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Coherent nuclear resonant scattering of X-rays: Time and space picture

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The problem of coherent resonant scattering of X-rays by an ensemble of nuclei is solved directly in time and space. In a first step the problem with a single coherently scattered beam is considered – nuclear forward scattering. The wave equation describing the propagation of the radiation through the nuclear ensemble is derived. It is a first order integro-differential equation. Its kernel is a double time function $K(t,\tilde{t})$ which represents the coherent single scattering response of the nuclear system at time t to excitation at $\tilde{t}$. The kernel is defined by the character of the interactions the nuclei experience with the environment and by the character of their motion. A general procedure of solution of the wave equation is introduced which is independent of the type of kernel. In a second step the wave equation is generalized to the case of many coherently scattered beams, which is, e.g., the case of nuclear Bragg diffraction. Kernels of the wave equations are derived for some particular cases: collective motion of nuclei in space, thermal lattice vibrations, time-independent hyperfine interactions, and time-dependent hyperfine interactions due to external magnetic-field switching.

Keywords: nuclear resonance, X-rays, coherent scattering, time dependence

1. Introduction

Presentation in the frequency domain has usually been used to describe nuclear resonance scattering phenomena. This had natural reasons, since the main experimental technique in use, the Mössbauer spectroscopy, allowed one to measure frequency dependencies of nuclear resonant absorption, emission, or scattering.

In 1986 nuclear resonant spectroscopy in the time domain, which uses pulsed synchrotron radiation as a source, was introduced by Gerdau et al. [1]. This experimental technique has been intensively developed since that time. It is a technique complementary to Mössbauer spectroscopy providing equal, additional or even unique information (for a review see, e.g., [2,3] and articles in this book). Time spectra of the coherent response of nuclear systems to the excitation with synchrotron radiation pulses are measured in cases of nuclear forward scattering (NFS), nuclear Bragg scattering (NBS), grazing incidence, nuclear small angle scattering, etc.

Time spectra of NFS and NBS of synchrotron radiation were first considered theoretically by Kagan et al. [4]. Fourier transformation of the frequency dependences of the transmission and reflection amplitudes, which had been obtained earlier in [5–8], were used to calculate the time spectra of NFS and NBS. Important features which arise in thick samples – speed-up of the nuclear decay and dynamic beats – were considered. Trammell and Hannon [9] pointed for the first time to an important effect which arises in samples possessing multiple nuclear resonances – quantum beats. Numerous subsequent theoretical papers [10–15] have used Fourier transformation from the frequency to the time domain to consider different scattering geometries and different cases of interactions of nuclei with their environment.

The Fourier transformation technique can be used to calculate the time spectra provided the frequency spectra of transmission or (and) reflection are known for the given sample. However, this is not always the case. One approach, which does not require initial knowledge of the frequency spectra, is based on the solution of the scattering problems directly in time and space. The methods of direct calculations in time and space arose originally with the need to describe experiments of nuclear resonance scattering with time-dependent external perturbations of hyperfine interactions [16,17] and nuclear motion [18–20]. This approach was later also used to handle NFS problems in case of time-independent hyperfine interactions [21].

In [22] the technique of evaluation of the NFS time spectra directly in time and space was generalized to nuclear systems experiencing any type of hyperfine interactions and any character of spatial motion. In the present paper this approach is generalized one step further with the purpose of handling problems with several coherently scattered beams as they take place, e.g., in the case of Bragg diffraction.

In the first part of the paper (section 2) the scattering problem of a single coherently scattered beam is considered: nuclear forward scattering. A wave equation is derived for the propagation of X-rays directly in time and space through a homogeneous nuclear resonant medium: sections 2.1, 2.4. The equation is a first order integro-differential 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: section 2.3. $K(t,\tilde{t})$ describes the coherent single scattering response in the forward direction of the nuclear system at time t to an excitation at $\tilde{t}$. A procedure for solving the wave equation is discussed that is independent of the type of the kernel: section 2.5.

In the next part of the paper (section 3) the wave equation is generalized to the case of several coherently scattered beams, which is, e.g., the case of nuclear Bragg scattering. The kernel of the set of multiple beam wave equations is the double-time self-correlation function of the nuclear ensemble $K_{m\widetilde{m}}(t,\tilde{t})$, which bears very similar physical information as in the case of NFS: it is a coherent single scattering response of the nuclear system at time t to the excitation at $\tilde{t}$. However, it additionally acquires an explicit double-momentum dependence due to different wave vectors of the incident $\boldsymbol{k}_{\widetilde{m}}$ and scattered $\boldsymbol{k}_m$ beams. The solution of the set of equations in a single scattering approximation is discussed: section 3.3.

Examples of the nuclear self-correlation functions – the kernels of the wave equations – are given in the following particular cases: collective motion of the nuclear ensemble in space: section 4.1.1; thermal lattice vibrations: section 4.1.2; time-independent hyperfine interactions: section 4.2; time-dependent hyperfine interactions due to external magnetic field switching: section 4.3.

2. Nuclear forward scattering

2.1. *Formulation of the problem*

In this section we will consider the nuclear resonant scattering problem with single coherently scattered X-ray beam: nuclear forward scattering (NFS). The nuclei are supposed to be moving in space, or (and) located in an environment with hyperfine interactions which in general may be time-dependent. 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 purpose of this section is to derive directly in time and space the wave equation of propagation of radiation in such media and to develop a procedure for its solution.

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

$$\nabla^2\boldsymbol{e} - \operatorname{grad}\operatorname{div}\boldsymbol{e} - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\boldsymbol{e} = \frac{4\pi}{c^2}\frac{\partial}{\partial t}\boldsymbol{i} \tag{2.1}$$

with $\boldsymbol{i}(\boldsymbol{r},t)$ the macroscopic current density induced by the radiation in the system of resonant nuclei. The incident radiation is represented as a plane wave modulated in time:

$$\boldsymbol{e}_{\text{in}}(\boldsymbol{r},t) = \boldsymbol{\mathcal{E}}(t)\mathrm{e}^{\mathrm{i}(\tilde{\boldsymbol{k}}\boldsymbol{r}-\widetilde{\omega}t)} \tag{2.2}$$

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

The solution of eq. (2.1) for the field propagating in the sample in the primary beam direction is sought for in the form

$$\boldsymbol{e}(\boldsymbol{r},t) = \boldsymbol{E}(z,t)\mathrm{e}^{\mathrm{i}(\tilde{\boldsymbol{k}}\boldsymbol{r}-\widetilde{\omega}t)} \tag{2.3}$$

with an envelope $\boldsymbol{E}(z,t)$ varying slowly in time and space compared with the exponent. The envelope only contains 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 sought for is of the same form:

$$\boldsymbol{i}(\boldsymbol{r},t) = \boldsymbol{I}(z,t)\mathrm{e}^{\mathrm{i}(\tilde{\boldsymbol{k}}\boldsymbol{r}-\widetilde{\omega}t)}. \tag{2.4}$$

The presentation by eqs. (2.3), (2.4) is known as the slowly varying envelope approximation. In this case the second order Maxwell wave equation (2.1) reduces to a set of first order differential equations for the envelopes $\boldsymbol{E}(z,t)$ and $\boldsymbol{I}(z,t)$:

$$\gamma\frac{\partial}{\partial z}\boldsymbol{E}(z,t) = -\frac{2\pi}{c}\boldsymbol{I}(z,t) \tag{2.5}$$

with $\gamma = \tilde{k}_z/\tilde{k}$. By using (2.2) and (2.3) the boundary conditions for eq. (2.5) read as follows:

$$\boldsymbol{E}(0,t) = \boldsymbol{\mathcal{E}}(t). \tag{2.6}$$

Wave equations for the slowly varying envelopes of the radiation field and induced current density were first introduced in optics (see, e.g., [23]) and in X-ray diffraction in crystals [24]. Recently it was also used in the analysis of nuclear resonant scattering problems [16,18,19,22].

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

$$S(t) \propto \left|\boldsymbol{E}(L,t)\right|^2 = \sum_s \left|E^s(L,t)\right|^2. \tag{2.7}$$

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

2.2. Current density of the nuclear ensemble

In each particular case the right hand side of eq. (2.5), 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) 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

$$\boldsymbol{i}(\boldsymbol{r}, t) = \sum_\alpha \int \frac{\mathrm{d}\boldsymbol{k}}{(2\pi)^3} \left\langle \boldsymbol{i}_\alpha(\boldsymbol{k}, t) \mathrm{e}^{\mathrm{i}\boldsymbol{k}(\boldsymbol{r}-\boldsymbol{r}_\alpha)} \right\rangle. \tag{2.8}$$

The broken brackets $\langle\cdot\rangle$ in eq. (2.8) mean a 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:

$$\boldsymbol{i}_\alpha(\boldsymbol{k}, t) = \mathrm{Tr}\left\{\hat{\boldsymbol{j}}(\boldsymbol{k}) \hat{\rho}_\alpha(t)\right\}. \tag{2.9}$$

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

$$\mathrm{i}\hbar \frac{\partial \hat{\rho}_\alpha(t)}{\partial t} = \left[\widehat{\mathcal{H}}_\alpha(t) + \widehat{\mathcal{H}}_\alpha^\gamma(t), \hat{\rho}_\alpha(t)\right]. \tag{2.10}$$

$\widehat{\mathcal{H}}_\alpha(t)$ in eq. (2.10) is the Hamiltonian of a nucleus α with the following components:

$$\widehat{\mathcal{H}}_\alpha(t) = E_\lambda - \mathrm{i}\frac{\Gamma_0}{2}\delta_{\lambda,\mathrm{e}} + \widehat{\mathcal{H}}^{\mathrm{hf}}_{\alpha_\lambda}(t). \tag{2.11}$$

E_λ is the energy in the ground ($\lambda = \mathrm{g}$) or in the excited ($\lambda = \mathrm{e}$) state defined by internal nuclear interactions. Each nucleus is also characterized by the energy $E_0 = E_\mathrm{e} - E_\mathrm{g}$ of the nuclear transition $\mathrm{e} \Rightarrow \mathrm{g}$, 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 Hamiltonian $\widehat{\mathcal{H}}^{\mathrm{hf}}_{\alpha_\lambda}(t)$ represents hyperfine interactions which, generally speaking, are time-dependent. Its actual form will be specified later.

$\widehat{\mathcal{H}}^{\gamma}_\alpha(t)$ in (2.10) is the Hamiltonian representing the interaction of the nucleus with the radiation field. It is given by the standard expression: $\widehat{\mathcal{H}}^{\gamma}_\alpha(t) = -c^{-1}\int \mathrm{d}\boldsymbol{r}\hat{\boldsymbol{j}}(\boldsymbol{r}-\boldsymbol{r}_\alpha)\boldsymbol{A}(\boldsymbol{r},t)$ [25] with $\boldsymbol{A}(\boldsymbol{r},t)$ the vector potential and with $\hat{\boldsymbol{j}}(\boldsymbol{r}-\boldsymbol{r}_\alpha)$ here and $\hat{\boldsymbol{j}}(\boldsymbol{k})$ in (2.9) 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. (2.3) and applying the slowly varying envelope approximation we obtain for the interaction Hamiltonian [16,18]

$$\widehat{\mathcal{H}}^{\gamma}_\alpha(t) = \mathrm{i}\tilde{\omega}^{-1}\hat{\boldsymbol{j}}\big(-\tilde{\boldsymbol{k}}\big)\boldsymbol{E}(z_\alpha,t)\mathrm{e}^{\mathrm{i}(\tilde{\boldsymbol{k}}\boldsymbol{r}_\alpha - \tilde{\omega}t)}. \tag{2.12}$$

We assume that the interaction (2.12) of the nuclei with the radiation field is weak compared to the other interactions given by (2.11). This allows us to use perturbation theory and to obtain in second order (resonant scattering is a second-order scattering process) the following expression for the current density (see [26]):

$$\boldsymbol{i}_\alpha(\boldsymbol{k},t) = \frac{1}{\mathrm{i}\hbar}\mathrm{Tr}\left\{\check{\boldsymbol{j}}_\alpha(\boldsymbol{k},t)\int_{-\infty}^{t}\big[\check{\mathcal{H}}^{\gamma}_\alpha(\tilde{t}),\check{\rho}_\alpha(-\infty)\big]\,\mathrm{d}\tilde{t}\right\}. \tag{2.13}$$

Here, the symbols with the accent ˇ denote interaction picture operators defined according to the rule

$$\check{\Lambda}_\alpha(t) = \widehat{U}^{-1}_\alpha(t,-\infty)\hat{\Lambda}_\alpha(t)\widehat{U}_\alpha(t,-\infty) \tag{2.14}$$

with the evolution operator

$$\widehat{U}_\alpha(t_2,t_1) = \widehat{T}\exp\left\{-\frac{\mathrm{i}}{\hbar}\int_{t_1}^{t_2}\widehat{\mathcal{H}}_\alpha(t)\,\mathrm{d}t\right\} \tag{2.15}$$

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

The trace in (2.13) is calculated by using a full set of nuclear state vectors $|\alpha_\lambda\rangle$ which, in the present paper, are chosen as eigenstate vectors of the nuclear Hamiltonian $\widehat{\mathcal{H}}_\alpha(-\infty)$ of eq. (2.11) taken at $t = -\infty$. It is assumed that initially at $t = -\infty$ all the sub-levels in the ground state are populated equally, i.e., $\langle\alpha_\mathrm{g}|\hat{\rho}_\alpha(-\infty)|\alpha_\mathrm{g}\rangle = 1/(2I_\mathrm{g}+1)$, while those in the excited state are not populated at all, i.e., $\langle\alpha_\mathrm{e}|\hat{\rho}_\alpha(-\infty)|\alpha_\mathrm{e}\rangle = 0$. 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:

$$i^{s}_{\alpha}(\boldsymbol{k},t) = \frac{1}{\hbar\widetilde{\omega}(2I_{\mathrm{g}}+1)} \sum_{\alpha_{\mathrm{g}},\alpha_{\mathrm{e}},\tilde{s}} j^{s}_{\alpha_{\mathrm{g}}\alpha_{\mathrm{e}}}(\boldsymbol{k},t) \int_{-\infty}^{t} j^{\tilde{s}}_{\alpha_{\mathrm{e}}\alpha_{\mathrm{g}}}\big(-\tilde{\boldsymbol{k}},\tilde{t}\big) E^{\tilde{s}}\big(z_{\alpha},\tilde{t}\big) \mathrm{e}^{\mathrm{i}(\tilde{\boldsymbol{k}}\boldsymbol{r}_{\alpha} - \widetilde{\omega}\tilde{t})}\,\mathrm{d}\tilde{t}. \tag{2.16}$$

The two matrix elements $j^{s}_{\alpha_{\mathrm{g}}\alpha_{\mathrm{e}}}(\boldsymbol{k},t)$ and $j^{\tilde{s}}_{\alpha_{\mathrm{e}}\alpha_{\mathrm{g}}}(-\tilde{\boldsymbol{k}},\tilde{t})$ in (2.16) represent two stages of the nuclear resonance scattering process via one of the possible nuclear transitions $\langle\alpha_{\mathrm{g}}| \Leftrightarrow |\alpha_{\mathrm{e}}\rangle$. The matrix element $j^{\tilde{s}}_{\alpha_{\mathrm{e}}\alpha_{\mathrm{g}}}(-\tilde{\boldsymbol{k}},\tilde{t})$ 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 $j^{s}_{\alpha_{\mathrm{g}}\alpha_{\mathrm{e}}}(\boldsymbol{k},t)$ describes de-excitation of the nucleus at time instant t with emission of the radiation in the polarization state s and with wave vector $\boldsymbol{k}$. The matrix elements $j^{s}_{\alpha_{\mathrm{g}}\alpha_{\mathrm{e}}}(\boldsymbol{k},t)$ and $j^{\tilde{s}}_{\alpha_{\mathrm{e}}\alpha_{\mathrm{g}}}(-\tilde{\boldsymbol{k}},\tilde{t})$ are calculated according to (2.14), (2.15), in particular

$$j^{s}_{\alpha_{\mathrm{g}},\alpha_{\mathrm{e}}}(\boldsymbol{k},t) = \sum_{\tilde{\alpha}_{\mathrm{g}},\tilde{\alpha}_{\mathrm{e}}} U^{-1}_{\alpha_{\mathrm{g}}\tilde{\alpha}_{\mathrm{g}}}(t,-\infty) j^{s}_{\tilde{\alpha}_{\mathrm{g}}\tilde{\alpha}_{\mathrm{e}}}(\boldsymbol{k}) U_{\tilde{\alpha}_{\mathrm{e}}\alpha_{\mathrm{e}}}(t,-\infty). \tag{2.17}$$

For the calculation of the nuclear current density matrix elements $j^{s}_{\alpha_{\mathrm{g}}\alpha_{\mathrm{e}}}(\tilde{\boldsymbol{k}})$ we refer to text books [25,27] and papers [8,12,28,29].

Inserting (2.16) into (2.8) we obtain the following expression for the s-component of the coherent current density induced in the nuclear ensemble:

$$i^{s}(\boldsymbol{r},t) = \int \frac{\mathrm{d}\boldsymbol{k}}{(2\pi)^{3}} \mathrm{e}^{\mathrm{i}(\boldsymbol{k}\boldsymbol{r} - \widetilde{\omega}t)} \sum_{\alpha,\tilde{s}} \mathrm{e}^{\mathrm{i}(\tilde{\boldsymbol{k}}-\boldsymbol{k})\boldsymbol{R}_{\alpha}} \int_{-\infty}^{t} F^{s\tilde{s}}_{\alpha}\big(\boldsymbol{k},\tilde{\boldsymbol{k}},t,\tilde{t}\big) E^{\tilde{s}}\big(z_{\alpha},\tilde{t}\big)\,\mathrm{d}\tilde{t}, \tag{2.18}$$

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

$$\begin{aligned} F^{s\tilde{s}}_{\alpha}\big(\boldsymbol{k},\tilde{\boldsymbol{k}},t,\tilde{t}\big) = {} & \frac{1}{\hbar\widetilde{\omega}(2I_{\mathrm{g}}+1)} \big\langle \mathrm{e}^{-\mathrm{i}\boldsymbol{k}\boldsymbol{u}_{\alpha}(t)} \mathrm{e}^{\mathrm{i}\tilde{\boldsymbol{k}}\boldsymbol{u}_{\alpha}(\tilde{t})} \big\rangle \\ & \times \mathrm{e}^{\mathrm{i}\,\widetilde{\omega}(t-\tilde{t})} \sum_{\alpha_{\mathrm{g}},\alpha_{\mathrm{e}}} \big\langle j^{s}_{\alpha_{\mathrm{g}}\alpha_{\mathrm{e}}}(\boldsymbol{k},t) j^{\tilde{s}}_{\alpha_{\mathrm{e}}\alpha_{\mathrm{g}}}\big(-\tilde{\boldsymbol{k}},\tilde{t}\big) \big\rangle. \end{aligned} \tag{2.19}$$

It is clear from (2.18) as well as from (2.13), (2.16) that $\tilde{t} \leqslant t$, which has the evident meaning that the time of excitation $\tilde{t}$ always precedes the time of de-excitation t.

2.3. Self-correlation function

Let us combine into groups the terms in the sum $\sum_{\alpha}$ of (2.18) with the same values of $F^{s\tilde{s}}_{\alpha}(\boldsymbol{k},\tilde{\boldsymbol{k}},t,\tilde{t})$. 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, etc., are examples of such groups. Thus, in the sum over the whole nuclear ensemble in eq. (2.18) we single out the sums $\sum'_{\alpha}$ 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

$$\sum_{\alpha}{}' \exp\left[\mathrm{i}\left(\tilde{\boldsymbol{k}}-\boldsymbol{k}\right)\boldsymbol{R}_{\alpha}\right] = (2\pi)^3 N_0 w_{\beta}\delta\left(\tilde{\boldsymbol{k}}-\boldsymbol{k}\right), \tag{2.20}$$

where N_0 is the number of all resonant nuclei per unit volume and w_β the relative weight of 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 [30]. As a result we finally obtain the expression for the coherent part of the macroscopic nuclear current density. It has the form of eq. (2.4) with the s-component of the envelope of the macroscopic nuclear current density given by

$$I^{s}(z,t) = \frac{c}{2\pi}\frac{\sigma_{\mathrm{R}}N_0\Gamma_0}{4\hbar}\int_{-\infty}^{t}\sum_{\tilde{s}} K^{s\tilde{s}}\left(t,\tilde{t}\right)E^{\tilde{s}}\left(z,\tilde{t}\right)\mathrm{d}\tilde{t}. \tag{2.21}$$

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

$$K^{s\tilde{s}}\left(t,\tilde{t}\right) = \sum_{\beta} L_{\beta}^{s\tilde{s}}\left(t,\tilde{t}\right)M_{\beta}\left(t,\tilde{t}\right), \tag{2.22}$$

$$L_{\beta}^{s\tilde{s}}\left(t,\tilde{t}\right) = \mathrm{e}^{\mathrm{i}\,\widetilde{\omega}(t-\tilde{t})}X_{\beta}\sum_{\beta_{\mathrm{g}},\beta_{\mathrm{e}}}\left\langle j_{\beta_{\mathrm{g}}\beta_{\mathrm{e}}}^{s}\left(\tilde{\boldsymbol{k}},t\right)j_{\beta_{\mathrm{e}}\beta_{\mathrm{g}}}^{\tilde{s}}\left(-\tilde{\boldsymbol{k}},\tilde{t}\right)\right\rangle, \tag{2.23}$$

$$M_{\beta}\left(t,\tilde{t}\right) = \left\langle \mathrm{e}^{-\mathrm{i}\tilde{\boldsymbol{k}}\boldsymbol{u}_{\beta}(t)}\mathrm{e}^{\mathrm{i}\tilde{\boldsymbol{k}}\boldsymbol{u}_{\beta}(\tilde{t})}\right\rangle, \tag{2.24}$$

$$X_{\beta} = \frac{4\tilde{k}}{c^2(2I_{\mathrm{e}}+1)\Gamma_{\gamma}}w_{\beta}. \tag{2.25}$$

The factor

$$\sigma_{\mathrm{R}} = \frac{4\pi}{\tilde{k}^2}\cdot\frac{2I_{\mathrm{e}}+1}{2(2I_{\mathrm{g}}+1)}\cdot\frac{\Gamma_{\gamma}}{\Gamma_0} \tag{2.26}$$

in (2.21) is the cross-section of the nuclear resonance absorption. Γ_γ in (2.25) is the full radiative width of the nuclear transition e $\Rightarrow$ g.

The function $L_{\beta}^{s\tilde{s}}(t,\tilde{t})$ in (2.23) can be presented in an alternative way:

$$L_{\beta}^{s\tilde{s}}\left(t,\tilde{t}\right) = \mathrm{e}^{\mathrm{i}\,\widetilde{\omega}(t-\tilde{t})}X_{\beta}\sum_{\beta_{\mathrm{g}},\beta_{\mathrm{e}}}\left\langle\left[U_{\beta}^{-1}\left(t,\tilde{t}\right)j^{s}\left(\tilde{\boldsymbol{k}}\right)U_{\beta}\left(t,\tilde{t}\right)\right]_{\beta_{\mathrm{g}}\beta_{\mathrm{e}}}\right\rangle j_{\beta_{\mathrm{e}}\beta_{\mathrm{g}}}^{\tilde{s}}\left(-\tilde{\boldsymbol{k}}\right). \tag{2.27}$$

For this we have used eqs. (2.17) and the composition law $\widehat{U}(t_3,t_1) = \widehat{U}(t_3,t_2)\widehat{U}(t_2,t_1)$ valid for the evolution operators [31].

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

$$K^{s\tilde{s}}(t,t) = \delta^{s\tilde{s}}, \tag{2.28}$$

which can be easily verified by taking into account that according to (2.24) $M_\beta(t,\tilde{t}\,) = 1$ independent of how the nuclei are moving, that according to (2.15) $U_\beta(t,\tilde{t}\,) = 1$ independent of the type of hyperfine interactions, and by the relation

$$\sum_{\beta_{\rm g},\beta_{\rm e}} j^{s}_{\beta_{\rm g}\beta_{\rm e}}(\tilde{\boldsymbol{k}}\,)\, j^{\tilde{s}}_{\beta_{\rm e}\beta_{\rm g}}(-\tilde{\boldsymbol{k}}\,) = \delta^{s\tilde{s}}\frac{\Gamma_\gamma(2I_{\rm e}+1)c^2}{4\tilde{k}}. \tag{2.29}$$

The latter can be proved by using the optical theorem.

The function $M_\beta(t,\tilde{t})$ in eqs. (2.22), (2.24) represents spatial motion of the nuclei belonging to a group β, while $L^{s\tilde{s}}_{\beta}(t,\tilde{t})$ in eqs. (2.22), (2.23) 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 [32] in connection with neutron scattering. It was later used by Singwi and Sjölander [33] 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 [34] and Blume and Tjon [35] to evaluate the Mössbauer line shape in the presence of time-dependent hyperfine fields.

The self-correlation functions typically used had a $(t-\tilde{t})$ dependence. The function defined by eqs. (2.22)–(2.24) contains both time variables separated. As shown in section 2.6.4 the self-correlation functions $K(t,\tilde{t})$ which are reduced to $\mathcal{K}(t-\tilde{t})$ describe coherent *elastic* scattering, while the self-correlation function $K(t,\tilde{t})$ with separated time variables describes coherent *inelastic* scattering.

Elastic scattering occurs under the conditions of time-independent hyperfine interactions, as discussed in section 4.2. It may also occur under the conditions of time-dependent hyperfine interactions or if the positions of nuclei in space are changing in time. 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 a $(t-\tilde{t})$ dependence of the self-correlation function. Thermal vibrations of nuclei in space (section 4.1.2) is such an example.

Nonstochastic variations in time usually cause inelastic scattering. It occurs if, e.g., collective synchronized motion of nuclei in space takes place (section 4.1.1), or synchronized time-dependent hyperfine interactions are induced (section 4.3).

2.4. NFS wave equation

By using eqs. (2.5), (2.21) we obtain the wave equation for propagation of X-rays in the nuclear resonant medium:

$$\frac{\partial E^{s}(\xi,\tau)}{\partial\xi} = -\int_{-\infty}^{\tau}\sum_{\tilde{s}} K^{s\tilde{s}}(\tau,\tilde{\tau})\,E^{\tilde{s}}(\xi,\tilde{\tau})\,{\rm d}\tilde{\tau}. \tag{2.30}$$

Here dimensionless space and time variables

$$\xi = \frac{1}{4}\sigma_{\rm R}N_0\frac{z}{\gamma}, \qquad \tau = \frac{t\Gamma_0}{\hbar} \tag{2.31}$$

are used. The dimensionless space variable ξ scales with the so-called effective resonance thickness $T_{\mathrm{R}} = \sigma_{\mathrm{R}} N_0 L$. The boundary condition of the wave equation (2.30) is given by (2.6).

The wave equation (2.30) can also be represented in short as follows:

$$\frac{\partial \boldsymbol{E}(\xi,\tau)}{\partial \xi} = -\widehat{D}\big(\tau,\tilde{\tau}\big)\boldsymbol{E}\big(\xi,\tilde{\tau}\big), \tag{2.32}$$

$$\widehat{D}\big(\tau,\tilde{\tau}\big) = \int_{-\infty}^{\tau} \widehat{K}\big(\tau,\tilde{\tau}\big)\,\mathrm{d}\tilde{\tau}. \tag{2.33}$$

Equation (2.32) is formally 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 $-\mathrm{i}\widehat{D}$ plays the role of the "time"- (i.e., ξ-independent) Hamiltonian. Different techniques of solving the Schrödinger equation exist. To solve (2.32) we use a method similar to that of quantum-mechanical perturbation theory.

2.5. General solution of the NFS wave equation

The general solution of the wave equation (2.30) can be given as a power series of ξ:

$$\boldsymbol{E}(\xi,\tau) = \sum_{p=0}^{\infty} \frac{(-\xi)^p}{p!}\boldsymbol{E}^{(p)}(\tau). \tag{2.34}$$

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

$$\boldsymbol{E}^{(0)}(\tau) = \boldsymbol{\mathcal{E}}(\tau). \tag{2.35}$$

The other terms, $\boldsymbol{E}^{(p)}(\tau)$, can be obtained after substitution of eq. (2.34) into eq. (2.32). This results in the recursion relation

$$\boldsymbol{E}^{(p)}(\tau) = \widehat{D}\big(\tau,\tilde{\tau}\big)\boldsymbol{E}^{(p-1)}(\tilde{\tau}) \tag{2.36}$$

with the help of which and of eqs. (2.33), (2.35) all the amplitudes $\boldsymbol{E}^{(p)}(\tau)$ in eq. (2.34) can be calculated. The $\boldsymbol{E}^{(p)}(\tau)$ will be referred to as multiple scattering amplitudes of order p.

By using eqs. (2.34)–(2.36) the solution of the wave equation can also be represented in the following compact form:

$$\boldsymbol{E}(\xi,\tau) = \exp\big[-\xi\widehat{D}\big(\tau,\tilde{\tau}\big)\big]\boldsymbol{\mathcal{E}}\big(\tilde{\tau}\big). \tag{2.37}$$

It is remarkable that the solution of the wave equation, given by eqs. (2.34)–(2.36), is general and independent of the explicit form of the nuclear interactions, which are hidden in the kernel $K^{s\tilde{s}}(\tau,\tilde{\tau})$. Equations (2.34)–(2.36) 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. (2.22)–(2.27). Then, to calculate the time response given by (2.34), one applies the general procedure based on the recursion relations (2.36). Such a procedure was implemented in the program MOTIF [36].

In a few particular cases one can obtain analytical solutions.

2.6. *Particular solutions of the NFS wave equation*

2.6.1. *Single scattering approximation*

One such case 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 (2.34). By using eqs. (2.33)–(2.36) the radiation field in this approximation reads

$$E^s(\xi,\tau) = \mathcal{E}^s(\tau) - \xi \int_{-\infty}^{\tau} \sum_{\tilde{s}} K^{s\tilde{s}}(\tau,\tilde{\tau})\mathcal{E}^{\tilde{s}}(\tilde{\tau})\,\mathrm{d}\tilde{\tau}. \tag{2.38}$$

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

$$\boldsymbol{\mathcal{E}}(\tau) = \boldsymbol{e}^0\mathcal{E}\delta(\tau), \tag{2.39}$$

which corresponds to the conditions of experiments with pulsed synchrotron radiation, then eq. (2.38) simplifies to

$$E^s(\xi,\tau) = \delta(\tau)\delta^{s0} - \xi K^{s0}(\tau,0). \tag{2.40}$$

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. (2.40) and everywhere in the following that $\mathcal{E} = 1$.

The nuclear response is given by the second term of eq. (2.40). The single scattering solution (2.40) 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.

2.6.2. *Immediate response*

At $t = 0+$ (i.e., immediately after the excitation) the solution (2.40) 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. Let us calculate this immediate response.

By using eqs. (2.40), (2.28) and (2.31) we obtain for the response in the forward direction immediately after excitation

$$E^s(\xi,0+) = -\xi = -\frac{\sigma_{\mathrm{R}}N_0L}{4\gamma}\delta^{s0}. \tag{2.41}$$

The immediate response of the nuclear system to instantaneous excitation given by eq. (2.41) 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 nuclear motion, which is discussed in [22] and in section 4.1.2.

2.6.3. 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: $\widehat{\mathcal{H}}^{\mathrm{hf}}_{\beta_\lambda} = 0$. In this case the evolution operator (2.15) is a c-value:

$$U_\beta\bigl(t,\tilde{t}\,\bigr) = \exp\left[-\frac{\mathrm{i}}{\hbar}\left(E_\lambda - \mathrm{i}\frac{\Gamma_0}{2}\delta_{\lambda,\mathrm{e}}\right)\bigl(t-\tilde{t}\,\bigr)\right],$$

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. (2.22)–(2.27) we obtain that the single resonance correlation function is elastic: $K^{s\tilde{s}}(\tau,\tilde{\tau}) = \mathcal{K}^{s\tilde{s}}(\tau-\tilde{\tau})$, and given by

$$\mathcal{K}^{s\tilde{s}}(\tau) = \delta^{s\tilde{s}}\psi(\tau), \tag{2.42}$$

where

$$\psi(\tau) = \exp\left[\mathrm{i}\left(\frac{\hbar\widetilde{\omega}-E_0}{\Gamma_0}+\frac{\mathrm{i}}{2}\right)\tau\right]\theta(\tau), \tag{2.43}$$

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

There is no polarization mixing or 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 (2.39). As a result, by using eqs. (2.33)–(2.36) and the self-correlation function (2.42) 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:

$$E(\xi,\tau) = \delta(\tau) - \psi(\tau)\xi\sum_{p=1}^{\infty}\frac{(-\xi\tau)^{p-1}}{(p-1)!p!} \tag{2.44}$$

$$= \delta(\tau) - \psi(\tau)\xi\frac{\mathrm{J}_1(2\sqrt{\xi\tau})}{\sqrt{\xi\tau}}. \tag{2.45}$$

The solution (2.45) is in agreement with the result obtained earlier in [4] by using Fourier transformation of the frequency-dependent transmission 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 [37,38].

The terms in the sum of (2.44) are the multiple scattering amplitudes of 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 from the excitation time $\tau = 0$) the solution (2.44) can be restricted to the term with $p = 1$. This actually represents the single scattering approximation discussed in section 2.6.1. 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 from $\tau = 0$, then other terms in the sum of (2.44), which represent multiple scattering, start to play a significant role. As a result, according to (2.45), the response acquires the modulation $\mathrm{J}_1(2\sqrt{\xi\tau})/\sqrt{\xi\tau}$ which alters the natural decay by additional damping and oscillations. The latter are often referred to as dynamical beats, which were observed both in experiments with radioactive sources [16,39,40] and with synchrotron radiation [41]. The faster damping is often referred to as coherent speed-up of the decay in NFS [4,40,42,43].

How many scattering events in a sample with thickness parameter ξ does a photon arriving in the detector at time τ experience? In other words, how many terms in the sum of (2.44) 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 $p! = \sqrt{2\pi}p^{p+0.5}\mathrm{e}^{-p}$ for factorials in the denominator of (2.44). Under these assumptions the terms in the sum of (2.44) can be approximated by $(\xi\tau\mathrm{e}^2/p^2)^{p-1}/\sqrt{2\pi}$. Thus, the required number of scattering events can be estimated to be $p_{\xi\tau} \geqslant \mathrm{e}\sqrt{\xi\tau}$.

By using solution (2.45) and the wave equation (2.5) one can also obtain the following expression for the coherent nuclear current density induced in the sample by the instantaneous radiation pulse (2.39):

$$I(\xi,\tau) = \frac{c}{8\pi} N_0 \sigma_{\mathrm{R}} \mathrm{J}_0\bigl(2\sqrt{\xi\tau}\,\bigr)\psi(\tau). \tag{2.46}$$

It shows that directly after the excitation at $\tau = 0$ there exists a homogeneous distribution of the nuclear currents over the whole sample: $I(\xi, 0+) = \text{const}$. Later the nuclear currents acquire an inhomogeneous spatial distribution which varies with time. According to eq. (2.5) the nuclear ensemble radiates in the forward direction a field $E(L,\tau) = -(2\pi/c\gamma)\int_0^L I(z,\tau)\,\mathrm{d}z$. At those instants, when the net coherent current density $\int_0^L I(z,\tau)\,\mathrm{d}z = 0$, i.e., when the currents interfere destructively, the nuclei do not radiate coherently in a forward direction. These instants correspond to the minima of the dynamical beats.

2.6.4. Particular solutions in the frequency domain

An alternative solution procedure to that given by eqs. (2.34)–(2.36) is to transform the wave equation (2.32) 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

$$\frac{\partial \boldsymbol{E}(\xi,\varepsilon)}{\partial \xi} = -\int_{-\infty}^{+\infty} \frac{\mathrm{d}\tilde{\varepsilon}}{2\pi} \widehat{K}\bigl(\varepsilon,\tilde{\varepsilon}\bigr)\boldsymbol{E}\bigl(\xi,\tilde{\varepsilon}\bigr), \tag{2.47}$$

where

$$\widehat{K}(\varepsilon, \tilde{\varepsilon}) = \int\int \mathrm{e}^{\mathrm{i}\varepsilon\tau} \mathrm{e}^{-\mathrm{i}\tilde{\varepsilon}\tilde{\tau}} \widehat{K}(\tau, \tilde{\tau})\, \mathrm{d}\tau\, \mathrm{d}\tilde{\tau}. \tag{2.48}$$

The set of eqs. (2.47) describes, generally speaking, the inelastic coherent forward scattering with the matrix operator $\widehat{K}(\varepsilon, \tilde{\varepsilon})$ representing the scattering of the radiation from the state with frequency $\widetilde{\omega} + \tilde{\varepsilon}\Gamma_0/\hbar$ to another state with frequency $\widetilde{\omega} + \varepsilon\Gamma_0/\hbar$. Only for some special types of kernels $\widehat{K}(\varepsilon, \tilde{\varepsilon})$ it is possible to obtain the solution of (2.47) (see, e.g., [18] 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

$$\widehat{K}\big(\varepsilon, \tilde{\varepsilon}\big) = \widehat{\mathcal{K}}(\varepsilon) 2\pi\delta\big(\varepsilon - \tilde{\varepsilon}\big). \tag{2.49}$$

In the time domain this corresponds to the relation

$$\widehat{K}\big(\tau, \tilde{\tau}\big) = \widehat{\mathcal{K}}\big(\tau - \tilde{\tau}\big), \tag{2.50}$$

which is verified by substituting (2.50) into (2.48). In this case the integral in eq. (2.47) vanishes and the integro-differential equation reduces to the linear differential equation

$$\frac{\partial \boldsymbol{E}(\xi, \varepsilon)}{\partial \xi} = -\widehat{\mathcal{K}}(\varepsilon)\boldsymbol{E}(\xi, \varepsilon), \tag{2.51}$$

the solution of which can be obtained in general form (see, e.g., [8,44]):

$$\boldsymbol{E}(\xi, \varepsilon) = \exp\big[-\xi\widehat{\mathcal{K}}(\varepsilon)\big]\boldsymbol{\mathcal{E}}(\varepsilon), \tag{2.52}$$

with $\boldsymbol{\mathcal{E}}(\varepsilon)$ the frequency spectrum of the incident radiation. The time dependence of NFS is obtained by reverse Fourier transformation [4,12–15].

3. Nuclear Bragg diffraction

In Bragg diffraction several coherently scattered beams arise. The method of evaluation of the time spectra of nuclear resonance scattering with a single coherent beam, presented in the previous sections, will now be generalized to the case of several coherently scattered beams.

As in the case of NFS the problem is solved by using Maxwell's wave equation (2.1). The incident beam is given as before by eq. (2.2). Our sample now is a single crystal plate, containing resonant nuclei. Taking into account the periodicity of the crystal lattice we represent the radiation field inside the crystal as a Bloch wave

$$\boldsymbol{e}(\boldsymbol{r}, t) = \sum_m \boldsymbol{E}_m(z, t)\mathrm{e}^{\mathrm{i}(\boldsymbol{k}_m\boldsymbol{r} - \widetilde{\omega}t)}, \tag{3.1}$$

which is a sum of plane wave components with wave vectors $\boldsymbol{k}_m = \boldsymbol{k}_0 + \boldsymbol{h}_m$. Here, $\boldsymbol{k}_0 \equiv \tilde{\boldsymbol{k}}$ is the wave vector of the incident radiation as in section 2.1. The sum in (3.1) includes all reciprocal lattice vectors $\boldsymbol{h}_m$ of the crystal. In frames of the linear response the induced current density is also a Bloch wave:

$$\boldsymbol{i}(\boldsymbol{r},t) = \sum_m \boldsymbol{I}_m(z,t)\mathrm{e}^{\mathrm{i}(\boldsymbol{k}_m\boldsymbol{r}-\widetilde{\omega}t)}. \tag{3.2}$$

Equations (3.1)–(3.2) are generalization of eqs. (2.3), (2.4) to the multiple beam case. For the same reasons as in section 2.1 we assume that $\boldsymbol{E}_m(z,t)$ and $\boldsymbol{I}_m(z,t)$ vary slowly compared to the fast exponential functions (slowly varying envelope approximation). In this approximation the second order Maxwell wave equation (2.1) reduces to a set of first order differential equations for $\boldsymbol{E}_m(z,t)$ and $\boldsymbol{I}_m(z,t)$:

$$\left(\gamma_m\frac{\partial}{\partial z} + \mathrm{i}\frac{\tilde{k}}{2}a_m\right)\boldsymbol{E}_m(z,t) = -\frac{2\pi}{c}\boldsymbol{I}_m(z,t), \tag{3.3}$$

where

$$a_m = \frac{k_m^2 - \tilde{k}^2}{\tilde{k}^2}, \qquad \gamma_m = \frac{k_{mz}}{\tilde{k}}, \tag{3.4}$$

with k_{mz} the z component of $\boldsymbol{k}_m$.

A set of equations of this type was introduced by Takagi [24] to handle problems of X-ray diffraction in spatially inhomogeneous crystals. It was also introduced in [19] to treat time-dependent nuclear diffraction problems.

The number of equations in (3.3) is equal to the number of amplitudes $\boldsymbol{E}_m$ to be considered. The latter is determined by the number of parameters a_m with values close to zero. In other words, it is equal to the number of reciprocal lattice points which lie near the Ewald sphere ($|\boldsymbol{k}_m| \simeq \tilde{k}$) and obey the Bragg law.

3.1. Nuclear current density

The components of the induced nuclear current density $\boldsymbol{I}_m(z,t)$ are evaluated in a very similar way as in section 2.2. One has only to bear in mind two additional points. Firstly, the radiation field in the crystal is now given by a sum of plane waves (3.1) instead of the single plane wave (2.3), which modifies, e.g., the expression for the interaction Hamiltonian (2.12). The sum of the plane waves should be taken there. Secondly, the sum of (2.20) is now proportional to $\sum_{\widetilde{m}}\delta(\boldsymbol{k}_{\widetilde{m}} - \boldsymbol{k})$ rather than to $\delta(\tilde{\boldsymbol{k}} - \boldsymbol{k})$. Taking these facts into account we arrive at the following expressions for the components of the induced nuclear current density in the crystal (cf. similar expression (2.21) for the case of a single coherent beam):

$$I_m^s(z,t) = \frac{c}{2\pi}\frac{\sigma_{\mathrm{R}}N_0\Gamma_0}{4\hbar}\int_{-\infty}^{t}\sum_{\widetilde{m},\tilde{s}} K_{m\widetilde{m}}^{s\tilde{s}}(t,\tilde{t}\,)E_{\widetilde{m}}^{\tilde{s}}(z,\tilde{t}\,)\,\mathrm{d}\tilde{t}. \tag{3.5}$$

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

$$K^{s\tilde{s}}_{m\widetilde{m}}(t,\tilde{t}) = \sum_{\beta} L^{s\tilde{s}}_{m\widetilde{m}(\beta)}(t,\tilde{t}) M_{m\widetilde{m}(\beta)}(t,\tilde{t}), \tag{3.6}$$

$$L^{s\tilde{s}}_{m\widetilde{m}(\beta)}(t,\tilde{t}) = \mathrm{e}^{\mathrm{i}\widetilde{\omega}(t-\tilde{t})} X_{\beta} \sum_{\beta_{\mathrm{g}},\beta_{\mathrm{e}}} \left\langle j^{s}_{\beta_{\mathrm{g}}\beta_{\mathrm{e}}}(\boldsymbol{k}_m, t) j^{\tilde{s}}_{\beta_{\mathrm{e}}\beta_{\mathrm{g}}}(-\boldsymbol{k}_{\widetilde{m}}, \tilde{t}) \right\rangle, \tag{3.7}$$

$$M^{s\tilde{s}}_{m\widetilde{m}(\beta)}(t,\tilde{t}) = \left\langle \mathrm{e}^{-\mathrm{i}\boldsymbol{k}_m \boldsymbol{u}_{\beta}(t)} \mathrm{e}^{\mathrm{i}\boldsymbol{k}_{\widetilde{m}} \boldsymbol{u}_{\beta}(\tilde{t})} \right\rangle. \tag{3.8}$$

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

$$K^{s\tilde{s}}_{m\widetilde{m}}(t,t) = \delta^{s\tilde{s}}. \tag{3.9}$$

The self-correlation function $K^{s\tilde{s}}_{m\widetilde{m}}(t,\tilde{t})$ given by eqs. (3.6)–(3.8) is a generalization of the self-correlation function $K^{s\tilde{s}}(t,\tilde{t})$ introduced in the case of nuclear forward scattering eqs. (2.22)–(2.27). The correlation function $K^{s\tilde{s}}_{m\widetilde{m}}(t,\tilde{t})$ acquires an explicit double-momentum dependence, specified by indices m and $\widetilde{m}$, since now the wave vectors of the incident $\boldsymbol{k}_{\widetilde{m}}$ and scattered $\boldsymbol{k}_m$ waves have to be distinguished. If they are equal, the function $K^{s\tilde{s}}_{m\widetilde{m}}(t,\tilde{t}) \equiv K^{s\tilde{s}}(t,\tilde{t})$ of (2.22)–(2.27) with $\tilde{\boldsymbol{k}} = \boldsymbol{k}_m$.

3.2. *Multiple beam wave equation*

By using (3.3), (3.5) the set of wave equations for the components of the radiation field in the crystal can be written as

$$\left(\gamma_m \frac{\partial}{\partial z} + \mathrm{i}\frac{\tilde{k}}{2} a_m\right) E^{s}_{m}(z,\tau) = -\frac{\sigma_{\mathrm{R}} N_0}{4} \int_{-\infty}^{\tau} \sum_{\widetilde{m},\tilde{s}} K^{s\tilde{s}}_{m\widetilde{m}}(\tau,\tilde{\tau}) E^{\tilde{s}}_{\widetilde{m}}(z,\tilde{\tau})\,\mathrm{d}\tilde{\tau}. \tag{3.10}$$

In the general case of multiple diffraction in a perfect crystal the boundary conditions for the set of eqs. (3.10) have the following form:

$$\begin{aligned} \boldsymbol{E}_0(0,t) &= \boldsymbol{\mathcal{E}}(t) && \text{(forward beam)}, \\ \boldsymbol{E}_m(0,t) &= 0 \quad \text{if } \gamma_m > 0 && \text{(Laue reflected beams)}, \\ \boldsymbol{E}_m(L,t) &= 0 \quad \text{if } \gamma_m < 0 && \text{(Bragg reflected beam)}. \end{aligned} \tag{3.11}$$

The wave equations (3.10) together with the boundary conditions (3.11) allow us to evaluate the time spectrum $S_m(t) \propto |\boldsymbol{E}_m(z,t)|^2$ of any coherently scattered beam. The general case of multiple diffraction discussed here includes forward scattering ($m = 0$) and two-beam diffraction ($m = 0, 1$) with one diffracted beam as particular cases. The solution of the wave equation with one coherently scattered beam was discussed in section 2.5. A procedure for the numerical solution of the wave equation (3.10) in the case of two-beam diffraction was implemented in the program NERT [45].

3.3. Solution in the single scattering approximation

In this paper we will confine ourselves to the analysis of the solution of the multiple beam wave equation (3.10) in the single scattering approximation. By doing this we will assume that the incident radiation pulse is very short, as given by eq. (2.39). In the zeroth approximation we take $E^s_m(z,\tau) = \delta^{s0}\delta_{m0}\delta(\tau)$. Inserting this into the right-hand side of (3.10) we obtain the equation for the radiation field amplitudes in the single scattering approximation:

$$\left(\gamma_m \frac{\partial}{\partial z} + \mathrm{i}\frac{\tilde{k}}{2}a_m\right) E^s_m(z,\tau) = -\frac{\sigma_{\mathrm{R}} N_0}{4} K^{s0}_{m0}(\tau,0). \tag{3.12}$$

Its solution is

$$E^s_m\big(z^*,\tau\big) = \mathrm{i}\frac{\sigma_{\mathrm{R}} N_0}{2\tilde{k}a_m}\left[1 - \exp\left(-\mathrm{i}\frac{\tilde{k}a_m}{2\gamma_m}z^*\right)\right] K^{s0}_{m0}(\tau,0), \tag{3.13}$$

where $z^* = z$ for the forward and Laue-reflected beams, and $z^* = L - z$ for the Bragg-reflected beams.

If the Bragg condition for the reciprocal vector $\boldsymbol{h}_m$ is fulfilled, i.e., $a_m \ll 1/(L\tilde{k})$, eq. (3.13) reduces to

$$E^s_m\big(z^* = L,\tau\big) = -\frac{\sigma_{\mathrm{R}} N_0 L}{4\gamma_m} K^{s0}_{m0}(\tau,0), \tag{3.14}$$

which is valid both for transmitted, Laue- and Bragg-reflected beams.

The single scattering solutions in the form of (3.13), (3.14) provide a clear physical interpretation of the kernel $K^{s\tilde{s}}_{m\widetilde{m}}(\tau,\tilde{\tau})$: the kernel gives the coherent single scattering response of the nuclear system in the direction $\boldsymbol{k}_m$ at time t to the excitation at $\tilde{t}$ with radiation of wave vector $\boldsymbol{k}_{\widetilde{m}}$. The superscripts $\tilde{s}$ and s represent the states of polarization of the incoming and scattered radiation.

4. Kernels in particular cases

In each particular case the nuclear self-correlation function (2.22)–(2.27) – the kernel of the NFS wave equation (2.30) – should be calculated. For the rest the evaluation procedure of the time spectrum of NFS is standard and given by eqs. (2.7), (2.34)–(2.36). Similar arguments are valid in case of multiple beam scattering, and concern the self-correlation function $K^{s\tilde{s}}_{m\widetilde{m}}(t,\tilde{t})$ (3.6)–(3.8), which is the kernel of the multiple beam wave equation (3.10).

As was already discussed, the self-correlation function $K^{s\tilde{s}}(t,\tilde{t})$ is a particular case of $K^{s\tilde{s}}_{m\widetilde{m}}(t,\tilde{t})$. The latter reduces to $K^{s\tilde{s}}(t,\tilde{t})$, provided the wave vectors of the incident and scattered waves are equal: $\boldsymbol{k}_m = \boldsymbol{k}_{\widetilde{m}}$. Unless mentioned we shall further discuss the self-correlation function in its general form, given by (3.6)–(3.8).

In sections 2.6.1 and 3.3 it was pointed out that the amplitude of the time spectrum in single scattering approximation is proportional to the self-correlation function.

The self-correlation function thus shows important features of the corresponding time spectrum without further solution of the multiple scattering wave equations (2.30) or (3.10).

The explicit form of the self-correlation function is determined by the trajectory of motion $\boldsymbol{u}_\beta(t)$ which enters (2.24), (3.8), and the evolution operator $\widehat{U}_\beta$ (2.15) which enters (2.27), (2.17), (3.7). The evolution operator in turn is specified by the Hamiltonian $\widehat{\mathcal{H}}_\beta(t)$ (2.11) and essentially by the type of hyperfine interactions $\widehat{\mathcal{H}}^{\mathrm{hf}}_{\beta_\lambda}(t)$.

In this section we calculate the kernels for different particular cases of nuclear motion and hyperfine interactions. We first consider the influence of nuclear motion in space (section 4.1). As a next step we consider time-independent hyperfine interactions (section 4.2), and time-dependent hyperfine interactions in the particular case of switching the direction of the magnetic hyperfine field (section 4.3).

4.1. Motion in space

In this section we discuss the effect of motion in space only and therefore do not specify the part of the self-correlation function $L^{s\tilde{s}}_{m\widetilde{m}(\beta)}(t,\tilde{t})$ representing other degrees of freedom. According to definition (3.8) the effect of resonant nuclei in motion on coherent scattering 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 de-excitation. These positions enter the phase factors $\exp[-\mathrm{i}\boldsymbol{k}_m\boldsymbol{u}_\beta(t)]\exp[+\mathrm{i}\boldsymbol{k}_{\widetilde{m}}\boldsymbol{u}_\beta(\tilde{t})]$ in the motional part $M_{m\widetilde{m}(\beta)}(t,\tilde{t})$ of the self-correlation function (3.8).

4.1.1. Collective motion

Let us first consider a simple case where the nuclei are moving as a rigid ensemble with a single displacement vector $\boldsymbol{u}_\beta(t) = \boldsymbol{u}(t)$.[1] The self-correlation function (3.6)–(3.8) becomes

$$K^{s\tilde{s}}_{m\widetilde{m}}\big(t,\tilde{t}\big) = \mathrm{e}^{-\mathrm{i}\boldsymbol{k}_m\boldsymbol{u}(t)}\mathrm{e}^{+\mathrm{i}\boldsymbol{k}_{\widetilde{m}}\boldsymbol{u}(\tilde{t})}\sum_\beta L^{s\tilde{s}}_{m\widetilde{m}(\beta)}\big(t,\tilde{t}\big). \tag{4.1}$$

No statistical averaging of the motional part is required in this case. The same nuclear ensemble but at rest, $\boldsymbol{u}(t) = \mathrm{const} = 0$, is characterized by the self-correlation function $K_{m\widetilde{m}}(t,\tilde{t}) = \sum_\beta L^{s\tilde{s}}_{m\widetilde{m}(\beta)}(t,\tilde{t})$. Let us assume that $\boldsymbol{E}^{(0)}_m(z,t)$ are the solutions of the wave equation (3.10) with such a kernel. Then the solution of the wave equation for the moving system with the self-correlation function (4.1) is given by

$$\boldsymbol{E}_m(z,t) = \boldsymbol{E}^{(0)}_m(z,t)\mathrm{e}^{-\mathrm{i}\boldsymbol{k}_m\boldsymbol{u}(t)}. \tag{4.2}$$

This is verified by substituting (4.1)–(4.2) into (3.10). Thus, according to (4.2), the collective motion of all nuclei produces a phase modulation of the re-emitted radiation. If, e.g., the sample is moving with constant velocity $\boldsymbol{v}$, so that $\boldsymbol{u}(t) = \boldsymbol{v}t$, the

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[1] We restrict ourselves to the case of translational motion, which does not change the direction of re-emission, opposite to rotational motion, which does cause such a change – the lighthouse effect [46].

phase factor in (4.2) is given by $\exp(-\mathrm{i}\Omega_{\mathrm{D}}t)$, where $\Omega_{\mathrm{D}} = \tilde{\boldsymbol{k}}\boldsymbol{v}$ is simply a Doppler frequency shift. For more complicated motions the phase modulation is more sophisticated.

To reveal the phase modulation (4.2) due to collective motion one has to use a phase-sensitive detector. Since the time spectrum recorded with a usual non-phase-sensitive detector is proportional to the modulo square of the nuclear response (4.2), the phase information disappears. Thus, customarily the time spectrum of nuclear resonance scattering 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 [47]. It was also proved theoretically in [19] for the general case of multiple Bragg diffraction. To detect the phase modulation another nuclear resonance scatterer can be used, playing the role of the phase-sensitive detector [47,48].

4.1.2. 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., [49]). Each normal mode, or each phonon in quantum mechanical language, is characterized by its momentum $\boldsymbol{q}$, branch number ν, dispersion law $\omega_{\nu}(\boldsymbol{q})$ and state of polarization $\boldsymbol{e}_{\boldsymbol{q}\nu}(\beta)$. The phonon occupation numbers $\bar{n}_{\boldsymbol{q}\nu}$ 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 the self-correlation function (2.24), (3.8). Here, we consider only elastic scattering, 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. (3.8) over the phonon occupation numbers. The result is well known (see, e.g., [50,49]). We reproduce it here without derivation:

$$\begin{aligned} M_{m\widetilde{m}(\beta)}\bigl(t-\tilde{t}\bigr) &= \bigl\langle \mathrm{e}^{-\mathrm{i}\boldsymbol{k}_m\boldsymbol{u}_{\beta}(t)}\mathrm{e}^{+\mathrm{i}\boldsymbol{k}_m\boldsymbol{u}_{\beta}(\tilde{t})}\bigr\rangle \\ &= \exp\Bigl\{-\frac{1}{2}\bigl\langle[\boldsymbol{k}_m\boldsymbol{u}_{\beta}]^2\bigr\rangle - \frac{1}{2}\bigl\langle[\boldsymbol{k}_{\widetilde{m}}\boldsymbol{u}_{\beta}]^2\bigr\rangle + \bigl\langle\boldsymbol{k}_m\boldsymbol{u}_{\beta}(t)\ \boldsymbol{k}_{\widetilde{m}}\boldsymbol{u}_{\beta}\bigl(\tilde{t}\bigr)\bigr\rangle\Bigr\}, \end{aligned} \tag{4.3}$$

$$\begin{aligned} &\bigl\langle u^{i}_{\beta}(t)u^{j}_{\beta}\bigl(\tilde{t}\bigr)\bigr\rangle \\ &\quad = \frac{\hbar}{2nm_{\beta}}\sum_{\boldsymbol{q}\nu}\frac{e^{i}_{\boldsymbol{q}\nu}(\beta)e^{*j}_{\boldsymbol{q}\nu}(\beta)}{\omega_{\nu}(\boldsymbol{q})}\bigl[\bigl(\bar{n}_{\boldsymbol{q}\nu}+1\bigr)\mathrm{e}^{-\mathrm{i}\omega_{\nu}(\boldsymbol{q})(t-\tilde{t})} + \bar{n}_{\boldsymbol{q}\nu}\mathrm{e}^{\mathrm{i}\omega_{\nu}(\boldsymbol{q})(t-\tilde{t})}\bigr]. \end{aligned} \tag{4.4}$$

Here, m_{β} is the mass of the atom in position β of the crystal unit cell, n is the number of unit cells in the sample, $i, j = 1, 2, 3$ denote vector components in the Cartesian reference system.

The nuclear self-correlation function (4.3), (4.4) representing thermal lattice vibrations, generally speaking, is time-dependent. This time dependence should show up in the time spectra of coherent nuclear resonance scattering. In particular, as shown in [22], the time spectra of NFS are proportional to $|M_{mm}(t)|^2$. This property offers a new possibility of studying thermal motions, since $|M_{mm}(t)|^2$ contains the time-dependent displacement function $\langle u^i_\beta(t) u^j_\beta(0)\rangle$. NFS time spectra in some particular cases of thermal vibrations were calculated in [22]. It is remarkable that such studies are equivalently possible in solids, liquids and gases.

The interval is very short, $\simeq 10^{11}$–10^{13} s^{-1}, where the time dependence caused by thermal vibrations shows up in coherent resonant scattering. Although it is beyond the reach of present-day experimental techniques, in our opinion the problem could be solved by mapping the time-dependence of photon emission into an angular dependence of photon emission. Due to the recently demonstrated lighthouse effect [46] this mapping can be realized with the help of fast spinning the nuclear resonance samples irradiated with synchrotron radiation. Presently available spinning rates beyond 30 kHz would allow one to transform an extremely short time interval of 10^{-13} s into a measurable angular change in photon emission of 0.02 μrad.

The time-dependent term $\langle u^i_\beta(t) u^j_\beta(\tilde{t})\rangle$ vanishes if $(t-\tilde{t}) \gg \omega_{\mathrm{ph}}^{-1}$, where ω_{ph} is a characteristic phonon frequency, $\omega_{\mathrm{ph}} \simeq 10^{13}$–$10^{11}$ s^{-1}. For the rest of the time the motional part of the self-correlation function is time-independent and given by

$$M_{m\widetilde{m}(\beta)}(t-\tilde{t}) = \exp\left\{-\frac{1}{2}\left\langle[\boldsymbol{k}_m\boldsymbol{u}_\beta]^2\right\rangle - \frac{1}{2}\left\langle[\boldsymbol{k}_{\widetilde{m}}\boldsymbol{u}_\beta]^2\right\rangle\right\} = f_\beta^{1/2}(\boldsymbol{k}_m) f_\beta^{1/2}(\boldsymbol{k}_{\widetilde{m}}), \tag{4.5}$$

where $f_\beta(\boldsymbol{k}_m)$ is the Lamb–Mössbauer factor giving the probability of elastic resonance absorption or emission in the direction of the photon wave vector $\boldsymbol{k}_m$; $f_\beta(\boldsymbol{k}_m)$ can be calculated by using eq. (4.4) taken at $t=\tilde{t}$.

The interval where the motional part (4.3), (4.4) is time-dependent is usually short compared to the lifetime of the low-lying nuclear levels: $\tau_0 = \hbar/\Gamma_0 \geqslant 10^{-12}$ s. It is also short compared to typical times of hyperfine interactions. In the following sections the influence of the hyperfine interactions will be discussed. Therefore, the motional part of the self-correlation function will be assumed in the following to be time-independent and to be represented in accordance with (4.5) with the help of the Lamb–Mössbauer factors $f_\beta(\boldsymbol{k}_m)$ and $f_\beta(\boldsymbol{k}_{\widetilde{m}})$.

4.2. *Time-independent hyperfine interactions*

Let $|\beta_\lambda\rangle$ be eigenvectors and $\varepsilon_{\beta_\lambda}$ eigenenergies of the time-independent hyperfine interaction Hamiltonian $\widehat{\mathcal{H}}^{\mathrm{hf}}_{\beta_\lambda}$. If $|\beta_\lambda\rangle$ and $\varepsilon_{\beta_\lambda}$ are known, the matrix elements of the evolution operator can be readily evaluated from (2.15):

$$\mathcal{U}_{\beta_\lambda\tilde{\beta}_\lambda}(t_2,t_1) = \delta_{\beta_\lambda\tilde{\beta}_\lambda}\exp\left[-\frac{\mathrm{i}}{\hbar}\left(E_\lambda - \mathrm{i}\frac{\Gamma_0}{2}\delta_{\lambda,\mathrm{e}} + \varepsilon_{\beta_\lambda}\right)(t_2-t_1)\right]. \tag{4.6}$$

Hereafter the notation $\mathcal{U}$ is reserved for the evolution operator for time-independent hyperfine interactions. Inserting (4.6) into (3.7), (2.17) and by using (4.5) for the motional part we obtain the result that the self-correlation function (3.6) is purely elastic, i.e., $K^{s\tilde{s}}_{m\widetilde{m}}(t,\tilde{t}) = \mathcal{K}^{s\tilde{s}}_{m\widetilde{m}}(t-\tilde{t})$, and is given by

$$\mathcal{K}^{s\tilde{s}}_{m\widetilde{m}}(t) = \eta(t) \sum_{\ell\equiv\{\beta,\beta_{\mathrm{g}},\beta_{\mathrm{e}}\}} \mathcal{A}^{s\tilde{s}}_{m\widetilde{m}(\ell)} \mathrm{e}^{-\mathrm{i}\Omega_\ell t}, \tag{4.7}$$

$$\eta(t) = \exp\left[\frac{\mathrm{i}}{\hbar}(\hbar\widetilde{\omega} - E_0 + \mathrm{i}\Gamma_0/2)t\right]\theta(t), \tag{4.8}$$

$$\mathcal{A}^{s\tilde{s}}_{m\widetilde{m}(\ell)} = X_\beta f_\beta^{1/2}(\boldsymbol{k}_m) j^{s}_{\beta_{\mathrm{g}}\beta_{\mathrm{e}}}(\boldsymbol{k}_m) j^{\tilde{s}}_{\beta_{\mathrm{e}}\beta_{\mathrm{g}}}(-\boldsymbol{k}_{\widetilde{m}})\, f_\beta^{1/2}(\boldsymbol{k}_{\widetilde{m}}) \tag{4.9}$$

with $\hbar\Omega_\ell = \varepsilon_{\beta_{\mathrm{e}}} - \varepsilon_{\beta_{\mathrm{g}}}$ the corrections to the transition energies E_0 arising from the hyperfine interaction. Unless this causes ambiguities we use for brevity a joint index $\ell \equiv \{\langle\beta_{\mathrm{g}}| \Leftrightarrow |\beta_{\mathrm{e}}\rangle, \beta\}$ to denote both the transition between the ground and excited nuclear states and the group number β.[2]

According to (2.40) or (3.14) the nuclear response in single scattering approximation is proportional to the nuclear self-correlation function. In the present case it is a sum of monochromatic components with frequencies Ω_ℓ: (4.7)–(4.8). The amplitude $\mathcal{A}^{s0}_{m0(\ell)}$ of each emitted frequency component is proportional to the product of the emission ($j^{s}_{\beta_{\mathrm{g}}\beta_{\mathrm{e}}}(\boldsymbol{k}_m)$) and the absorption ($j^{0}_{\beta_{\mathrm{e}}\beta_{\mathrm{g}}}(-\boldsymbol{k}_{\widetilde{m}})$) matrix elements. The interference of the different monochromatic components results in a time spectrum with periodic modulation called quantum beat [1,9]. 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 NFS time spectra under conditions of time-independent hyperfine interactions by using the procedure described in this paper and of fits of experimental spectra are presented in [21,36,56–58].

4.3. Switching of the magnetic hyperfine fields

In this section we present an example of calculating the nuclear self-correlation functions in the particular case of time-dependent hyperfine interactions. Directions of

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[2] The Fourier image $\int_0^\infty \exp(\mathrm{i}\omega t)\mathcal{K}^{s\tilde{s}}_{00}(t)\,\mathrm{d}t$ of the correlation function (4.7),

$$\mathcal{K}^{s\tilde{s}}_{00}(\omega) = \mathcal{K}^{s\tilde{s}}(\omega) = \sum_\ell \frac{\mathrm{i}\mathcal{A}^{s\tilde{s}}_{00(\ell)}\Gamma_0}{\hbar(\widetilde{\omega}+\omega) - E_0 - \Omega_\ell\hbar + \mathrm{i}\Gamma_0/2},$$

together with (2.52) and the relations $\omega = \varepsilon\Gamma_0/\hbar$, $\widetilde{\omega} = \tilde{\varepsilon}\Gamma_0/\hbar$, give the well known frequency transmission function through the nuclear resonance medium with time-independent hyperfine interactions.

the magnetic hyperfine fields are switched (rotated) from $\boldsymbol{n}_0^{(\beta)}$ to $\boldsymbol{n}_0^{(\beta')}$ instantaneously at a definite time t':

$$\boldsymbol{B}^{(\beta)}(t) = B_0^{(\beta)}\big[\boldsymbol{n}_0^{(\beta)}\theta\big(t'-t\big) + \boldsymbol{n}_0^{(\beta')}\theta\big(t-t'\big)\big]. \tag{4.10}$$

The rotation is specified by the three Eulerian angles $\{\chi_1', \chi_2', \chi_3'\} = \boldsymbol{\chi}'$. The Hamiltonian for a magnetic hyperfine field changing in time in general is

$$\widehat{\mathcal{H}}^{\mathrm{hf}}_{\beta_\lambda}(t) = -\mu_\lambda \frac{\widehat{\boldsymbol{J}}_\lambda \boldsymbol{B}^{(\beta)}(t)}{J_\lambda}. \tag{4.11}$$

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 (4.7)–(4.9) derived in section 4.2.

After switching ($t > t'$) the hyperfine field (4.10) and the Hamiltonian (4.11) 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

$$|\beta_\lambda\rangle = \sum_{\beta'_\lambda} |\beta'_\lambda\rangle \mathcal{D}_{\beta'_\lambda\beta_\lambda}\big(\boldsymbol{\chi}'\big), \qquad \langle\beta_\lambda| = \sum_{\beta'_\lambda} \mathcal{D}^{-1}_{\beta_\lambda\beta'_\lambda}\big(\boldsymbol{\chi}'\big)\langle\beta'_\lambda|. \tag{4.12}$$

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 $\mathcal{D}^{(I_\lambda)}_{m'_\lambda m_\lambda}(\boldsymbol{\chi}')$ [27,55].

The unknown values, which have yet 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 $\widehat{U}(t_2, t_1) = \widehat{\mathcal{U}}(t_2, t')\widehat{\mathcal{U}}(t', t_1)$, of the fact that the Hamiltonian (4.11) is time-independent although different in the time intervals (t_2, t') and (t', t_1) (see (4.10)), of definition (2.15), and of relation (4.12). The matrix elements of the evolution operator then become

$$U_{\beta'_\lambda\beta_\lambda}(t_2, t_1) = \mathcal{U}_{\beta'_\lambda\beta'_\lambda}\big(t_2, t'\big)\mathcal{D}_{\beta'_\lambda\beta_\lambda}\mathcal{U}_{\beta_\lambda\beta_\lambda}\big(t', t_1\big), \tag{4.13}$$

$$U^{-1}_{\beta_\lambda\beta'_\lambda}(t_2, t_1) = \mathcal{U}^{-1}_{\beta_\lambda\beta_\lambda}\big(t', t_1\big)\mathcal{D}^{-1}_{\beta_\lambda\beta'_\lambda}\mathcal{U}^{-1}_{\beta'_\lambda\beta'_\lambda}\big(t_2, t'\big). \tag{4.14}$$

It was taken into account that only the diagonal matrix elements $\mathcal{U}_{\beta_\lambda\beta_\lambda}$ of the unperturbed evolution operator (4.6) have nonzero values. Combining (4.13), (4.14) with (3.7), (2.17) we obtain the following expressions for the correlation function after switching ($t > t' > \tilde{t}$):

$$K^{s\tilde{s}}_{m\widetilde{m}}\big(t, \tilde{t}\big) = \eta\big(t-\tilde{t}\big) \sum_{\ell' \equiv \{\beta, \beta'_{\mathrm{g}}, \beta'_{\mathrm{e}}\}} \mathcal{A}^{s\tilde{s}}_{m\widetilde{m}(\ell')}\big(\boldsymbol{\chi}', t'-\tilde{t}\big)\mathrm{e}^{-\mathrm{i}\Omega_{\ell'}(t-t')}, \tag{4.15}$$

$$\begin{aligned}\mathcal{A}^{s\tilde{s}}_{m\widetilde{m}(\ell')}\big(\boldsymbol{\chi}', t'-\tilde{t}\big) = {} & X_\beta f_\beta^{1/2}(\boldsymbol{k}_m) f_\beta^{1/2}(\boldsymbol{k}_{\widetilde{m}}) j^s_{\beta'_{\mathrm{g}}\beta'_{\mathrm{e}}}(\boldsymbol{k}_m) \\ & \times \sum_{\beta_{\mathrm{g}}, \beta_{\mathrm{e}}} S'_{\ell'\ell}\big(\boldsymbol{\chi}', t'-\tilde{t}\big) j^{\tilde{s}}_{\beta_{\mathrm{e}}\beta_{\mathrm{g}}}(-\boldsymbol{k}_{\widetilde{m}}),\end{aligned} \tag{4.16}$$

$$S'_{\ell'\ell}(\chi', t' - \tilde{t}) = \mathcal{D}_{\beta'_{\mathrm{g}}\beta_{\mathrm{g}}}(\chi')\mathcal{D}_{\beta'_{\mathrm{e}}\beta_{\mathrm{e}}}(\chi')\,\mathrm{e}^{-\mathrm{i}\Omega_{\ell}(t'-\tilde{t})}. \tag{4.17}$$

The correlation function (4.15) has a structure similar to that of the unperturbed correlation function (4.7).[3] However, this similarity is formal. Firstly, the time variables t and $\tilde{t}$ enter (4.15)–(4.17) independently. Thus, unlike the unperturbed correlation function, given by eq. (4.7), the correlation function of eq. (4.15) describes *inelastic* coherent scattering. Secondly, the amplitudes $\mathcal{A}^{s\tilde{s}}_{m\tilde{m}(\ell')}(\chi', t' - \tilde{t})$ of the monochromatic components $\Omega_{\ell'}$ are different. They are built by the interference of all initially excited transitions $\beta_{\mathrm{g}} \Rightarrow \beta_{\mathrm{e}}$. The interference shows up in eq. (4.16) as the sum of the current density matrix elements $j^{\tilde{s}}_{\beta_{\mathrm{e}}\beta_{\mathrm{g}}}(-\tilde{\boldsymbol{k}})$ with amplitudes given by $S'_{\ell'\ell}(\chi', t' - \tilde{t})$ in eq. (4.17). By varying the angle χ' of switching and the switching time t' one can change the amplitudes $S'_{\ell'\ell}$ and can thus control the interference pattern and the coherently re-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 [16,17,51,53,54,59] or in the time reversal of the time spectrum [52]. E.g., one can make the interference totally destructive and thus suppress the coherent re-emission [17,59]. However, this by no means implies that the nuclear excitation is destroyed. By the next switching at the proper time and with the proper angle one can restore constructive interference and again see coherently re-emitted radiation [17,59].

This second switching is described similarly to the procedure outlined above. E.g., 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. (4.12) with the substitutions $\beta_{\lambda} \to \beta'_{\lambda}$ and $\beta'_{\lambda} \to \beta''_{\lambda}$. Further, it can be shown that the nuclear self-correlation function is described by the same eqs. (4.15), (4.16), however, with the replacement

$$\ell' \to \ell'' \quad \text{and} \quad S'_{\ell'\ell}(\chi', t' - \tilde{t}) \to S''_{\ell''\ell}(\chi'', t'' - \tilde{t}),$$

where $S''_{\ell''\ell}(\chi'', t'' - \tilde{t})$ is now defined as

$$S''_{\ell''\ell}(\chi'', t'' - \tilde{t}) = \sum_{\beta'_{\mathrm{e}},\beta'_{\mathrm{g}}} S'_{\ell''\ell'}(\chi'', t'' - t')\,S'_{\ell'\ell}(\chi', t' - \tilde{t}). \tag{4.18}$$

Examples of transformations occurring after the second switching were presented in [17,53]. Any subsequent switching is described similarly.

Some additional details of the theory of nuclear resonance scattering in the case of switching the magnetic hyperfine fields can be found in [16,17,51,52,45]. Examples of evaluations of NFS time spectra under conditions of switching the magnetic hyperfine fields by using the procedure described in the present paper and fits of experimental spectra are presented in [17].

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[3] In the particular case of pure magnetic interactions which we consider here, the frequencies $\Omega_{\ell'}$ 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.

5. Conclusions

A general approach for solving nuclear resonant coherent scattering problems directly in time and in space is presented. It is based on the solution of the first order integro-differential equations with a kernel which is a double-time nuclear self-correlation function $K_{m\widetilde{m}}(t,\tilde{t})$. The kernel represents the coherent single scattering response of the nuclear system at time t in the direction $\boldsymbol{k}_m$ to the excitation at $\tilde{t}$ with radiation of wave vector $\boldsymbol{k}_{\widetilde{m}}$. The explicit form of the kernel is defined by the type of interactions the nuclei experience with their environment and by the character of their motion in space. The kernels for some particular cases of hyperfine interactions and motion in space are presented. A general procedure is given for the solution of the wave equation with a single coherent beam. The multiple beam coherent scattering problem can be solved analytically in single scattering approximation. The multiple scattering, multiple beam problems can be solved numerically.

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