilon}\tilde{\tau}}\hat{K}(\tau,\tilde{\tau})d\tau d\tilde{\tau}. \tag{48}$$

The set of equations (47) describes, generally speaking, the inelastic coherent forward scattering with the matrix operator $\hat{K}(\varepsilon,\tilde{\varepsilon})$ representing the scattering of the radiation from the state with frequency $\tilde{\omega}+\tilde{\varepsilon}\Gamma_0/\hbar$ to another state with frequency $\tilde{\omega}+\varepsilon\Gamma_0/\hbar$. Only for some special types of kernels $\hat{K}(\varepsilon,\tilde{\varepsilon})$ it is possible to obtain the solution of Eq. (47) (see, e.g., Ref. 10, where the coherent inelastic scattering from vibrated nuclei was considered) but not in the general case. There is, however, a rather large subset of problems for which one can obtain a general solution, namely, for the *elastic* nuclear forward scattering when

$$\hat{K}(\varepsilon,\tilde{\varepsilon})=\hat{K}_1(\varepsilon)2\pi\delta(\varepsilon-\tilde{\varepsilon}). \tag{49}$$

In the time domain this corresponds to the relation

$$\hat{K}(\tau,\tilde{\tau})=\hat{K}_1(\tau-\tilde{\tau}), \tag{50}$$

which is verified by substitution of Eq. (50) into Eq. (48). In this case the integral in Eq. (47) vanishes and the integro-differential equation reduces to the linear differential equation

$$\frac{\partial \boldsymbol{E}(\xi,\varepsilon)}{\partial\xi}=-\hat{K}_1(\varepsilon)\boldsymbol{E}(\xi,\varepsilon), \tag{51}$$

the solution of which can be obtained in general form (see, e.g., Ref. 37):

$$\boldsymbol{E}(\xi,\varepsilon)=\exp[-\xi\hat{K}_1(\varepsilon)]\boldsymbol{\mathcal{E}}(\varepsilon), \tag{52}$$

with $\boldsymbol{\mathcal{E}}(\varepsilon)$ being the frequency spectrum of the incident radiation. The time dependence of NFS is obtained by reverse Fourier transformation.[3,5–8]

V. KERNELS IN PARTICULAR CASES

In each particular case the nuclear self-correlation function (22)–(27)—the kernel of the wave equation (30)—

should be calculated. For the rest the evaluation procedure of the time spectrum of NFS is standard and given by Eqs. (34)–(36), and (7).

The explicit form of the kernel is determined by the trajectory of motion $\boldsymbol{u}_\beta(t)$ that enters Eq. (24), and the evolution operator $\hat{U}_\beta(t,\tilde{t})$ that enters Eq. (27). The evolution operator (15) in turn is specified by the Hamiltonian $\hat{\mathcal{H}}_\beta(t)$ (11) and essentially by the type of the hyperfine interactions $\hat{\mathcal{H}}^{\mathrm{hf}}_{\beta_\lambda}(t)$.

In the present section we calculate the kernels for different particular cases of nuclear motion and hyperfine interactions and use them for calculating analytically the time spectra of NFS in the single scattering approximation. We consider first the influence of nuclear motion in space on NFS time spectra, Sec. V A. As a next step we consider time-independent hyperfine interactions, Sec. V B; time-dependent hyperfine interactions, in particular, stochastic fluctuations of the magnetic hyperfine field, Sec. V C; and switching of the magnetic hyperfine field direction synchronized with the nuclear excitation, Sec. V D.

A. Motion in space

In this section we discuss the effect of spatial motion only and therefore do not specify the part of the self-correlation function $L^{s\tilde{s}}_\beta(t,\tilde{t})$ representing other degrees of freedom. According to the definition (22)–(24) the effect of resonant nuclei in motion on NFS is totally defined by their position $\boldsymbol{u}_\beta(\tilde{t})$ at the moment $\tilde{t}$ of excitation and their position $\boldsymbol{u}_\beta(t)$ at the moment t of deexcitation. These positions are entering the phase factors $\exp[-i\tilde{\boldsymbol{k}}\boldsymbol{u}_\beta(t)]\exp[+i\tilde{\boldsymbol{k}}\boldsymbol{u}_\beta(\tilde{t})]$ in the motional part $M_\beta(t,\tilde{t})$ of the self-correlation function (24).

1. Collective motion

Let us first consider a simple case where the nuclei move as a rigid ensemble with a single displacement vector $\boldsymbol{u}_\beta(t) = \boldsymbol{u}(t)$. The self-correlation function (22)–(24) becomes

$$K^{s\tilde{s}}(t,\tilde{t}) = e^{-i\tilde{\boldsymbol{k}}\boldsymbol{u}(t)} e^{+i\tilde{\boldsymbol{k}}\boldsymbol{u}(\tilde{t})} \sum_\beta L^{s\tilde{s}}_\beta(t,\tilde{t}). \qquad (53)$$

No statistical averaging of the motional part is required in this case. The same nuclear ensemble but at rest $\boldsymbol{u}(t) = \mathrm{const}$ is characterized by the self-correlation function $K^{s\tilde{s}}_0(t,\tilde{t}) = \Sigma_\beta L^{s\tilde{s}}_\beta(t,\tilde{t})$. Let us assume that $\boldsymbol{E}_0(\xi,t)$ is the solution of the wave equation (30) with such a kernel. Then the solution of the wave equation for the moving system with the self-correlation function (53) is given by

$$\boldsymbol{E}(\xi,t) = \boldsymbol{E}_0(\xi,t) e^{-i\tilde{\boldsymbol{k}}\boldsymbol{u}(t)}. \qquad (54)$$

This is verified by substituting Eqs. (53) and (54) into Eq. (30). Thus, according to Eq. (54), the collective motion of all nuclei produces a phase modulation of the reemitted radiation. If, e.g., the sample is moving with constant velocity $\boldsymbol{v}$, so that $\boldsymbol{u}(t) = \boldsymbol{v}t$, then the phase factor in Eq. (54) is given by $\exp(-i\Omega_D t)$, where $\Omega_D = \tilde{\boldsymbol{k}}\boldsymbol{v}$ is simply a Doppler frequency shift. For more complicated motions the phase modulation is more sophisticated.

To reveal the phase modulation (54) due to collective motion one has to use a phase-sensitive detector. The time spectrum recorded with a usual non-phase-sensitive detector is given by Eq. (7). Since it is proportional to the modulo square of the nuclear response (54) the phase information is lost. Thus customarily the time spectrum of NFS is not sensitive to the collective motion of nuclei. Experimentally the insensitivity of the time spectra to the collective motion of nuclei was first proved in Ref. 38. It was shown theoretically in Ref. 12 that this conclusion is also true in a more general case of multiple Bragg diffraction. To observe the phase modulation another nuclear resonance scatterer should be used, playing the role of the phase-sensitive detector.[38,39]

2. Diffusive motion

A large set of problems that can be studied by using nuclear resonant scattering concerns diffusion of atoms in solids and liquids.[26] These problems are treated by specifying again the motional part of the self-correlation function. Diffusion is a stochastic process. Therefore one has to perform statistical averaging of the motional part (24) of the self-correlation function. Depending on the diffusion model $M_\beta(t,\tilde{t})$ takes different forms. Despite these differences there is a general feature: due to the fact that the time instant of excitation and emission is in no way correlated with the diffusion, the motional part of the self-correlation function should acquire in this case a $(t-\tilde{t})$ time dependence:

$$\langle e^{-i\tilde{\boldsymbol{k}}\boldsymbol{u}_\beta(t)} e^{i\tilde{\boldsymbol{k}}\boldsymbol{u}_\beta(\tilde{t})} \rangle = M_\beta(t-\tilde{t}), \qquad (55)$$

which describes elastic nuclear forward scattering.

In the particular case of free diffusion discussed in Ref. 26, the motional part of the self-correlation is given by

$$M_\beta(t-\tilde{t}) = e^{-D_\beta \tilde{k}^2 (t-\tilde{t})}, \qquad (56)$$

where D_β is the diffusion coefficient of atoms belonging to the group β. Different diffusion models were reviewed recently in Ref. 6 in connection with NFS. The first experimental studies of diffusion by using NFS were performed in Ref. 40.

3. Thermal lattice vibrations

Atoms bound in a crystal experience thermal vibrations. According to the theory of lattice dynamics in the harmonic approximation the displacement vector $\boldsymbol{u}_\beta(t)$ from the equilibrium position is given by a sum of displacements in the so called normal modes (see, e.g., Ref. 41). Each normal mode, or each phonon in the quantum mechanical language, is characterized by the momentum $\boldsymbol{q}$, branch number ν, dispersion law $\omega_\nu(\boldsymbol{q})$ and the state of polarization $\boldsymbol{e}_{\boldsymbol{q}\nu}(\beta)$. The phonon occupation numbers $\overline{n_{\boldsymbol{q}\nu}}$ at a given temperature characterize the intensity of thermal vibrations. The effect of the thermal lattice vibrations on the time dependence of NFS exhibits itself via the motional part of self-correlation function (24). We consider only elastic scattering here when the phonon state before and after scattering is the same, although in the intermediate state when the nuclear ensemble is excited, it may be different. That means we have to perform the thermal average of Eq. (24) over the phonon occupation numbers.

The result is well known (see, e.g. Refs. 42 and 41). We reproduce it here without derivation:

$$M_\beta(t-\tilde{t})=\langle e^{-i\tilde{\boldsymbol{k}}\boldsymbol{u}_\beta(t)}e^{+i\tilde{\boldsymbol{k}}\boldsymbol{u}_\beta(\tilde{t})}\rangle$$
$$=\exp\{-\langle(\tilde{\boldsymbol{k}}\boldsymbol{u}_\beta)^2\rangle+\langle[\tilde{\boldsymbol{k}}\boldsymbol{u}_\beta(t)][\tilde{\boldsymbol{k}}\boldsymbol{u}_\beta(\tilde{t})]\rangle\}, \tag{57}$$

$$\langle[\tilde{\boldsymbol{k}}\boldsymbol{u}_\beta(t)][\tilde{\boldsymbol{k}}\boldsymbol{u}_\beta(\tilde{t})]\rangle$$
$$=\frac{\hbar}{2nm_\beta}\sum_{\boldsymbol{q}\nu}\frac{|\tilde{\boldsymbol{k}}\boldsymbol{e}_{\boldsymbol{q}\nu}(\beta)|^2}{\omega_\nu(\boldsymbol{q})}$$
$$\times[(\overline{n}_{\boldsymbol{q}\nu}+1)e^{-i\omega_\nu(\boldsymbol{q})(t-\tilde{t})}+\overline{n}_{\boldsymbol{q}\nu}\,e^{i\omega_\nu(\boldsymbol{q})(t-\tilde{t})}]. \tag{58}$$

Here m_β is the mass of the atom in group β and n is the number of the unit cells in the sample.

It is clear that independent of how the nuclei are moving $M_\beta(t-\tilde{t})=1$ at $t=\tilde{t}$. If $(t-\tilde{t})\gg\omega_{\mathrm{ph}}^{-1}$, where ω_{ph} is a typical phonon frequency, then the term (58) vanishes and the self-correlation function becomes

$$M_\beta(t-\tilde{t})=\exp\{-\langle[\boldsymbol{k}\boldsymbol{u}_\beta]^2\rangle\}=f_\beta(\tilde{\boldsymbol{k}}), \tag{59}$$

where $f_\beta(\tilde{\boldsymbol{k}})$ is the Lamb-Mössbauer factor giving the probability of elastic resonance absorption or emission in the direction of the photon wave vector $\tilde{\boldsymbol{k}}$. The factor $f_\beta(\tilde{\boldsymbol{k}})$ can be calculated by using Eq. (58) taken at $t=\tilde{t}$.

An important question is, what happens in between? According to Eqs. (57) and (58) the self-correlation function should decay from 1 to $f_\beta(\tilde{\boldsymbol{k}})$ with a law defined by the phonon spectra $\omega_\nu(\boldsymbol{q})$.

Let us derive as an example the decay law in a particular case of a cubic crystal with one atom ($\beta=1$) in its unit cell. In this case the thermal vibrations are anisotropic, i.e., $\langle u^i(t)u^l(\tilde{t})\rangle=\delta^{il}\langle u(t)u(\tilde{t})\rangle$, where $i,l=1,2,3$ denote Cartesian components of the displacement vector $\boldsymbol{u}$. As a result the self-correlation function (57) takes the form

$$M(t)=\exp\{-\tilde{k}^2[\langle u^2\rangle-\langle u(t)u(0)\rangle]\}, \tag{60}$$

where

$$\langle u(t)u(0)\rangle=\frac{\hbar}{2nm}\sum_{\boldsymbol{q}\nu}\frac{1}{\omega_\nu(\boldsymbol{q})}[(\overline{n}_{\boldsymbol{q}\nu}+1)e^{-i\omega_\nu(\boldsymbol{q})t}$$
$$+\overline{n}_{\boldsymbol{q}\nu}e^{i\omega_\nu(\boldsymbol{q})t}]. \tag{61}$$

Here we have put $\tilde{t}=0$. Following the usual procedure we replace the summation over $\boldsymbol{q}$ by integration over phonon frequencies ω and introduce the phonon density of states $D(\omega)$. Equation (61) may be then transformed to

$$\langle u(t)u(0)\rangle=\frac{\hbar}{m}\int_0^\infty\frac{d\omega D(\omega)}{\omega}\left[(\overline{n}(\omega)+\tfrac{1}{2})\cos\omega t-\frac{i}{2}\sin\omega t\right]. \tag{62}$$

If the crystal temperature T is high, the phonon occupation numbers $n(\omega)\simeq kT/\hbar\omega\gg1$ and Eq. (62) reduces to

$$\langle u(t)u(0)\rangle=\frac{kT}{m}\int_0^\infty d\omega D(\omega)\omega^{-2}\cos\omega t. \tag{63}$$

In the Debye approximation the phonon density of states is given by $D(\omega)=3\omega^2/\omega_D^3$, with ω_D being the Debye frequency—the cutoff frequency of the density function. In this approximation the integration in Eq. (63) is performed in the limits $[0,\omega_D]$ and we obtain:

$$\langle u(t)u(0)\rangle=\langle u^2\rangle\frac{\sin\omega_D t}{\omega_D t},\quad\langle u^2\rangle=\frac{3kT}{m\omega_D^2}. \tag{64}$$

The self-correlation function (57) in a cubic crystal, in the high-temperature limit, and under assumption of the Debye approximation for the phonon density of states finally takes the form

$$M(t)=\exp\left\{-\tilde{k}^2\langle u^2\rangle\left[1-\frac{\sin\omega_D t}{\omega_D t}\right]\right\}. \tag{65}$$

The Debye frequency is typically $\omega_D\simeq10^{13}\ \mathrm{s}^{-1}$. Therefore the self-correlation function decays from 1 to the level given by the Lamb-Mossbauer factor $\exp(-\tilde{k}^2\langle u^2\rangle)$ within a few 10^{-13} s.

This time interval is usually much shorter than the lifetimes of the low-lying nuclear transitions, which are typically $\tau_0=\hbar/\Gamma_0>10^{-12}$ s. Therefore in this time interval the single scattering approximation is valid, Sec. IV C, and according to Eqs. (7) and (40) the NFS time spectrum

$$S(t)\propto\xi^2|M(t)|^2$$

with $M(t)$ given by Eq. (57) in the general case of harmonic vibrations, or by Eqs. (60),(61), and (65) in particular cases. The NFS spectra in this time range bear information on the time dependence of atomic vibrations via $\langle u(t)u(0)\rangle$, as well as on the phonon spectra $\omega_\nu(\boldsymbol{q})$. It is noteworthy, that the spectra in this time range are nonzero independent of how small the value of the Lamb-Mössbauer factor is. Even in liquids and gases, where the value of the Lamb-Mössbauer factor tends to zero, one can observe a strong NFS signal in this time range.

The time interval is very short, where the time dependence caused by thermal vibrations shows up in NFS, and it is beyond the reach of present-day experimental techniques. However, in future one may hope it will be possible to perform measurements of NFS in this range and thus to perform studies of atomic motions and not only in solids, but in liquids, gases, etc. as well. The problem to our mind could be solved by mapping time dependence of photon emission into angular dependence of photon emission. Due to the recently demonstrated light-house effect[43] such mapping can be realized with the help of fast spinning nuclear resonance samples irradiated with synchrotron radiation. Presently available spinning rates beyond 30 kHz could allow an extremely short time interval of 10^{-13} s to be transformed into a measurable angular change in photon emission of 0.02 μrad. The nuclear transitions with lifetimes slightly more than ω_D^{-1} would be preferable in such experiments.

In the following analysis the influence of the hyperfine interactions on the NFS time spectra will be discussed. Their influence is far beyond the time region of influence of thermal vibrations, discussed in the present section. Therefore the motional part of the self-correlation function will be assumed further to be time independent and to be represented in accordance with Eq. (59) with the help of the Lamb-Mössbauer factor $f_\beta(\tilde{\boldsymbol{k}})$.

B. Time-independent hyperfine interactions

Let $|\beta_\lambda\rangle$ be eigenvectors and ϵ_{β_λ} eigenenergies of the time-independent hyperfine interaction Hamiltonian $\hat{\mathcal{H}}^{\mathrm{hf}}_{\beta_\lambda}$. If $|\beta_\lambda\rangle$ and ϵ_{β_λ} are known, the matrix elements of the evolution operator can be readily evaluated by Eq. (15):

$$\mathcal{U}_{\beta_\lambda\tilde{\beta}_\lambda}(t_2,t_1)=\delta_{\beta_\lambda\tilde{\beta}_\lambda} \times\exp\left[-\frac{i}{\hbar}\left(E_\lambda-i\frac{\Gamma_0}{2}\delta_{\lambda,e}+\epsilon_{\beta_\lambda}\right)(t_2-t_1)\right]. \tag{66}$$

Hereafter the notation $\hat{\mathcal{U}}$ is reserved for the evolution operator under the condition of time-independent hyperfine interactions. Inserting Eq. (66) into Eq. (27) and by using Eq. (59) for the motional part we obtain the result that the complete self-correlation function (22) is purely elastic, i.e., $K^{s\tilde{s}}(t,\tilde{t})=K_1^{s\tilde{s}}(t-\tilde{t})$, and is given by

$$K_1^{s\tilde{s}}(t)=\eta(t)\sum_{l\equiv\{\beta,\beta_g,\beta_e\}}\mathcal{A}_l^{s\tilde{s}}e^{-i\Omega_l t}, \tag{67}$$

$$\eta(t)=\exp\left[\frac{i}{\hbar}(\hbar\tilde{\omega}-E_0+i\Gamma_0/2)t\right]\theta(t), \tag{68}$$

$$\mathcal{A}_l^{s\tilde{s}}=X_\beta f_\beta(\tilde{\boldsymbol{k}})j^{s}_{\beta_g\beta_e}(\tilde{\boldsymbol{k}})j^{\tilde{s}}_{\beta_e\beta_g}(-\tilde{\boldsymbol{k}}) \tag{69}$$

with $\hbar\Omega_l=\epsilon_{\beta_e}-\epsilon_{\beta_g}$ being the corrections to the transition energies E_0 arising due to the hyperfine interaction. Unless this causes ambiguities we use for brevity a joint index $l\equiv\{\langle\beta_g|\Leftrightarrow|\beta_e\rangle,\beta\}$ to denote both the transition between the ground and excited nuclear states and the group number β.[44]

From Eqs. (40) and (67) we obtain in single scattering approximation for the s-polarization component of the emitted radiation:

$$E^s(\xi,t)\propto-\xi\eta(t)\sum_l\mathcal{A}_l^{s0}e^{-i\Omega_l t}. \tag{70}$$

The nuclear response given by Eq. (70) is a sum of monochromatic components with the frequencies Ω_l. The amplitude $\mathcal{A}_l^{s0}$ of each emitted frequency component is proportional to the product of the absorption $j^0_{\beta_e\beta_g}(-\tilde{\boldsymbol{k}})$ and the emission $j^s_{\beta_g\beta_e}(\tilde{\boldsymbol{k}})$ matrix elements. The interference of the different monochromatic components results in a time spectrum with periodic modulation called quantum beat.[4,45] The quantum beat pattern is defined both by the transition frequencies and by the matrix elements of the nuclear transition currents and thus bears the information on the hyperfine interactions experienced by the nuclei.

Examples of evaluations of the NFS time spectra under conditions of time-independent hyperfine interactions by using the procedure described in the present paper and fits of experimental spectra are presented in Refs. 9 and 29.

C. Fluctuating magnetic hyperfine field

In this and in the next section we present examples of calculations of the nuclear self-correlation functions for two particular cases of time-dependent hyperfine interactions. In both cases the direction of the magnetic hyperfine fields is assumed to change in time. In the case, considered in the present section, the direction of the hyperfine field stochastically changes at each nucleus. In the second case, considered in the next section, the direction of the hyperfine field is supposed to change at all nuclei simultaneously at a definite time.

The Hamiltonian for magnetic hyperfine field changing in time reads

$$\hat{\mathcal{H}}^{\mathrm{hf}}_{\beta_\lambda}(t)=-\mu_\lambda\frac{\hat{\boldsymbol{J}}_\lambda\boldsymbol{B}^{(\beta)}(t)}{J_\lambda}. \tag{71}$$

First we calculate the time spectrum of NFS in the presence of a fluctuating magnetic hyperfine field that jumps randomly between the values $B^{(\beta)}$ and $-B^{(\beta)}$ along $\boldsymbol{n}_0^{(\beta)}$:

$$\boldsymbol{B}^{(\beta)}(t)=B_0^{(\beta)}\boldsymbol{n}_0^{(\beta)}f(t). \tag{72}$$

Here $f(t)$ is a random function of time taking only the values ±1. Such a stochastic model was used in Ref. 28 to calculate the Mössbauer resonance line shape of a nucleus in an environment with fluctuating atomic spin direction.

For simplicity we assume here pure magnetic hyperfine interactions (for a more general cases see Ref. 28). The eigenvectors $|\beta_\lambda\rangle$ of the Hamiltonian $\hat{\mathcal{H}}^{\mathrm{hf}}_{\beta_\lambda}(-\infty)$ are equal in this case to the eigenvectors $|m_\lambda\rangle$ of the nuclear spin-projection operator, with the projection on the magnetic hyperfine field direction $\boldsymbol{n}_0^{(\beta)}$. Here m_λ is the magnetic quantum number. In this particular case the eigenenergies are given by $\epsilon_{\beta_\lambda}=-\mu_\lambda m_\lambda B^{(\beta)}/J_\lambda$. Under these conditions and by using Eqs. (15),(71), and (72) we obtain for the evolution operator

$$U_{\beta_\lambda\tilde{\beta}_\lambda}(t_2,t_1)=\delta_{\beta_\lambda\tilde{\beta}_\lambda}\exp\left\{-\frac{i}{\hbar}\left[\left(E_\lambda-i\frac{\Gamma_0}{2}\delta_{\lambda,e}\right)(t_2-t_1)+\epsilon_{\beta_\lambda}\int_{t_1}^{t_2}f(t)dt\right]\right\}. \tag{73}$$

The self-correlation function (22)–(27) can then be readily evaluated,

$$K^{s\tilde{s}}(t,\tilde{t})=\eta(t-\tilde{t})\sum_l\mathcal{A}_l^{s\tilde{s}}\exp\left\{-i\Omega_l\int_{\tilde{t}}^{t}f(T)dT\right\}. \tag{74}$$

Here $\mathcal{A}_l^{s\tilde{s}}$ is given by Eq. (69) and Ω_l in this particular case of pure magnetic interactions by

$$\hbar\Omega_l=(\mu_g m_g/J_g-\mu_e m_e/J_e)B^{(\beta)}. \tag{75}$$

One still has to perform the stochastic average of the phase factors $\exp\{-i\Omega_l\int_{\tilde{t}}^{t}f(T)\,dT\}$ in Eq. (74). As long as the hyperfine field fluctuations in no way correlate with the time instants of absorption $\tilde{t}$ and emission t the result of averaging should depend on the difference $(t-\tilde{t})$ only. This means that the self-correlation function reduces to $K^{s\tilde{s}}(t,\tilde{t})=K_1^{s\tilde{s}}(t-\tilde{t})$, i.e., to the function for pure elastic coherent scattering. The actual result of averaging depends on the details of the stochastic model. In the simplest case, where the fluctuations of the hyperfine field direction are described by a single parameter W—the mean frequency of fluctuations—the averaging results in (see Ref. 28 for details):

$$\left\langle \exp\left\{ -i\Omega_l \int_0^t f(T)dT \right\} \right\rangle = \left[\cos(x_l W t) + \frac{1}{x_l} \sin(x_l W t) \right] e^{-Wt} \tag{76}$$

with $x_l=(\Omega_l^2/W^2-1)^{1/2}$. The self-correlation function in the presence of the fluctuating magnetic hyperfine field becomes

$$K_1^{s\tilde{s}}(t)=\eta(t)\sum_l \mathcal{A}_l^{s\tilde{s}} \left[\cos(x_l W t) + \frac{1}{x_l} \sin(x_l W t) \right] e^{-Wt}. \tag{77}$$

Let us consider two limiting cases. If the mean frequency of the fluctuations is very low $(W \ll \Omega_l)$, the so called slow relaxation limit, we obtain $x_l W \simeq \Omega_l$ and $x_l \gg 1$. Then the self-correlation function reduces to

$$K_1^{s\tilde{s}}(t)=\eta(t)\sum_l \mathcal{A}_l^{s\tilde{s}} \cos(\Omega_l t)\, e^{-Wt}. \tag{78}$$

Since $\cos(\Omega_l t)=[\exp(i\Omega_l t)+\exp(-i\Omega_l t)]/2$ the function (78) can be interpreted as a superposition of two unperturbed nuclear self-correlation functions (67) with constant magnetic hyperfine fields $B^{(\beta)}$ and $-B^{(\beta)}$, respectively. Besides the natural decay factor $\exp(-\Gamma_0 t/2)$ of $\eta(t)$ here appears an additional deexcitation factor $\exp(-Wt)$, which causes the faster nuclear coherent response. This corresponds to the broadening of the nuclear resonances in the frequency spectrum.

In the other limiting case of very fast relaxation $(W \gg \Omega_l)$ we obtain $x_l \simeq i$ and as a result the self-correlation function becomes

$$K_1^{s\tilde{s}}(t)=\eta(t)\sum_l \mathcal{A}_l^{s\tilde{s}}. \tag{79}$$

The dependence on the magnetic field disappears, since the fluctuations of the magnetic hyperfine field are so rapid, that the nuclei see its average value to be zero.

First experimental studies and evaluations of NFS time spectra in paramagnetic environments with fluctuating atomic spins were reported recently in Refs. 46 and 47. Experimental evidence of superparamagnetic fluctuations seen by NFS were given in Ref. 47.

D. Switching of the magnetic hyperfine fields

A very different picture arises if the magnetic hyperfine fields are ‘‘fluctuating’’ regularly in time and in space. In other words, the directions of the hyperfine fields are switched (rotated) from $\boldsymbol{n}_0^{(\beta)}$ to $\boldsymbol{n}_0^{(\beta')}$ instantaneously and simultaneously at each nucleus within each group β at a definite time instant t':

$$\boldsymbol{B}^{(\beta)}(t)=B_0^{(\beta)}[\boldsymbol{n}_0^{(\beta)}\theta(t'-t)+\boldsymbol{n}_0^{(\beta')}\theta(t-t')]. \tag{80}$$

The rotation is specified by the three Eulerian angles: $\{\chi_1',\chi_2',\chi_3'\}=\boldsymbol{\chi}'$.

Before switching $(t<t')$ the direction and the value of the magnetic hyperfine field are constant. Therefore the correlation function in this time interval is equal to the unperturbed correlation function (67)–(69) derived in Sec. V B.

After switching $(t>t')$ the hyperfine field (80) and the Hamiltonian (71) are again time independent; however, different from the initial ones. Eigenvectors $|\beta_\lambda'\rangle$ of the Hamiltonian associated with the new direction of the hyperfine fields are related to the eigenvectors $|\beta_\lambda\rangle$ of the Hamiltonian before switching through the transformation

$$|\beta_\lambda\rangle=\sum_{\beta_\lambda'} |\beta_\lambda'\rangle \mathcal{D}_{\beta_\lambda'\beta_\lambda}(\boldsymbol{\chi}'), \quad \langle\beta_\lambda|=\sum_{\beta_\lambda'} \mathcal{D}^{-1}_{\beta_\lambda\beta_\lambda'}(\boldsymbol{\chi}')\langle\beta_\lambda'|. \tag{81}$$

Here $\mathcal{D}_{\beta_\lambda'\beta_\lambda}(\boldsymbol{\chi}')$ is a unitary matrix. In the particular case, when the eigenvectors $|\beta_\lambda\rangle$ and $|\beta_\lambda'\rangle$ are the eigenvectors of the nuclear spin-projection operator, it coincides with the matrix of finite rotations $\mathcal{D}^{(J_\lambda)}_{m_\lambda' m_\lambda}(\boldsymbol{\chi}')$.[48,19]

The unknown values, which still have to be defined in order to calculate the correlation function after switching, are the matrix elements of the evolution operator. To do this we make use of the composition law $\hat{U}(t_2,t_1)=\hat{\mathcal{U}}(t_2,t')\,\hat{\mathcal{U}}(t',t_1)$, of the fact that the Hamiltonian (71) is time independent although different in the time intervals (t_2,t') and (t',t_1), see Eq. (80), of the definition (15), and of the relation (81). The matrix elements of the evolution operator then become

$$U_{\beta_\lambda'\beta_\lambda}(t_2,t_1)=\mathcal{U}_{\beta_\lambda'\beta_\lambda'}(t_2,t')\,\mathcal{D}_{\beta_\lambda'\beta_\lambda}\,\mathcal{U}_{\beta_\lambda\beta_\lambda}(t',t_1), \tag{82}$$

$$U^{-1}_{\beta_\lambda\beta_\lambda'}(t_2,t_1)=\mathcal{U}^{-1}_{\beta_\lambda\beta_\lambda}(t',t_1)\,\mathcal{D}^{-1}_{\beta_\lambda\beta_\lambda'}\,\mathcal{U}^{-1}_{\beta_\lambda'\beta_\lambda'}(t_2,t'). \tag{83}$$

It was taken into account that only the diagonal matrix elements $\mathcal{U}_{\beta_\lambda\beta_\lambda}$ of the unperturbed evolution operator (66) have nonzero values. Combining Eqs. (82) and (83) with Eq. (27) we obtain the following expressions for the correlation function after switching $(t>t'>\tilde{t})$:

$$K^{s\tilde{s}}(t,\tilde{t})=\eta(t-\tilde{t})\sum_{l'\equiv\{\beta,\beta_g',\beta_e'\}} \mathcal{A}_{l'}^{s\tilde{s}}(\boldsymbol{\chi}',t'-\tilde{t})e^{-i\Omega_{l'}(t-t')}, \tag{84}$$

$$\mathcal{A}_{l'}^{s\tilde{s}}(\boldsymbol{\chi}',t'-\tilde{t})=X_\beta f_\beta(\tilde{\boldsymbol{k}})\, j_{l'}^{s}(\tilde{\boldsymbol{k}})\sum_{\beta_g,\beta_e} S_{l'l}'(\boldsymbol{\chi}',t'-\tilde{t})j_l^{\tilde{s}}(-\tilde{\boldsymbol{k}}), \tag{85}$$

$$S_{l'l}'(\boldsymbol{\chi}',t'-\tilde{t})=\mathcal{D}_{\beta_g'\beta_g}(\boldsymbol{\chi}')\mathcal{D}_{\beta_e'\beta_e}(\boldsymbol{\chi}')\, e^{-i\Omega_l(t'-\tilde{t})}. \tag{86}$$

The correlation function (84) has a structure similar to that of the unperturbed correlation function (67).[49] However this similarity is only formal. First, the time variables t and $\tilde{t}$ are entering Eqs. (84)–(86) independently. Thus unlike the unperturbed correlation function, given by Eq. (67), the correlation function of Eq. (84) describes *inelastic* coherent scattering. Second, the amplitudes $\mathcal{A}^{s\tilde{s}}_{l'}(\boldsymbol{\chi}',t'-\tilde{t})$ of the monochromatic components $\Omega_{l'}$ are different. They are built up by the interference of all the initially excited transitions l. The interference shows up in Eq. (85) as the sum of the current density matrix elements $j^{\tilde{s}}_{l}(-\tilde{\boldsymbol{k}})$ with amplitudes given by $S'_{l'l}(\boldsymbol{\chi}',)$ in Eq. (86). By varying the angle $\boldsymbol{\chi}'$ of switching and the switching time t' one can change the amplitudes $S'_{l'l}$ and thus can control the interference pattern and the coherently emitted intensity. The switching may result in the emission of new frequency and polarization components as well as in the suppression of already excited ones[11,13,50–52] or in the time reversal of the time spectrum.[53] For example, one can make the interference totally destructive and thus suppress the coherent reemission.[13] However, this implies by no means that the nuclear excitation is destroyed. By the next switching at proper time and with proper angle one can restore constructive interference and see again coherently emitted intensity.[13]

This second switching is described similarly to the procedure outlined above. For example, if at time t'' the magnetic hyperfine field is now switched to $\boldsymbol{n}_0^{(\beta'')}$, the eigenvectors $|\beta''_\lambda\rangle$ of the Hamiltonian associated with this new direction of the hyperfine field are related to the previous eigenvectors $|\beta'_\lambda\rangle$ through Eq. (81) with the substitutions $\beta_\lambda \rightarrow \beta'_\lambda$ and $\beta'_\lambda \rightarrow \beta''_\lambda$. Further it can be shown that the nuclear self-correlation function is described by the same Eqs. (84) and (85); however, with the replacement $l' \rightarrow l''$, and $S'_{l'l}(\boldsymbol{\chi}',t'-\tilde{t}) \rightarrow S''_{l''l}(\boldsymbol{\chi}'',t''-\tilde{t})$, where $S''_{l''l}(\boldsymbol{\chi}'',t''-\tilde{t})$ is now defined as

$$S''_{l''l}(\boldsymbol{\chi}'',t''-\tilde{t}) = \sum_{\beta'_e,\beta'_g} S'_{l''l'}(\boldsymbol{\chi}'',t''-t')\,S'_{l'l}(\boldsymbol{\chi}',t'-\tilde{t}). \tag{87}$$

Examples of transformations occurring after the second switching were presented in Refs. 50 and 13. Any subsequent switching is described similarly.

Some additional details of the theory of nuclear resonant scattering for the case of switching the magnetic hyperfine fields can be found in Refs. 11,13,50, and 53. Examples of evaluations of the NFS time spectra under conditions of switching of the magnetic hyperfine fields by using the procedure described in the present paper and fits of experimental spectra are presented in Ref. 13.

VI. CONCLUSIONS

A general approach is presented of solving nuclear resonant forward scattering problems directly in time and in space. It is based on the solution of the first-order integro-differential equation with a kernel that is a double time nuclear self-correlation function $K(t,\tilde{t})$. The kernel represents a coherent single scattering response in forward direction of their spatial system at time t to the excitation at $\tilde{t}$. The form of the kernel is defined by the type of interactions the nuclei experience with their environment and by the character of their spatial motion. A general procedure is introduced for the solution of the wave equation. It is independent of the type of the kernel. The kernels for some particular cases of hyperfine interactions and nuclear motion in space were presented. Examples of the NFS time spectra under conditions of time-independent as well as time-dependent hyperfine interactions obtained by the direct calculations in time and space can be found, e.g., in Refs. 9,13, and 29.

The solution procedure directly in time and space can also be applied to other scattering problems, such as nuclear Bragg diffraction, nuclear resonant small angle scattering, etc. For these cases the wave equation (30) should be changed to describe along with the forward scattered amplitude also the radiation components scattered at nonzero angles. In Ref. 12 an appropriate wave equation for multiple Bragg diffraction was introduced. In such problems the double time self-correlation function should acquire additionally a double momentum dependence $K^{s\tilde{s}}(t,\tilde{t},\boldsymbol{k},\tilde{\boldsymbol{k}})$, since the wave vector $\boldsymbol{k}$ of the scattered and $\tilde{\boldsymbol{k}}$ of the incident photons are now different.

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[1] E. Gerdau, and U. van Bürck, in *Resonant Anomalous X-ray Scattering, Theory and Applications*, edited by G. Materlik, C. J. Sparks, and K. Fischer (North-Holland, Amsterdam, 1994), p. 589.

[2] G. V. Smirnov, Hyperfine Interact. **97/98**, 551 (1996).

[3] Yu. Kagan, A. M. Afanas'ev, and V. G. Kohn, J. Phys. C **12**, 615 (1979).

[4] G. T. Trammell and J. P. Hannon, Phys. Rev. B **18**, 165 (1978); **19**, 3835 (1979).

[5] W. Sturhahn and E. Gerdau, Phys. Rev. B **49**, 9285 (1994).

[6] G. V. Smirnov and V. G. Kohn, Phys. Rev. B **52**, 3356 (1995); **57**, 5788 (1998).

[7] L. Deák, L. Bottyàn, D. L. Nagy, and H. Spiering, Phys. Rev. B **53**, 6158 (1996).

[8] M. Haas, E. Realo, H. Winkler, W. Meyer-Klaucke, A. X. Trautwein, O. Leupold, and H. D. Rüter, Phys. Rev. B **56**, 14 082 (1997).

[9] Yu. V. Shvyd'ko, U. van Bürck, W. Potzel, P. Schindelmann, E. Gerdau, O. Leupold, J. Metge, H. D. Rüter, and G. V. Smirnov, Phys. Rev. B **57**, 3552 (1998).

[10] Yu. V. Shvyd'ko and G. V. Smirnov, J. Phys.: Condens. Matter **4**, 2663 (1992).

[11] Yu. V. Shvyd'ko, S. L. Popov, and G. V. Smirnov, J. Phys.: Condens. Matter **5**, 1557 (1993); **5**, 7047 (1993).

[12] V. G. Kohn and Yu. V. Shvyd'ko, J. Phys.: Condens. Matter **7**, 7589 (1995).

[13] Yu. V. Shvyd'ko, T. Hertrich, U. van Bürck, E. Gerdau, O. Leupold, J. Metge, H. D. Rüter, S. Schwendy, G. V. Smirnov, W. Potzel, and P. Schindelmann, Phys. Rev. Lett. **77**, 3232 (1996).

[14] T. Mitsui, T. Shimizu, Y. Imai, Y. Yoda, X. W. Zhang, H. Takei, T. Harami, and S. Kikuta, Jpn. J. Appl. Phys., Part 1 **36**, 6525 (1997).

[15] L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Wiley, New York, 1975), Sec. 1.6.

[16] S. Takagi, Acta Crystallogr. **15**, 1311 (1962); J. Phys. Soc. Jpn. **26**, 1239 (1968).

[17] A. I. Akhiezer and V. B. Berestetskii, *Quantum Electrodynamics* (Wiley, New York, 1965).

[18] K. Blum, *Density Matrix Theory and Applications* (Plenum Press, New York, 1981).

[19] V. B. Berestetskii, E. M. Lifshitz, and L. P. Pitaevskii, *Relativistic Quantum Theory* (Pergamon, New York, 1971).

[20] J. P. Hannon and G. T. Trammell, Phys. Rev. **186**, 306 (1969).

[21] Yu. M. Kagan and A. M. Afanas'ev, Z. Naturforsch. Teil A **28**, 1351 (1973).

[22] P. A. Aleksandrov, Zh. Éksp. Teor. Fiz. **67**, 728 (1974) [Sov. Phys. JETP **40**, 360 (1975)].

[23] Yu. V. Shvyd'ko, A. I. Chumakov, A. Q. R. Baron, E. Gerdau, R. Rüffer, A. Bernhard, and J. Metge, Phys. Rev. B **54**, 14 942 (1996).

[24] A. Messiah, *Quantum Mechanics* (North-Holland Publishing, Amsterdam, 1962), Vol. II.

[25] L. Van Hove, Phys. Rev. **55**, 190 (1955).

[26] K. S. Singwi and A. Sjölander, Phys. Rev. **120**, 1093 (1960).

[27] A. M. Afanas'ev and Yu. Kagan, Zh. Éksp. Teor. Fiz. **45**, 1660 (1963) [Sov. Phys. JETP **18**, 1139 (1964)].

[28] M. Blume and J. A. Tjon, Phys. Rev. **165**, 446 (1967).

[29] MOTIF.23, a program for fitting time spectra of nuclear resonant forward scattering, available from the anonymous ftp://i2aix04.desy.de/pub/motif/ or from the author via e-mail: Yuri.Shvydko@desy.de. The User guide to Motif is the postscript file motif_23.ps. It includes also examples of time spectra evaluations.

[30] D. C. Burnham and R. Y. Chiao, Phys. Rev. **188**, 667 (1969).

[31] A. Laubereau and W. Kaiser, Rev. Mod. Phys. **50**, 607 (1978).

[32] F. J. Lynch, R. E. Holland, and M. Hamermesh, Phys. Rev. **120**, 513 (1960).

[33] Yu. V. Shvyd'ko, S. L. Popov, and G. V. Smirnov, Pis'ma. Zh. Éksp. Teor. Fiz. **53**, 217 (1991) [JETP Lett. **53**, 231 (1991)].

[34] U. van Bürck, D. P. Siddons, J. B. Hastings, U. Bergmann, and R. Hollatz, Phys. Rev. B **46**, 6207 (1992).

[35] J. B. Hastings, D. P. Siddons, U. van Bürck, R. Hollatz, and U. Bergmann, Phys. Rev. Lett. **66**, 770 (1991).

[36] Yu. V. Shvyd'ko, G. V. Smirnov, S. L. Popov, and T. Hertrich, Pis'ma. Zh. Éksp. Teor. Fiz. **53**, 69 (1991) [JETP Lett. **53**, 69 (1991)].

[37] M. Blume and O. C. Kistner, Phys. Rev. **171**, 417 (1968).

[38] Yu. V. Shvyd'ko, A. I. Chumakov, G. V. Smirnov, V. G. Kohn, T. Hertrich, U. van Bürck, E. Gerdau, H. D. Rüter, J. Metge, and O. Leupold, Europhys. Lett. **22**, 305 (1993).

[39] G. V. Smirnov, U. van Bürck, J. Arthur, S. L. Popov, A. Q. R. Baron, A. I. Chumakov, S. L. Ruby, W. Potzel, and G. S. Brown, Phys. Rev. Lett. **77**, 183 (1996).

[40] B. Sepiol, A. Meyer, G. Vogl, R. Rüffer, A. I. Chumakov, and A. Q. R. Baron, Phys. Rev. Lett. **76**, 3220 (1996).

[41] A. A. Maradudin, E. W. Montroll, G. H. Weiss, and I. P. Ipatova, *Theory of Lattice Dynamics in the Harmonic Approximation* (Academic, New York, 1971).

[42] G. T. Trammell, Phys. Rev. **126**, 1045 (1962).

[43] R. Röhlsberger, A. Bernhard, E. Burkel, O. Leupold, and E. Gerdau, HASYLAB Annual Report 1997 (unpublished), p. 933; R. Röhlsberger, T. S. Toellner, W. Sturhahn, K. W. Quast, E. E. Alp, A. Bernhard, E. Burkel, O. Leupold, and E. Gerdau (unpublished).

[44] The Fourier image $\int_0^\infty \exp(i\omega t) K_1^{s\tilde{s}}(t)dt$ of the correlation function (67)

$$K_1^{s\tilde{s}}(\omega)=\sum_l \frac{i\mathcal{A}_l^{s\tilde{s}}\Gamma_0}{\hbar(\tilde{\omega}+\omega)-E_0-\Omega_l\hbar+i\Gamma_0/2}$$

together with Eq. (52) and the relations $\omega=\varepsilon\Gamma_0/\hbar$, $\tilde{\omega}=\tilde{\varepsilon}\Gamma_0/\hbar$ give the well-known frequency transmission function through the nuclear resonance medium with time-independent hyperfine interactions.

[45] E. Gerdau, R. Rüffer, R. Hollatz, and J. P. Hannon, Phys. Rev. Lett. **57**, 1141 (1986).

[46] O. Leupold, A. Bernhard, E. Gerdau, J. Jäschke, H. D. Rüter, Yu. V. Shvyd'ko, H. de Waard, E. E. Alp, P. Hession, M. Hu, W. Sturhahn, J. Sutter, T. Toellner, A. I. Chumakov, J. Metge, and R. Rüffer, Hyperfine Interact. **113**, 81 (1998).

[47] H. Winkler, W. Meyer-Klaucke, S. Schwendy, A. X. Trautwein, B. F. Matzanke, O. Leupold, H. D. Rüter, M. Haas, E. Realo, D. Mandon, and R. Weiss, Hyperfine Interact. **113**, 443 (1998).

[48] L. D. Landau and E. M. Lifshitz, *Quantum Mechanics. Non-Relativistic Theory* (Pergamon, New York, 1970).

[49] In the particular case of pure magnetic interactions that we consider here the frequencies $\Omega_{l'}$ corresponding to the new directions of the magnetic hyperfine field are equal to Ω_l. In general, if the nuclei experience combined magnetic and electric quadrupole interactions these frequencies may change after switching.

[50] Yu. V. Shvyd'ko, Hyperfine Interact. **90**, 287 (1994).

[51] Yu. V. Shvyd'ko, A. I. Chumakov, G. V. Smirnov, T. Hertrich, U. van Bürck, H. D. Rüter, O. Leupold, J. Metge, and E. Gerdau, Europhys. Lett. **26**, 215 (1994).

[52] S. Kikuta, Hyperfine Interact. **90**, 335 (1994).

[53] Yu. V. Shvyd'ko, T. Hertrich, J. Metge, O. Leupold, E. Gerdau, and H. D. Rüter, Phys. Rev. B **52**, R711 (1995).