J. Phys.: Condens. Matter 7 (1995) 7589–7600. Printed in the UK

Coherent scattering of a synchrotron radiation pulse by nuclei in vibrating crystals

V G Kohn† and Yu V Shvyd'ko‡

† Russian Research Centre 'Kurchatov Institute', Moscow 123182, Russia
‡ Institut für Experimentalphysik, Universität Hamburg, D-22761 Hamburg, Germany

Received 20 February 1995, in final form 23 June 1995

Abstract. A theory describing the time evolution of coherent scattering of a synchrotron radiation pulse by a nuclear ensemble in a crystal subjected to acoustic vibrations is presented for the case where the nuclei are vibrated in unison. A general multiple-diffraction scattering is considered which involves scattering in the forward direction as well as the two-beam Bragg or Laue diffraction as particular cases. The vibrations cause only the phase shift of the resonantly scattered radiation field which depends on the positions of nuclei at the moments of excitation and de-excitation. The time evolution as a result turns out to be insensitive to vibrations. It is shown that the time evolution of scattering from two vibrating crystals contains information on their relative motion.

1. Introduction

Mössbauer energy spectra are known to be very sensitive to nuclear motions. Harmonic vibrations with a frequency f result in satellite lines which are spaced out with the period $2\pi f\hbar$ [1] (see e.g. the review [2] and references therein). The small natural width of the nuclear γ-resonance Γ_0 and the short wavelength of the Mössbauer radiation λ_0 define the scale of frequency and amplitude sensitivity of the Mössbauer spectra to vibrations. For example, in the case of 14.413 keV γ-resonance of ^{57}Fe nuclei the corresponding values are $\Gamma_0 = 7.068 \times 10^6\ \mathrm{s}^{-1}$ and $\lambda_0 = 0.861$ Å. The different aspects of the effect of vibrations on Mössbauer energy absorption and diffraction spectra were considered theoretically in [3–10].

The influence of vibrations on the Mössbauer time spectra was investigated experimentally recently in [11]. The effect of vibrations in the important particular case, where the nuclei were vibrating in unison, on the time evolution of the nuclear Bragg diffraction in a perfect ^{57}FeBO$_3$ crystal was studied. The time spectra were measured by exciting the nuclei in the crystal by synchrotron radiation pulses (for reviews on the Mössbauer spectroscopy in the time domain see e.g. [12–14]). The influence of vibrations in the same sample on the diffraction energy spectra was investigated earlier in [10]. It was shown that unlike the energy spectra the time spectra are not affected by the vibrations excited in the ^{57}FeBO$_3$ crystal. This result was explained in [11] by the fact that after the excitation by a synchrotron radiation pulse the nuclei vibrating in unison cause only the phase modulation of the scattered radiation. Installation of the resonant nuclear absorber—the second ^{57}FeBO$_3$ crystal—downstream from the vibrating crystal revealed an effect of vibrations in the time spectra of scattering from both crystals.

0953-8984/95/387589+12$19.50

We present a theory which was developed to describe the results of the experiment [11] and was used there to fit the data. The theory is more general than the case investigated experimentally. The influence of nuclear vibrations on the coherent nuclear scattering is considered in a general multiple-diffraction case which includes the forward scattering as well as the two-beam Bragg or Laue diffraction as particular cases. The theory is restricted to the unison mode of nuclear vibrations which corresponds for instance to ultrasound vibrations with the wavelength exceeding dimensions of the effective coherent scattering volume in a crystal. The time dependence of nuclear coherent scattering from ultrasound with short wavelength or from phonons is quite different and will be treated elsewhere.

In section 2 we derive general formulae useful for calculating the Mössbauer time spectra of *inelastic coherent* scattering. The dynamic equations for the multiple diffraction in the vibrating crystal are derived in section 3. The solutions of these equations in general form are obtained in section 4. The theory confirms the experimentally observed insensitivity of Mössbauer time spectra to the unison vibrations of nuclei. This result is generalized to any type of coherent scattering and to any form of unison vibrations.

In section 5 of the paper general equations for calculating the time spectra of the successive scattering from two or more vibrating crystals are presented. Two particular examples are discussed. The first one is the scattering from the vibrating Bragg reflector and the absorber moving with constant velocity. The second example is the nuclear forward scattering from two resonance absorbers. It is shown that the time spectra contain information on the relative motion of the samples even if in that case the vibrations are not synchronized with the synchrotron radiation pulse.

2. General formulae

To describe the time evolution of the coherent scattering of the synchrotron radiation pulse from a nuclear ensemble in a crystal we shall follow the general procedure proposed in [15] which has exploited the formalism of response functions. The method of [15] was developed to describe *elastic coherent* scattering, namely, a spatially coherent scattering without the change of the radiation frequency. We shall generalize the formalism to the cases of *inelastic coherent* scattering, a spatially coherent process with the change of the frequency. This type of scattering may occur due to nuclear vibrations, time-dependent hyperfine interactions, etc.

We shall specify the type of inelastic scattering in the next section. Here we shall consider the inelastic scattering in a general way by introducing a time-dependent scattering amplitude $R^{ss'}(t,\omega)$ which is the response of a scatterer on the monochromatic wave of frequency ω and polarization s'. Inelastic scattering due to nuclear vibrations [3, 6, 9, 16] or time-dependent hyperfine interactions [17, 18] results in the appearance of new frequency components in the spectrum of the scattered radiation. Therefore the inelastic scattering amplitude unlike the elastic one (cf. [15]) is time dependent. We shall assume also that the scatterer can change the state of polarization of the radiation from s' to s.

It is convenient to proceed from the coherent incident radiation field of general form $E_{\mathrm{in}}(t)$. The actual properties of the synchrotron radiation field will be taken into account later.

The electric field of scattered radiation then equals

$$E^{s}(t) = \int_{-\infty}^{\infty} \frac{\mathrm{d}\omega}{2\pi} R^{ss'}(t,\omega) E_{\mathrm{in}}^{s'}(\omega) \exp(-\mathrm{i}\omega t) \tag{1}$$

where $E_{\mathrm{in}}^{s}(\omega)$ is the Fourier transform of $E_{\mathrm{in}}^{s}(t)$. The formula (1) takes into account in an explicit form the phase relation between the incident and the scattered waves. It provides the general procedure of calculating the time dependence of the coherently scattered field via the Fourier transformation of the incident field.

The incident and scattered fields can be related directly with the help of the response function $G^{ss'}(t,t')$ which is defined as

$$G^{ss'}(t,t') = \int_{-\infty}^{\infty} \frac{\mathrm{d}\omega}{2\pi} R^{ss'}(t,\omega)\exp[-\mathrm{i}\omega(t-t')]. \tag{2}$$

The most general property of the scattering amplitude $R^{ss'}(t,\omega)$ is its analyticity in the upper half of the plane of complex ω values. As a result, the function $G^{ss'}(t,t')$ is the delaying function which equals zero at $t' > t$.

The expression for the time dependence of the scattered radiation field can be rewritten now as a matrix product (or a generalized convolution) of the incident radiation with the response function:

$$E^{s}(t) = \int_{-\infty}^{\infty} \mathrm{d}t' G^{ss'}(t,t') E_{\mathrm{in}}^{s'}(t'). \tag{3}$$

From this relation it is clear that t' represents the time instant of excitation while t is the time instant of de-excitation.

Let us now specify the incident radiation field. In our case it is a synchrotron radiation pulse. The synchrotron radiation pulses have a duration T, typically $\sim 10^{-10}$ s, which is defined mainly by the linear size of the electron (positron) bunches in the storage ring. The synchrotron radiation pulse itself is a sum of many elementary radiation pulses, each arising from an individual electron (positron) in the bunch. We associate the previously defined incident radiation field $E_{\mathrm{in}}^{s}(t)$ with one of these elementary radiation pulses. The individual duration of each elementary pulse, typically $\sim 10^{-11}$ s, is shorter than T and much shorter than the characteristic times of the nuclear scattering system, which are usually $> 10^{-8}$ s. Therefore $E_{\mathrm{in}}(t')$ can be represented with a good accuracy by the delta function: $E_{\mathrm{in}}^{s}(t') \propto \delta(t'-t_0)$. Elementary pulses arising from the different electrons are incoherent. Thus the intensity of the scattered radiation $I(s',t) \propto |E^{s}(t)|^2$ should be averaged over the t_0—the arrival time of each elementary pulse. As a result we obtain the general formula for the time dependence of inelastic coherent scattering of the synchrotron radiation pulse of duration T in the s' polarization state from a nuclear ensemble

$$I(s',t) = \frac{N_0}{T}\int_0^T \mathrm{d}t_0 \sum_s |G^{ss'}(t,t_0)|^2 \tag{4}$$

with N_0 being the number of photons in the unit frequency band per synchrotron radiation pulse. The intensity given by (4) is summed over the final polarization states s of the scattered radiation. In many practically important cases $t \gg T$ and we have simply

$$I(s',t) = N_0 \sum_s |G^{ss'}(t,0)|^2. \tag{5}$$

Thus the problem of calculating the time evolution of scattering from the nuclear ensemble is reduced to seeking the scattering amplitude $R^{ss'}(t,\omega)$ or the response function $G^{ss'}(t,t')$ of the scatterer.

3. Wave equation

Let us consider a vibrating crystal in the form of a plate of thickness L in the z-direction. Let the wavelength of the vibrations be so long that all the scattering parts of the crystal are vibrating in unison, i.e. the displacement $\boldsymbol{u}(t)$ does not depend on the space coordinate. To calculate the coherent electric field $\boldsymbol{E}(\boldsymbol{r},t)$ of the radiation scattered by nuclei in the vibrating crystal we shall use Maxwell's wave equation

$$\nabla^2\boldsymbol{E} - \operatorname{grad}\operatorname{div}\boldsymbol{E} - \frac{1}{c^2}\frac{\mathrm{d}^2}{\mathrm{d}t^2}\boldsymbol{E} = \frac{4\pi}{c^2}\frac{\mathrm{d}}{\mathrm{d}t}\boldsymbol{j} \tag{6}$$

where $\boldsymbol{j}(\boldsymbol{r},t)$ is the induced current density which is a quantum mechanical average of the current density operator. To find the scattering amplitude we consider a monochromatic incident plane wave with frequency ω and wave vector $\boldsymbol{k}_0$:

$$\boldsymbol{E}(\boldsymbol{r},t) = \mathcal{E}_0\exp(\mathrm{i}\boldsymbol{k}_0\cdot\boldsymbol{r} - \mathrm{i}\omega t). \tag{7}$$

Taking into account the periodicity of the crystal lattice we represent the field inside the crystal as the Bloch wave

$$\boldsymbol{E}(\boldsymbol{r},t) = \sum_m \boldsymbol{E}_m(z,t)\exp(\mathrm{i}\boldsymbol{k}_m\cdot\boldsymbol{r} - \mathrm{i}\omega t) \tag{8}$$

which is a sum of plane wave components with wave vectors $\boldsymbol{k}_m = \boldsymbol{k}_0 + \boldsymbol{h}_m$. This sum involves all the reciprocal lattice vectors $\boldsymbol{h}_m$ of the crystal. If the nuclei of the crystal are vibrating then the amplitudes $\boldsymbol{E}_m(z,t)$ depend on time. The amplitudes depend as well on the coordinate z due to refraction. Because of the same factors and within the framework of the linear response theory the induced current density is also the Bloch wave

$$\boldsymbol{j}(\boldsymbol{r},t) = \sum_m \boldsymbol{j}_m(z,t)\exp(\mathrm{i}\boldsymbol{k}_m\cdot\boldsymbol{r} - \mathrm{i}\omega t). \tag{9}$$

Since the refraction effect is small due to nuclear resonant and electronic scattering, the z-dependence of the quantities $\boldsymbol{E}_m(z,t)$ and $\boldsymbol{j}_m(z,t)$ varies slowly as compared to the fast oscillations of the exponential functions (this is the so-called slowly varying envelope approximation). We shall also consider the cases where the frequency of vibrations Ω is much less than the frequency of the radiation ω. These approximations allow us to reduce the second-order Maxwell equation to a first-order differential equation for the quantities $\boldsymbol{E}_m(z,t)$ and $\boldsymbol{j}_m(z,t)$ (see also [9, 17, 19])

$$\left(\gamma_m\frac{\mathrm{d}}{\mathrm{d}z} + \mathrm{i}\frac{K}{2}\alpha_m\right)\boldsymbol{E}_m(z,t) = -\frac{2\pi}{c}\boldsymbol{j}_m(z,t) \tag{10}$$

where

$$\alpha_m = (k_m^2 - K^2)/K^2 \qquad \gamma_m = k_{mz}/K \qquad K \equiv \omega/c \equiv 2\pi/\lambda. \tag{11}$$

In the linear approximation one can derive the following general expression for the components of the induced current density:

$$j_m^s(z,t) = -\frac{\mathrm{i}\omega}{4\pi}\sum_{m's'}[\hat{P}^{ss'}_{mm'}(t,\tau)E^{s'}_{m'}(z,\tau) + \chi^{ss'}_{mm'}F_{mm'}(t)E^{s'}_{m'}(z,t)] \tag{12}$$

where the first term describes the nuclear current by means of the causal integral operator while the second term corresponds to instantaneous electronic scattering. The integral operator has the following form provided the nuclei are vibrating in unison:

$$\hat{P}^{ss'}_{mm'}E^{s'}_{m'} = -\frac{\mathrm{i}}{2}\Gamma_0 f_m(t)\sum_{\mathrm{ge}} p^{ss'}_{mm'}(\mathrm{ge})\int_{-\infty}^{t}\mathrm{d}\tau\, e(\omega-\omega_{\mathrm{ge}}, t-\tau)f^*_{m'}(\tau)E^{s'}_{m'}(z,\tau) \tag{13}$$

where

$$e(\omega,t) = \exp\{\mathrm{i}(\omega + \mathrm{i}\Gamma/2)t\} \qquad f_m(t) = \exp\{-\mathrm{i}\boldsymbol{k}_m\cdot\boldsymbol{u}(t)\} \tag{14}$$

and

$$p_{mm'}^{ss'}(\mathrm{ge}) = -\frac{8\pi}{\omega^2(2I_0+1)V_0\Gamma_0}\sum_l \langle \mathrm{g}|\hat{j}_l^{s}(\boldsymbol{k}_m)|\mathrm{e}\rangle\langle \mathrm{e}|\hat{j}_l^{s'}(-\boldsymbol{k}_{m'})|\mathrm{g}\rangle$$
$$\times \exp\{\mathrm{i}(\boldsymbol{k}_{m'}-\boldsymbol{k}_m)\cdot\boldsymbol{\rho}_l - \tfrac{1}{2}[Z_l(\boldsymbol{k}_{m'})+Z_l(\boldsymbol{k}_m)]\}. \tag{15}$$

The derivation of this expression and notations are given in the appendix.

The second term in (12) corresponds to the potential electronic scattering. The latter cannot influence the time spectrum of the scattered radiation directly but in combination with nuclear scattering it can be of importance. In (12) $\chi_{mm'}^{ss'}$ is the Fourier transform of the complex electronic polarizability of the crystal at rest; $F_{mm'}(t) = f_m(t)f_{m'}^*(t)$ is a displacement phase factor.

The number of equations in (10) is equal to the number of amplitudes $\boldsymbol{E}_m$ which should be considered. The latter is determined by the number of parameters α_m having values close to zero. In other words it is equal to the number of reciprocal lattice points which lie near the Ewald sphere ($|\boldsymbol{k}_m| \simeq K$) and obey the Bragg law.

We shall consider the general case of multiple diffraction in a perfect crystal. The boundary conditions for the set of equations (10)–(12) then have the following form:

$$\begin{aligned}
&\boldsymbol{E}_0(0,t) = \boldsymbol{\mathcal{E}}_0 && \text{(forward beam)}\\
&\boldsymbol{E}_m(0,t) = 0 \quad \text{if } \gamma_m > 0 && \text{(Laue reflected beams)}\\
&\boldsymbol{E}_m(L,t) = 0 \quad \text{if } \gamma_m < 0 && \text{(Bragg reflected beam).}
\end{aligned} \tag{16}$$

Now the problem is formulated completely.

4. Scattering amplitude and response function

The problem, formulated in the previous section, can be solved by performing the following two operations. First we eliminate the time-dependent factors $f_m(t)$ from the equations (10)–(13) by the substitution

$$\boldsymbol{E}_m(z,t) = f_m(t)\boldsymbol{B}_m(z,t) \tag{17}$$

where $\boldsymbol{B}_m(z,t)$ are new unknown functions. The boundary conditions for $\boldsymbol{B}_m(z,t)$ are time dependent:

$$\begin{aligned}
&\boldsymbol{B}_0(0,t) = \boldsymbol{\mathcal{E}}_0 \exp\{\mathrm{i}\boldsymbol{k}_0\cdot\boldsymbol{u}(t)\}\\
&\boldsymbol{B}_m(0,t) = 0 \quad \text{if } \gamma_m > 0\\
&\boldsymbol{B}_m(L,t) = 0 \quad \text{if } \gamma_m < 0.
\end{aligned} \tag{18}$$

Henceforth we shall confine ourselves to the case of harmonic vibrations with amplitude $\boldsymbol{d}$, frequency Ω and initial phase φ

$$\boldsymbol{u}(t) = \boldsymbol{d}\sin(\Omega t + \varphi). \tag{19}$$

In this particular case the boundary condition for $\boldsymbol{B}_0(0,t)$ takes the following form after substitution of equation (19) in (18):

$$\boldsymbol{B}_0(0,t) = \boldsymbol{\mathcal{E}}_0 \exp\{\mathrm{i}b_0\sin(\Omega t+\varphi)\} = \boldsymbol{\mathcal{E}}_0 \sum_{n=-\infty}^{\infty} J_n(b_0)\exp\{\mathrm{i}n(\Omega t+\varphi)\} \tag{20}$$

where $J_n(b_0)$ is the Bessel function of integer index n; $b_m = \boldsymbol{k}_m\cdot\boldsymbol{d}$ is a dimensionless amplitude of vibrations in the direction of the wave vector $\boldsymbol{k}_m$ which is called the modulation index. In equation (20) a well known expansion of the $\exp\{ib_0\sin(\Omega t+\varphi)\}$ function was used [20].

The evidence of the second operation follows from the next observation. The integral over time in (13) could be calculated explicitly if $B_m(z,t)$ were the monochromatic wave. However the boundary conditions (18,20) do not allow this kind of solution. Instead we can find the solution for $B_m(z,t)$ as a sum of monochromatic waves

$$B_m(z,t)=\sum_{n=-\infty}^{\infty}B_m^{(n)}(z)\exp[\mathrm{i}n(\Omega t+\varphi)]. \tag{21}$$

Since the equations are linear with respect to the functions B_m their form is conserved for each spectral component $B_m^{(n)}$ as well. But now the time integral over time in (13) can be calculated explicitly because the functions $B_m^{(n)}$ do not depend on time. As a result we obtain the following set of equation for the amplitudes of monochromatic components $B_m^{(n)}$:

$$\left(\gamma_m\frac{\mathrm{d}}{\mathrm{d}z}+\mathrm{i}\frac{K}{2}\alpha_m\right)B_m^{s(n)}(z)=\mathrm{i}\frac{K}{2}\sum_{m's'}g_{mm'}^{ss'}(\omega-\Omega n)B_{m'}^{s'(n)}(z) \tag{22}$$

where

$$g_{mm'}^{ss'}(\omega)=\sum_{\mathrm{ge}}\frac{p_{mm'}^{ss'}(\mathrm{ge})\Gamma_0/2}{(\omega-\omega_{\mathrm{ge}}+\mathrm{i}\Gamma/2)}+\chi_{mm'}^{ss'}. \tag{23}$$

The boundary conditions for $B_m^{(n)}(z)$ take the form

$$\begin{aligned}&B_0^{(n)}(0)=\mathcal{E}_0J_n(b_0)\\&B_m^{(n)}(0)=0\quad\text{if }\gamma_m>0\\&B_m^{(n)}(L)=0\quad\text{if }\gamma_m<0.\end{aligned} \tag{24}$$

The equations (22) with the boundary conditions (24) represent the standard problem of γ-ray diffraction in a crystal at rest. Such a problem was solved earlier in many particular cases. For example in the case of forward scattering (one-beam diffraction) the solution involving no polarization state mixing is obtained from (19) directly

$$B_0^{s(n)}(z)=\exp\left[\mathrm{i}\frac{Kz}{2\gamma_0}g_{00}^{ss}(\omega-\Omega n)\right]B_0^{s(n)}(0). \tag{25}$$

In the Bragg and Laue cases of two-beam γ-ray diffraction the solutions were given in [21–24]. The case of multiple diffraction was considered recently in [25]. Therefore we shall assume that the functions $B_m^{(n)}(z)$ are known. The nth component of the scattering amplitude is then equal to

$$\mathcal{R}_{m0}^{ss'}(\omega-\Omega n)=\frac{B_m^{s(n)}(z_{out})}{B_0^{s'(n)}(0)} \tag{26}$$

where $z_{out}=L$ for the forward-scattered and for the Laue-reflected beams ($\gamma_m>0$), and $z_{out}=0$ for the Bragg-reflected beams ($\gamma_m<0$). As a result the total scattering amplitude $R_{m0}^{ss'}(t,\omega)$ of the vibrating crystal can be expressed through the scattering amplitude of the crystal at rest†

$$R_{m0}^{ss'}(t,\omega)=f_m(t)\sum_n\mathcal{R}_{m0}^{ss'}(\omega-\Omega n)J_n(b_0)\exp\{\mathrm{i}n(\Omega t+\varphi)\}. \tag{27}$$

---

† We denote hereafter the quantities for the crystal at rest by calligraphic symbols.

The substitution of this expression in (2) results in the general formula for the response function of the vibrating crystal

$$G^{ss'}_{m0}(t,t') = f_m(t)\mathcal{G}^{ss'}_{m0}(t-t')f_0^*(t') \tag{28}$$

where

$$\mathcal{G}^{ss'}_{m0}(t) = \int_{-\infty}^{\infty}\frac{\mathrm{d}\omega}{2\pi}\mathcal{R}^{ss'}_{m0}(\omega)\exp(-\mathrm{i}\omega t). \tag{29}$$

Equation (28) shows that acoustic vibrations lead only to the phase modulation of the response function for the crystal at rest through the factor

$$f_m(t)f_0^*(t') = \exp\{-\mathrm{i}\boldsymbol{k}_m\cdot\boldsymbol{u}(t) + \mathrm{i}\boldsymbol{k}_0\cdot\boldsymbol{u}(t')\} \tag{30}$$

which arises from the difference in positions of the nucleus at the moments of excitation and de-excitation. However, this phase factor disappears in the time spectrum—(4), (28). It is for this reason the time spectra for the vibrating crystal measured in [11] turned out to be the same as for the crystal at rest.

The result obtained can be explained in another way too. Let us transfer from the laboratory reference system, where the crystal is vibrating, to the one attached to the crystal. This transformation leads to the situation where the crystal is at rest and all other objects (the source of radiation and the detector) are vibrating. In the new reference system we obtain (28) directly. The crystal at rest is described by the response function $\mathcal{G}^{ss'}_{m0}(t-t')$ while the function (30) takes into account the phase shifts due to the displacement of the source and the detector. It is also worth noting that this result is valid not only for the harmonic vibrations given by equation (19). It can be generalized to any form of unison motion $\boldsymbol{u}(t)$.

5. Scattering by two crystals

5.1. General consideration

As was shown in [11] to reveal the vibrations in the time spectrum an additional nuclear resonance scatterer should be introduced. For this reason we shall generalize the problem and consider cases of successive scattering of the synchrotron radiation pulse from two crystals where both of the crystals (or one of them) are vibrating.

The radiation scattered by the first crystal is the incident radiation for the second crystal. Taking this into account and using (3) we obtain immediately that the response function for the system of two vibrating crystals is a convolution of the response functions of these crystals

$$\hat{G}_{21}(t,t') = \hat{G}_2(t,t'')\otimes\hat{G}_1(t'',t') \tag{31}$$

where the hat symbol denotes a matrix and the symbol ⊗ stands for

$$\begin{aligned}[]
[\hat{B}(t,t'')\otimes\hat{A}(t'',t')]^{ss'}_{mm'} &= \sum_{s''}\int_{t'}^{t}\mathrm{d}t''\,B^{ss''}_{mm''}(t,t'')A^{s''s'}_{m''m'}(t'',t') \\
&= f_m^{(B)}(t)\left\{\sum_{s''}\int_{t'}^{t}\mathrm{d}t''\,\mathcal{B}^{ss''}_{mm''}(t-t'')f_{m''}^{(B)*}(t'')f_{m''}^{(A)}(t'')\mathcal{A}^{s''s'}_{m''m'}(t''-t')\right\}f_{m'}^{(A)*}(t').
\end{aligned} \tag{32}$$

Index m'' denotes here the direction of the beam between the two crystals.

From these formulae one can observe that the time spectrum $I_{21}(t) \propto |G_{21}(t,0)|^2$ will be affected by the relative motion of the scatterer through the factor

$$F^{BA}_{m''}(t'') = f^{(B)*}_{m''}(t'') f^{(A)}_{m''}(t'') = \exp\{\mathrm{i}\boldsymbol{k}_{m''} \cdot [\boldsymbol{u}^B(t'') - \boldsymbol{u}^A(t'')]\} \qquad (33)$$

in equation (32). It is also worth noting that the procedure presented by the (31)–(32) allows us to consider successive scattering from any number of scatterers.

In the derivation presented above we have assumed that the distances from the source to the first crystal L_s, between the crystals L_c and from the second crystal to the detector L_d are small enough that the condition $\Delta\omega L_x/c \ll 1$ is fulfilled. Here $x = s, d, c$ and $\Delta\omega$ is the characteristic frequency interval of resonant scattering which is typically about $100\Gamma_0/\hbar$. If this condition is not fulfilled, for example the distance between the crystals L_c is large, then equation (31) should be revised by taking into account retardation effects. This will be accounted for by a response function of the empty space.

It is clear that the scattering amplitude of the empty space with the length L_x is $R_x(\omega) = \exp(\mathrm{i}KL_x) = \exp(\mathrm{i}\omega t_x)$ where $t_x = L_x/c$ with $x = s, d, c$. The quantities t_s, t_d and t_c represent the time of flight of the light through the empty space. The substitution of this function in (2) results in the response function $G_x(t,t') = \delta(t - t_x - t')$.

The resultant response function of the successive scattering from two crystals which takes the propagation in the empty spaces into account should be a convolution of five response functions instead of two in equation (31). Two of them are response functions of the crystals themselves while the other three are the response functions of the empty spaces around the crystals, which are simply delta functions. Performing explicitly the convolution with the delta functions we finally obtain the following formula instead of (31):

$$\hat{G}_{21}(t,t') = \hat{G}_2(t - t_d, t'' + t_c) \otimes \hat{G}_1(t'', t' + t_s). \qquad (34)$$

This formula describes the successive scattering from two crystals with allowance for the retardation effect due to the finite light velocity. The manifestation of this effect in the energy Mössbauer spectra was discussed in [7, 26]. This effect may play an important role provided t_c is comparable with the nuclear lifetime $\tau_0 = \hbar/\Gamma_0$. In the case of the 14.4 keV nuclear resonance in ^{57}Fe this corresponds to the distance L_c between the scatterers of a few tens of metres. These are special conditions. For the sake of simplicity we shall neglect this effect in the consideration of the following particular cases of double-crystal scattering.

5.2. Specific case considered in [11]

Let us consider the case of scattering by two crystals (figure 1(a)) where the first one is the vibrating Bragg reflector ($\hat{\mathcal{R}}_1$) and the second one is the resonance absorber ($\hat{\mathcal{R}}_2$) moving with constant velocity $\boldsymbol{v}$. This case was studied experimentally in [11].

The motion with constant velocity is a particular case of vibrations, namely with zero frequency. Under these conditions the phase factor (14) is reduced to $f_m(t) = \exp(-\mathrm{i}\omega_m t)$ with $\omega_m = \boldsymbol{k}_m \cdot \boldsymbol{v}$ which is the Doppler frequency shift in the direction of the wave vector $\boldsymbol{k}_m$.

According to the general equation (31) and by (14), (19), (23), (28), (29) we obtain

$$\hat{G}_{21}(t,t') = f_0^*(t') \sum_n J_n(b_{m'}) \exp\{-\mathrm{i}n(\Omega t + \varphi)\} \hat{\mathcal{G}}^{(n)}_{21}(t - t') \qquad (35)$$

where

$$\hat{\mathcal{G}}^{(n)}_{21}(t) = \int \frac{\mathrm{d}\omega}{2\pi} \hat{\mathcal{R}}_2(\omega + \Omega n - \omega_{m'}) \hat{\mathcal{R}}_1(\omega) \exp(-\mathrm{i}\omega t). \qquad (36)$$

Here index m' denotes the direction of the beam between the two crystals.

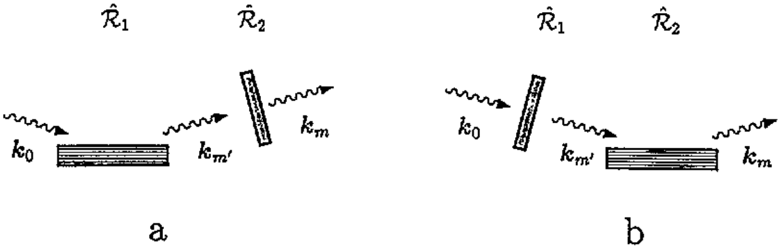

Figure 1. Double-crystal scattering arrangements. (a) The vibrating Bragg reflector ($\hat{\mathcal{R}}_1$) and the resonance absorber ($\hat{\mathcal{R}}_2$) moving with constant velocity. (b) The same as before, but with the opposite sequence of the scatterers.

If the absorber is the first and the reflector is the second (figure 1(b)) then a similar calculation leads to the different result

$$\hat{G}_{21}(t,t') = f_m(t)\sum_n J_n(b_{m'})\exp\{\mathrm{i}n(\Omega t'+\varphi)\}\hat{\mathcal{G}}_{21}^{(n)}(t-t') \qquad (37)$$

where

$$\hat{\mathcal{G}}_{21}^{(n)}(t) = \int\frac{\mathrm{d}\omega}{2\pi}\hat{\mathcal{R}}_2(\omega)\hat{\mathcal{R}}_1(\omega+\Omega n-\omega_{m'})\exp(-\mathrm{i}\omega t). \qquad (38)$$

In the following we shall assume that the synchrotron radiation pulses are not locked in phase with crystal vibrations. Therefore in calculating the intensity we have to average equation (5) additionally over the phase φ. As a result we obtain for the time spectrum

$$I(t) = N_0\sum_n W_n|\hat{\mathcal{G}}_{21}^{(n)}(t)|^2 \qquad (39)$$

where $W_n = J_n^2(b_{m'})$.

In practice it is difficult to excite vibrations with the amplitudes homogeneous over the whole volume of the crystal. However, if the amplitudes are homogeneous in sufficiently large volumes with linear dimensions about 10 μm (see e.g. the discussion in [10, 11]) then (39) is still applicable but with the weights W_n averaged over a distribution of the amplitudes or correspondingly over the modulation index. The Rayleigh function is known to be the distribution that gives a good agreement with experimental results

$$P(b) = (b/2b_R^2)\exp(-b^2/b_R^2). \qquad (40)$$

Here b_R is the most probable value of the modulation index. As a result of the averaging procedure we have to replace $W_n = J_n^2(b)$ by

$$W_n = \int_0^\infty \mathrm{d}b\,J_n^2(b)P(b) = \exp(-b_R^2/2)I_n(b_R^2/2). \qquad (41)$$

Here $I_n(x)$ is the modified Bessel function of integer index.

The scattering scheme considered in the present section can be used for measurements of the quasispectral characteristics of the radiation scattered from the vibrated crystal (as was performed e.g. in [11]). In this case the crystal moving with constant velocity is used as a resonance analyser. Experimentally the velocity dependence of the intensity integrated over the time region between successive synchrotron radiation pulses with exclusion of a short time window $0 < t < t_w$ just after the synchrotron radiation pulse is measured. This dependence is described by the following expression

$$\bar{I}(v) = N_0\sum_n W_n\int_{t_w}^\infty \mathrm{d}t\,|\hat{\mathcal{G}}_{21}^{(n)}(v,t)|^2. \qquad (42)$$

5.3. Forward scattering

In this section we shall consider a special arrangement of forward scattering by two identical crystals containing the resonant nuclei. The first crystal is vibrating while the second one is at rest ($\omega_{m'} = 0$). Each crystal is characterized by the same scattering amplitude $\mathcal{R}(\omega)$ and response function $\mathcal{G}(t)$. We assume again no phase locking with the synchrotron radiation pulses. In this case we may use (35), (36) and (39) for calculating the time spectrum.

We shall assume also that the frequency of vibrations Ω is so large that the distributions $\mathcal{R}(\omega)$ and $\mathcal{R}(\omega + \Omega n)$ practically do not overlap. In this case (36) can be calculated in an analytical form for all values of n except $n = 0$

$$\mathcal{G}_{21}^{(n)}(t) = \mathcal{G}(t)[1 + \exp(\mathrm{i}\Omega n)] \qquad n \neq 0. \tag{43}$$

Substitution of this function in (39) leads to the following expression for the time spectrum of the double-crystal system:

$$I(t) = N_0\left[W_0|\mathcal{G}_{21}^{(0)}(t)|^2 + 4|\mathcal{G}(t)|^2 \sum_{n\neq 0} W_n \cos^2(\Omega n/2)\right]. \tag{44}$$

If b_R is small enough then according to (41) the coefficients W_n decrease rapidly and we come to an interesting result. Although the crystal vibrations are not synchronized with the synchrotron radiation pulses the time spectrum has periodic modulation at the frequency of the crystal vibration. This effect can be directly used for studying excitations in the crystals caused by periodic perturbations.

Acknowledgments

The authors would like to thank Dr H D Rüter and J Metge for their helpful remarks on the manuscript. V G Kohn acknowledges the financial support of the Russian Foundation for Fundamental Research, grant No 93-02-2590 and the International Science Foundation, grant No N9C000. Yu V Shvyd'ko acknowledges the receipt of an Alexander von Humboldt Research Fellowship while at Hamburg University.

Appendix

To calculate the nuclear current density induced by an electromagnetic wave under multiple diffraction in a vibrating crystal we shall follow the method previously used in [9] for the similar calculations under the conditions of forward scattering.

It is convenient to start from the Fourier expansion of the nuclear current density in terms of momentum

$$\jmath(\boldsymbol{r}, t) = \int \frac{\mathrm{d}\boldsymbol{k}}{(2\pi)^3} \exp(\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}) \sum_a \exp(-\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}_a)\langle\psi_a(t)|\hat{\jmath}_a(\boldsymbol{k})|\psi_a(t)\rangle. \tag{A1}$$

Here the summation is performed over all nuclei of the crystal; $\psi_a(t)$ is the time-dependent wave function of the nucleus in the crystal site a with position vector $\boldsymbol{r}_a$; $\hat{\jmath}_a(\boldsymbol{k})$ is the nuclear current density operator in $\boldsymbol{k}$-space.

The current density for the individual nucleus in the first non-vanishing order of perturbation theory is equal to

$$\langle\psi_a(t)|\hat{\jmath}_a(\boldsymbol{k})|\psi_a(t)\rangle = \frac{1}{\mathrm{i}(2I_0+1)} \sum_{\mathrm{ge}} \int_{-\infty}^{t} \mathrm{d}\tau \exp\{-\mathrm{i}(\omega_{\mathrm{ge}} - \mathrm{i}\Gamma_0/2)(t-\tau)\}$$
$$\times \langle \mathrm{g}|\hat{\jmath}_a(\boldsymbol{k})|\mathrm{e}\rangle\langle \mathrm{e}|\hat{V}_a(\tau)|\mathrm{g}\rangle. \tag{A2}$$

Here $|\mathrm{g}\rangle$ and $|\mathrm{e}\rangle$ are the ground and excited state vectors of a nucleus respectively; I_0 is the nuclear spin in the ground state; ω_{ge} is the frequency of the corresponding resonance transition; $\hat{V}_a(t)$ is the time-dependent Hamiltonian of interaction of the ath nucleus with the γ-radiation. It is defined in a standard way

$$\hat{V}_a(t) = -c^{-1} \int \mathrm{d}\boldsymbol{r}\, \hat{\boldsymbol{j}}_a(\boldsymbol{r} - \boldsymbol{r}_a) \cdot \boldsymbol{A}(\boldsymbol{r}, t) \tag{A3}$$

where $\boldsymbol{A}(\boldsymbol{r}, t)$ is a vector potential. We shall consider the cases where the frequency of the vibration Ω is much less than the frequency of the radiation ω. Taking this into account and using the Coulomb gauge with a zero scalar potential we have the relation $\boldsymbol{A}(\boldsymbol{r}, t) = -\mathrm{i}c\omega^{-1}\boldsymbol{E}(\boldsymbol{r}, t)$. Representing $\boldsymbol{E}(\boldsymbol{r}, t)$ as a Bloch wave, equation (8), and using the slowly varying envelope approximation we obtain after simple transformations

$$\hat{V}_a(t) = \mathrm{i}\omega^{-1} \sum_m \hat{\boldsymbol{j}}_a(-\boldsymbol{k}_m)\boldsymbol{E}_m(z, t) \exp\{\mathrm{i}(\boldsymbol{k}_m \cdot \boldsymbol{r}_a - \omega t)\}. \tag{A4}$$

The position vector of the ath nucleus $\boldsymbol{r}_a$ can be presented as a sum of the equilibrium position vector $\boldsymbol{R}_a$ and the displacement vector from this position $\boldsymbol{u}_a(t)$ due to vibrations (both thermal and externally induced), i.e. $\boldsymbol{r}_a = \boldsymbol{R}_a + \boldsymbol{u}_a(t)$. The current density, given by equation (A1), should be averaged over the initial state of the crystal, assuming its thermal equilibrium. This will result in the appearance of the Lamb–Mössbauer factors $\exp[-Z_a(\boldsymbol{k}_m)]$ in the final expression for the current density. In the present paper we shall neglect coherent inelastic scattering from phonons, therefore we shall assume henceforth that $\boldsymbol{u}_a(t)$ describes the vibrations induced only by an external force. The possibility of inhomogeneous broadening of the nuclear resonance will be accounted for by replacing Γ_0 with Γ ($\Gamma \geqslant \Gamma_0$). Substitution of (A4) in (A2) and then of (A2) in (A1) results in

$$j^s(\boldsymbol{r}, t) = \int \frac{\mathrm{d}\boldsymbol{k}}{(2\pi)^3} \exp\{\mathrm{i}(\boldsymbol{k} \cdot \boldsymbol{r} - \omega t)\} \sum_a \sum_{m's'} \sum_{\mathrm{ge}} \frac{\langle \mathrm{g}|\hat{J}_a^s(\boldsymbol{k})|\mathrm{e}\rangle\langle \mathrm{e}|\hat{J}_a^{s'}(-\boldsymbol{k}_{m'})|\mathrm{g}\rangle}{\omega(2I_0 + 1)}$$
$$\times \exp\{-\mathrm{i}\boldsymbol{k} \cdot \boldsymbol{u}_a(t)\} \int_{-\infty}^{t} \mathrm{d}\tau\, e(\omega - \omega_{\mathrm{ge}}, t - \tau) \exp\{\mathrm{i}\boldsymbol{k}_{m'}\boldsymbol{u}_a(\tau)\} E_{m'}^{s'}(z, \tau). \tag{A5}$$

Here we use the notation (14) and

$$\langle \mathrm{g}|\hat{J}_a^s(\boldsymbol{k})|\mathrm{e}\rangle = \langle \mathrm{g}|\hat{j}_a^s(\boldsymbol{k})|\mathrm{e}\rangle \exp\{-\mathrm{i}\boldsymbol{k} \cdot \boldsymbol{R}_a - \tfrac{1}{2}Z_a(\boldsymbol{k})\} \tag{A6}$$

as a transition current between the states $\langle \mathrm{g}|$ and $\langle \mathrm{e}|$ at the ath site.

When all nuclei are vibrated in unison the vectors $\boldsymbol{u}_a$ do not depend on a. Let us represent the equilibrium position of nucleus as the sum $\boldsymbol{R}_a = \boldsymbol{r}_j + \boldsymbol{\rho}_l$, where $\boldsymbol{r}_j$ is a position of the unit cell and $\boldsymbol{\rho}_l$ is a position of nucleus inside the unit cell. Taking into account the relation $\boldsymbol{k}_m \cdot \boldsymbol{r}_j = 2\pi n$ and

$$\sum_j \exp(-\mathrm{i}\boldsymbol{k} \cdot \boldsymbol{r}_j) = \frac{(2\pi)^3}{V_0} \sum_n \delta(\boldsymbol{k} - \boldsymbol{k}_n) \tag{A7}$$

with V_0 as the unit cell volume we obtain finally after simple transformations (13)–(15).

References

[1] Ruby S L and Bolef D I 1960 *Phys. Rev. Lett.* 5 5

[2] Srivastava J K 1983 *Advances in Mössbauer Spectroscopy* ed B V Thosar, P K Iyengar, J K Srivastava and S C Bhargava (Amsterdam: Elsevier) p 761

[3] Asher J, Cranshaw T E and O'Connor D A 1974 *J. Phys. A: Math. Gen.* 7 410

[4] Mitin A V 1976 *Sov. Quantum Electron.* 3 840

[5] Monahan J E and Perlow G J 1979 *Phys. Rev.* A **20** 1499

[6] Tsankov L T 1980 *J. Phys. A: Math. Gen.* **13** 2959, 2969

[7] Baryshevsky V G 1985 *J. Phys. C: Solid State Phys.* **18** 191

[8] Dzyblik A Yu 1984 *Phys. Status Solidi* b **123** 53, **134** 503

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

[10] Shvyd'ko Yu V, Hertrich T, Sedov V E, Smirnov G V, van Bürck U, Mössbauer R L and Chumakov A I 1992 *Europhys. Lett.* **19** 723

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

[12] Rüffer R, Gerdau E, Grote M, Hollatz R, Röhlsberger R Rüter H D and Sturhahn W 1990 *Hyperfine Interact.* **61** 1279; 1991 *Nucl. Instrum. Methods* A **303** 495

[13] Rüffer R 1992 *Synchrotron Radiat. News* **5** 25

[14] Gerdau E and van Bürck U 1994 *Resonant Anomalous X-Ray Scattering* ed G Materlik, C J Sparks and K Fischer (Amsterdam: Elsevier) pp 589–607

[15] Kagan Yu M, Afanas'ev A M and Kohn V G 1979 *J. Phys. C: Solid State Phys.* **12** 615

[16] Popov S L, Smirnov G V and Shvyd'ko Yu V 1989 *Pis. Zh. Eksp. Teor. Fiz.* **49** 651 (Engl. Transl. 1989 *JETP Lett.* **49** 747); 1990 *Hyperfine Interact.* **58** 2463

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

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

[19] Allen L and Eberly J H 1975 *Optical Resonance and Two-Level Atoms* (New York: Wiley) part I, section 6

[20] Abramowitz M and Stegun I A (ed) 1964 *Handbook of Mathematical Functions* (New York: Dover)

[21] Afanas'ev A M and Kagan Yu 1965 *Zh. Eksp. Teor. Fiz.* **48** 327 (Engl. Transl. 1965 *Sov. Phys.–JETP* **21** 215)

[22] Kagan Yu, Afanas'ev A M and Perstnev I P 1968 *Zh. Eksp. Teor. Fiz.* **54** 1530 (Engl. Transl. 1968 *Sov. Phys.–JETP* **27** 819)

[23] Hannon J P and Trammell G T 1968 *Phys. Rev.* **169** 315

[24] Hannon J P and Trammell G T 1969 *Phys. Rev.* **186** 306

[25] Kohn V G 1994 *Zh. Eksp. Teor. Fiz.* **105** 665 (Engl. Transl. 1994 *Sov. Phys.–JETP* **78** 357)

[26] Bömmel H E 1962 *Proc. 2nd Int. Conf. on the Mössbauer Effect (Saclay)* (New York: Wiley) p 229