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Mössbauer filtration of synchrotron radiation: multipulse regime

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Abstract. Mössbauer filtration of synchrotron radiation (SR) in the multipulse regime is considered. Phase correlations causes interference between resonant responses of single pulses of SR sequence. Therefore, both the coherent amplification of response intensity and the variation in form of free-induction decay take place. Numerical analysis of theoretical expressions has been carried out.

1. Introduction

Recently experiments with synchrotron radiation (SR) have aroused a great amount of interest in Mössbauer spectroscopy. Usually experimenters investigate the time structure of the resonant response of the Mössbauer nuclei system to the SR pulse and modulation of time dependences of scattered radiation due to hyperfine splitting of nuclear levels. Experiments were made both on diffraction scattering [1] and forward scattering [2]. The frequency of SR 'flashes' was larger than the reverse lifetime Γ of the nuclear in order to study the time structure of the resonant response to a single SR pulse. However, in SR sources the regime can be realized when the time interval between pulses is shorter than $1/\Gamma$. In this case the incident radiation must be considered as the sequence of incoherent pulses with additional phase relations (this situation looks like that for an optical inteferometer with an incoherent source). Since the incoming wave is coherent to wave outgoing from the resonant sample, then interference effects for the intensity of scattered radiation are possible. They are essential only within the resonant response sequence when the coherent time of the single responses are of the order of the time intervals between them. It can cause both amplification of response signal and variation in its time structure. The problem is considered in this paper.

2. Formalism

The semiclassical Maxwell–Bloch equations formalism in the slowly changing approximation is used for the theoretical calculations [3].

The original system of equations for the γ-radiation field $A(r, t)$ and the density matrix ρ of the nuclear system has the form

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$$\begin{aligned}
&\Delta \boldsymbol{A} - (1/c^2)\partial^2 \boldsymbol{A}/\partial t^2 = -(4\pi/c)\langle j\rangle \\
&\langle j\rangle = \mathrm{Tr}(\rho j) = \sum \rho^{eg}_{0\beta} j^{ge}_{\beta 0} \\
&\partial \rho^{eg}_{0\beta}/\partial t = -\mathrm{i}(\omega_0 + \omega_{eg} + \Omega_{0\beta} - \mathrm{i}\Gamma/2)\rho^{eg}_{0\beta} - (\mathrm{i}/h)\rho^{(0)}_g j^{eg}_{0\beta} \cdot \boldsymbol{A} \\
&j^{ge}_{\beta 0} = \sum_m j^{ge}_{\beta 0}(r_m)\delta(r - r^0_m).
\end{aligned} \tag{1}$$

In (1), j is the nuclear transition current operator, ω_{eg} is the hyperfine component of γ-transition frequency (the quadrupole component is neglected), $\Omega_{0\beta}$ is the lattice vibration frequency ($\Omega_{00} = 0$), $\rho^{(0)}_g = 1/(2I_g + 1)$ is the equilibrium population of the ground nuclear state and r^0_m is the radius vector of a nucleus when there are no lattice vibrations. The system (1) is correct for all γ-optics domain excepting the γ-laser regime. According to [3], $\boldsymbol{A}(r, t)$, $\rho^{eg}_{0\beta}(r, t)$ can be written as

$$\begin{aligned}
&\boldsymbol{A}(r, t) = \sum_H \boldsymbol{a}_H(r, t)\exp\{-\mathrm{i}[\omega t - (k + H)\cdot r]\} \\
&\rho^{eg}_{0\beta}(r, t) = \sum_H \sigma^{eg}_{0\beta(H)}(r, t)\exp\{-\mathrm{i}[\omega t - (k + H)\cdot r]\}
\end{aligned} \tag{1a}$$

where H are the reciprocal-lattice vectors. The following conditions are true.

Firstly,

$$|\Delta \boldsymbol{a}_H| \ll |(k + H)\cdot\nabla)\boldsymbol{a}_H| \ll |(k + H)|^2 \cdot |\boldsymbol{a}_H|$$

(here, we are not considering the grazing-incidence case, when $|\Delta \boldsymbol{a}_H| \sim |(k+H)\cdot\nabla)\boldsymbol{a}_H|$ [4, 5]). Secondly,

$$|\partial^2 \boldsymbol{a}_H/\partial t^2| \ll (\omega/c)|\partial \boldsymbol{a}_H/\partial t| \ll (\omega^2/c^2)|\boldsymbol{a}_H| \qquad |k| = k \qquad k^2 = (\omega/c)^2.$$

Then, using (1a) and the above two conditions the shortened system of Maxwell–Bloch equations can be obtained:

$$\begin{aligned}
&(n_H\cdot\nabla)\boldsymbol{a}_H + (1/c)\partial \boldsymbol{a}_H/\partial t + \mathrm{i}(k\alpha_H/2)\boldsymbol{a}_H = 2(\pi\mathrm{i}/h)\langle j\rangle \\
&\partial \sigma^{eg}_{0\beta}/\partial t = -\mathrm{i}(\Delta - \omega_{eg} - \Omega_{0\beta} + \mathrm{i}\Gamma/2)\sigma^{eg}_{0\beta} - (\mathrm{i}/h)\rho^{(0)}_g j^{eg}_{0\beta}\cdot\boldsymbol{a}_H
\end{aligned} \tag{2}$$

where $n_H = (k + H)/|(k + H)|$, $\alpha_H = H\cdot(H + 2k)/k^2$. In order to solve the system (2), we make the substitution:

$$\begin{aligned}
&\boldsymbol{a}_H(r, t) = \int_{-\infty}^{\infty} \mathrm{d}\nu\, \boldsymbol{a}_H(r, \nu)\exp(-\mathrm{i}\nu t) \\
&\sigma^{eg}_{0\beta}(r, t) = \int_{-\infty}^{\infty} \mathrm{d}\nu\, \sigma^{eg}_{0\beta}(r, \nu)\exp(-\mathrm{i}\nu t).
\end{aligned} \tag{2a}$$

From (2) and (2a) we can obtain the system of equations for Fourier images $\boldsymbol{a}_H(r, \nu)$ for the case of coherent Mössbauer scattering ($\Gamma \ll \Omega_{0\beta}$ when $\beta \neq 0$):

$$\begin{aligned}
&(n_H\cdot\nabla)\boldsymbol{a}_H(r, \nu) + \frac{\mathrm{i}\nu}{c}\boldsymbol{a}_H(r, \nu) + \mathrm{i}k\frac{\alpha_H}{2}\boldsymbol{a}_H(r, \nu) \\
&\quad = \frac{-2\pi f\eta}{cV_0}\sum_{e,g} J_{ge}\sum_{H_1} S_{HH_1}\frac{J_{eg}\cdot\boldsymbol{a}_{H_1}(r, \nu)}{h(2I_g + 1)(\Delta + \nu + \omega_{eg} + \mathrm{i}\Gamma/2)}
\end{aligned} \tag{3}$$

where f is the Debye–Waller factor and η is the abundance of the resonant nuclei. The matrix elements $J_{eg} = f^{-1/2} j_{00}^{eg}$ are the same for all resonant nuclei of the sample. $S_{HH_1} = \sum_{\text{unit cell}} \exp[i(\boldsymbol{H} - \boldsymbol{H}_1) \cdot \boldsymbol{r}_j]$ is the nuclear structural factor. V_0 is the unit-cell volume and $\Delta = \omega - \omega_0$ is the Doppler shift of the incident radiation frequency.

Using [6], equation (3) can be considerably simplified when hyperfine splitting of nuclear levels is absent ($\omega_{eg} = 0$):

$$(\boldsymbol{n}_H \cdot \nabla) a_H^s(\boldsymbol{r}, \nu) + (i\nu/c) a_H^s(\boldsymbol{r}, \nu) + i(k\alpha_H/2) a_H^s(\boldsymbol{r}, \nu) = i(k/2) F_{HH_1}^s a_{H_1}^s(\boldsymbol{r}, \nu) \tag{3a}$$

where $a_H^s = (e_H^{s*} \cdot a_H)$, e_H^s is the unit transverse polarization vector. The nuclear scattering amplitude $F_{HH_1}^s$ has the form

$$F_{HH_1}^s = (1/4V_0 k) f \Gamma \sigma_{abs} S_{HH_1} P_{HH_1}^s / \Delta + \nu + i\Gamma/2$$

where σ_{abs} is the nuclear resonant absorption cross-section and $P_{HH_1}^s = (e_H^{s*} \cdot e_{H_1})$ is the nuclear polarization factor. The system (3a) may be easily generalized when one adds a Rayleigh electron scattering amplitude to $F_{HH_1}^s$. Its form can be found in [7]. To solve (3a) we use the substitution

$$a_H^s(\boldsymbol{r}, \nu) = a_H^s(\nu) \exp[ikp^s(\nu)z]$$

which is valid when the incident radiation is a plane wave. The solution of the corresponding algebraic system for $a_H^s(\nu)$ and $p^s(\nu)$ is already known. The values $a_H^s(\nu)$ are expressed via the amplitude of the incident radiation field $a_{inc}^s(\nu)$ and the boundary conditions for each specific case of scattering. Let us consider the following two cases of coherent scattering which can be realized in Mössbauer experiments with SR.

(1) First consider forward scattering without any diffraction process ($\boldsymbol{H} = \boldsymbol{0}$). Then for radiation passing through the sample of thickness L the expression

$$a_{fs}^s(\nu, L) = a_{inc}^s(\nu) \exp(ikF_{00}^s L/2) \tag{3b}$$

is correct.

(2) Next the two-wave approximation of diffraction scattering under Bragg geometry conditions in a thick crystal is considered. Then for the diffracted component of scattered radiation the expression

$$a_{ds}^s(\nu) = a_{inc}^s(\nu)\{F_{00}^s + F_{0H}^s + \alpha_H - [(F_{00}^s + F_{HH}^s + \alpha_H)^2 - 4F_{0H}^s F_{H0}^s]^{1/2}\}/2F_{0H}^s \tag{3c}$$

is true.

3. Time dependences of the multipulse regime

Let the sample containing the Mössbauer nuclei be subjected to a SR pulse sequence which has been reflected from an x-ray monochromator. It is assumed that the pulses have the same shape and duration T. Let t_i be the moment of emission of the ith quantum into the first pulse of the sequence. (It is continuously changing within the $[0, T]$ interval.) Then there is a quantum into the kth pulse, emitted at the $t_i + t_k$ moment, where t_k is the time

interval between the first and kth pulses. Summing over i and k we obtain the amplitude of the incident radiation field:

$$A^{s}_{inc}(r,t) = \sum_{i,k} \Psi(t_i) A^{s(i)}_{inc}(u - t_i - t_k)$$
$$A^{s(i)}_{inc}(u - t_i - t_k) = e^{s}_{inc}\theta(u - t_i - t_k)\exp[(-i\omega - \gamma/2)(u - t_i - t_k)]/V^{1/2} \tag{4}$$

where $u = t - n \cdot r/c$, $n = k/|k|$. $\theta(\ldots)$ is the step function. e^{s}_{inc} is the unit polarization vector of incident radiation. γ is the frequency distribution of SR pulses ($\gamma \gg 1/T \gg \Gamma$), $\Psi(t_i) = \theta(t_i) - \theta(t_i - T)$ and V is the sample volume.

Then the amplitude of scattered radiation field has the form

$$A^{s}_{sc}(t,r) = \sum_{i,k} \Psi(t_i) A^{s(i)}_{sc}(u - t_i - t_k) \tag{4a}$$

where the amplitude $A^{s(i)}_{sc}(u - t_i - t_k)$ can be calculated using (2a) and (3a)–(3c) [8].

For forward scattering it has the following form:

$$A^{s(i)}_{fs}(u - t_i - t_k) = e^{s}_{0}\theta(u - t_i - t_k)\exp[-i\omega(u - t_i - t_k)]\, G^{s}_{fs}(u - t_i - t_k)$$
$$G^{s}_{fs}(u - t_i - t_k) = i\exp[ik\chi^{s}_{00}L](\exp[-\gamma(u - t_i - t_k)/2] - (\Gamma/\gamma) \tag{4b}$$
$$\times \exp[(i\Delta - \Gamma/2)(u - t_i - t_k)](\beta/4\Gamma^2(u - t_i - t_k)^{1/2})J_1(\beta(u - t_i - t_k)^{1/2})$$

where χ^{s}_{ij} is the Rayleigh electron scattering amplitude, β is the nuclear resonant absorption coefficient [6] and J_1 is the Bessel function of first kind.

For diffraction scattering the following expression may be obtained:

$$A^{s(i)}_{ds}(u - t_i - t_k) = e^{s}_{H}\theta(u - t_i - t_k)\exp[-i\omega(u - t_i - t_k)]\, G^{s}_{dS}(\alpha_H, u - t_i - t_k)$$
$$G^{s}_{ds}(\alpha_H, u - t_i - t_k) = R\exp[-\gamma(u - t_i - t_k)/2]$$
$$\pm i\exp[i(\Delta + 2A + i\Gamma/2)(u - t_i - t_k)] \tag{4c}$$
$$\times (C/2xb^{s}_{0H}\gamma)J_1(C(u - t_i - t_k))/(u - t_i - t_k)$$

where

$$R = (\alpha_H + 2(F^{s}_{00} + F^{s}_{HH}) - [(\alpha_H + 2(F^{s}_{00} + \chi^{s}_{00})^2$$
$$- 4(F^{s}_{0H} + \chi^{s}_{0H})(F^{s}_{H0} + \chi^{s}_{H0})]^{1/2}/2(F^{s}_{0H} + \chi^{s}_{0H})(\nu = -i\gamma/2)$$

is the coefficient of total SR Rayleigh scattering,

$$C = 2(A^2 - B)^{1/2} \qquad x = 2F/(\alpha_H + 2\chi^{s}_{00}) \qquad b^{s}_{0H} = S_{0H}\, p^{s}_{0H}/n_0$$
$$A = [\alpha_H + 2\chi^{s}_{00} - 2\mathrm{Re}(\chi^{s}_{H0}b^{s}_{0H})]F/[(\alpha_H + 2\chi^{s}_{00})^2 - 4|\chi^{s}_{0H}|^2] \tag{4d}$$
$$B = (1 - |b^{s}_{0H}|^2)F^2/[(\alpha_H + 2\chi^{s}_{00})^2 - 4|\chi^{s}_{0H}|^2]$$
$$F = F^{s}_{00}(\Delta + \nu + i\Gamma/2).$$

In (4d), n_0 is the number of Mössbauer nuclei in the unit cell. When Rayleigh electron scattering is structurally forbidden, then (4d) is simplified to

$$C = x|b^{s}_{0H}| \qquad A = -x/2 \qquad R = -2x|b^{s}_{0H}|^2/\gamma b^{s}_{0H}.$$

Now let us obtain the intensity of scattered radiation. It is calculated as the modulus of the Poynting vector

$$I^{s}_{sc}(u) = (c/4\pi)h\omega|A^{s}_{sc}(u)|^2$$

or, using (4), (4*a*) and (4*b*),

$$I^{s}_{sc}(u) \sim \sum_{k,l}\sum_{i,j} \Psi(t_i)\Psi(t_j)\exp(i\varphi_{ij})\exp(i\varphi_{kl})G^{s}_{sc}(u-t_{ik})G^{s*}_{sc}(u-t_{jl}) \qquad (5)$$

where $\varphi_{ij} = \omega(t_i - t_j)$, $\varphi_{kl} = \omega(t_k - t_l)$ and $t_{ik} = t_i - t_k$. As SR is incoherent radiation, the moments of emission of quanta in a single pulse are uncorrelated with each other. So it is necessary to average the expression for the intensity of scattered radiation over the phase φ_{ij}. Therefore the quantity $\langle\exp(i\varphi_{ij})\rangle = (1/2\pi)\int_0^{2\pi} d\varphi_{ij}\exp(i\varphi_{ij})$ appears instead of $\exp(i\varphi_{ij})$ in (5). So (5) is not zero only when $i = j$. The phase φ_{kl} is not averaged because t_k and t_l are the defined values.

After this substitution we have

$$\langle I^{s}_{sc}(u)\rangle_{\varphi_{ij}} \sim \sum_{k,l}\sum_{i} \Psi(t_i)\exp(i\varphi_{kl})\,G^{s}_{sc}(u-t_{ik})G^{s*}_{sc}(u-t_{il}). \qquad (5a)$$

As the Mössbauer experiment is statistical, it is necessary to average (5*a*) over t_i. Then we obtain the expression

$$\langle\langle I^{s}_{sc}(u)\rangle_{\varphi_{ij}}\rangle_{t_i} \sim \frac{N}{T}\sum_{k,l}\exp(i\varphi_{kl})\int_{-\infty}^{\infty} dt_i\,\Psi(t_i)G^{s}_{sc}(u-t_{ik})G^{s*}_{sc}(u-t_{il}) \qquad (5b)$$

where N is the number of quanta in a single pulse of the sequence. For free-induction decay (FID) ($u > T + \max(t_k)$) the following expression is correct:

$$\langle\langle I^{s}_{sc}(u)\rangle_{\varphi_{ij}}\rangle_{t_i} \sim N\sum_{k,l}\exp(i\varphi_{kl})\,G^{s}_{res}(u-t_k)G^{s*}_{res}(u-t_l) \qquad (5c)$$

where

$$G^{s}_{res}(u) = G^{s}_{fs}(u) - i\exp(ik\chi^{s}_{00}L - \gamma u/2)$$
$$G^{s}_{res}(u) = G^{s}_{ds}(u) - R\exp[-\gamma u/2]$$

for forward and diffraction scattering, respectively. Equation (5*c*) shows that there may be interference of resonant responses of single pulses of sequence. The value and shape of the FID (5*c*) depend on the time intervals t_{kl}.

4. Numerical calculations and analysis

When $|t_{kl}| \geqslant \Gamma$ the time dependences of FID are similar to those in [9, 10]. Now let the condition $|t_{kl}| < \Gamma$ be fulfilled. For forward scattering, numerical calculations have been carried out for an α-Fe sample of thickness 7 μm [1]. For diffraction scattering, an α-Fe_2O_3 sample and pure nuclear diffraction (7,7,7) have been considered because in this case Rayleigh electron diffraction is essentially suppressed. The following dependences are shown in the figures:

$$P(\tau) = \exp\{-[\tau - \Gamma\max(t_k)]\}\langle\langle I^{s}_{sc}(\tau)\rangle_{\varphi_{ij}}\rangle_{t_i}$$

where $\tau = \Gamma t$, $t > \max(t_k)$. Calculating the particular values of the phase φ_{kl} we suppose that the time intervals t_k are defined with the precision 0.1 ns [1]. The main peculiarities of FID due to resonant response interference are analysed for a two-pulse SR sequence. In this case, using (5*c*), the following expressions for $P(\tau)(\tau > \tau_2 = \Gamma t_2)$ can be obtained:

(a) for forward scattering ($\Delta = 0$),

$$P(\tau) \sim [(\beta/4\tau)J_1^2((\beta\tau)^{1/2})\exp(-\tau_2) + [\beta/4(\tau-\tau_2)]J_1^2([\beta(\tau-\tau_2)]^{1/2}) + (\beta/2)\exp(-\tau_2/2)J_1((\beta\tau)^{1/2})J_1([\beta(\tau-\tau_2)]^{1/2})\cos\varphi_{12}/[\tau(\tau-\tau_2)]^{1/2} \tag{6a}$$

where $\beta \simeq 106.3$ [1];

(b) for diffraction scattering ($\Delta = -x_0$, α_H is the fixed value),

$$P^s(\alpha_H, \tau) \sim \tfrac{1}{4}[J_1^2(y^s\tau)\exp(-\tau_2)/\tau^2 + J_1^2(y^s(\tau-\tau_2))/(\tau-\tau_2)^2 + 2\exp(-\tau_2/2)J_1(y^s\tau)J_1(y^s(\tau-\tau_2))\cos\varphi_{12}] \tag{6b}$$

where $y^s = x_0|S_{0H}|P^s_{0H}/\Gamma$, $|S_{0H}| \simeq 0.7042$, $P^\pi_{0H} \simeq 0.279$ [2], $x_0 \simeq 46\Gamma$ [2]. First, we shall analyse (6*a*). If the phase $\varphi_{12} \to 2\pi$, then constructive interference takes place. In this case,

$$P(\tau) \sim \{(\beta/4\tau)^{1/2}J_1((\beta\tau)^{1/2})\exp(-\tau_2/2) + [\beta/4(\tau-\tau_2)]^{1/2}J_1([\beta(\tau-\tau_2)]^{1/2})\}^2. \tag{7a}$$

If $\tau_2 \ll \tau_0 = (3.8)^2/\beta$, the first null of the $J_1((\beta\tau)^{1/2})$ Bessel function, then the conditions

$$(\beta/4\tau)^{1/2}J_1((\beta\tau)^{1/2}) \simeq [\beta/4(\tau-\tau_2)]^{1/2}J_1([\beta(\tau-\tau_2)]^{1/2})\exp(\tau_2/2) \simeq 1$$

are fulfilled and

$$P(\tau) \sim [\beta/(\tau-\tau_2)]J_1^2([\beta(\tau-\tau_2)]^{1/2}) \text{(figure 1, curve 1).} \tag{7b}$$

This value is twice that for the case of simple sum of two responses ($\varphi_{12} = 0$).

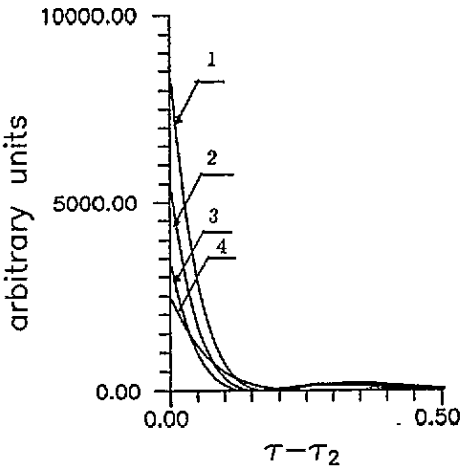

Figure 1. FID dependences for forward scattering (two-pulse regime) ($\tau_0/\Gamma \simeq 19.2$ ns): curve 1, $t_2 = 2.8$ ns ($\varphi_{12} \simeq 6.23$ rad); curve 2, $t_2 = 8.3$ ns ($\varphi_{12} \simeq 0.08$ rad); curve 3, $t_2 = 16.7$ ns ($\varphi_{12} \simeq 6.21$ rad); curve 4, $t_2 = 15.3$ ns ($\varphi_{12} \simeq 3.09$ rad).

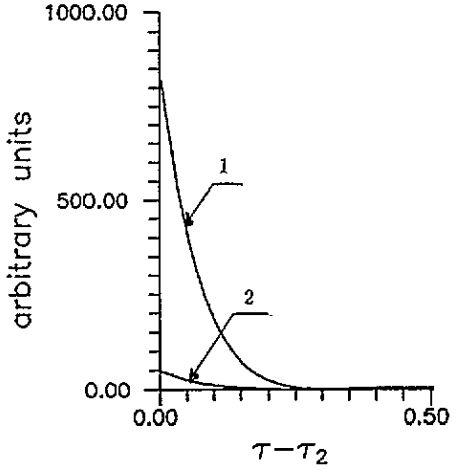

Figure 2. FID dependences for forward scattering (two-pulse regime): curve 1, $t_2 = 1.4$ ns ($\varphi_{12} \simeq 3.11$ rad); curve 2, $t_2 = 6.9$ ns ($\varphi_{12} \simeq 3.24$ rad).

When $\tau_2 < \tau_0$, then for $\tau > \tau_2$ the condition

$$(\beta/4\tau)^{1/2} J_1((\beta\tau)^{1/2}) < [\beta/4(\tau - \tau_2)]^{1/2} J_1([\beta(\tau - \tau_2)]^{1/2})$$

is fulfilled and interference effects are less essential (figure 1, curve 2).

For $\tau_2 \simeq \tau_0$ the condition

$$(\beta/4\tau)^{1/2} J_1((\beta\tau)^{1/2}) \ll [\beta/4(\tau - \tau_2)]^{1/2} J_1([\beta(\tau - \tau_2)]^{1/2})$$

is fulfilled and

$$P(\tau) \to [\beta/4(\tau - \tau_2)] J_1^2([\beta(\tau - \tau_2)]^{1/2}) \tag{7c}$$

(single-pulse response) (figure 1, curve 3).

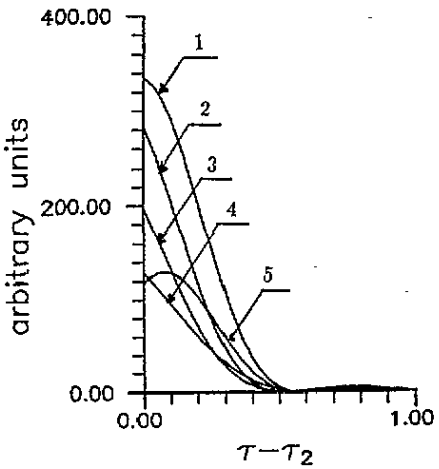

Figure 3. FID dependences for diffraction scattering (two-pulse regime) π polarization of incident radiation ($\tau_{(0)}/\Gamma \simeq 58.7$ ns): curve 1, $t_2 = 2.8$ ns; curve 2, $t_2 = 22.2$ ns ($\varphi_{12} \simeq 0.05$ rad); curve 3, $t_2 = 38.9$ ns ($\varphi_{12} \simeq 6.25$ rad); curve 4, $t_2 = 58.3$ ns ($\varphi_{12} \simeq 0.08$ rad); curve 5, $t_2 = 56.9$ ns ($\varphi_{12} \simeq 3.24$ rad).

If destructive interference takes place ($\varphi_{12} \to \pi$), then

$$P(\tau) \sim \{(\beta/4\tau)^{1/2} J_1((\beta\tau)^{1/2}) \exp(-\tau_2/2) - [\beta/4(\tau - \tau_2)]^{1/2} J_1([\beta(\tau - \tau_2)]^{1/2})\}^2. \tag{7d}$$

Once again, if $\tau_2 \ll \tau_0$, we have

$$P(\tau) \sim \{(\beta/4\tau)^{1/2} J_1((\beta\tau)^{1/2}) - [\beta/4(\tau - \tau_2)]^{1/2} J_1([\beta(\tau - \tau_2)]^{1/2})\}^2 \text{(figure 2, curve 1).} \tag{7e}$$

(In this case, for small $\tau - \tau_2 P(\tau) = (\beta^2/32)^2$ [11].)

If $\tau_2 \to \tau_0$, then $P(\tau)$ is given by (7c) (figure 2, curve 2; figure 1, curve 4).

The same discussion applies to diffraction scattering, when $\alpha_H = 0.68$ μrad [2] is the fixed value.

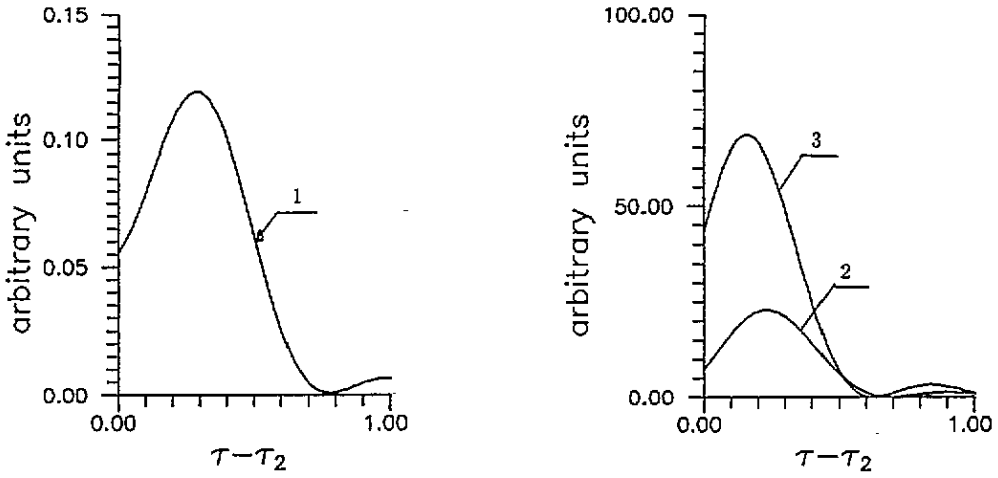

Figure 4. FID dependences for diffraction scattering (two-pulse regime) (π-polarization of incident radiation): curve 1, $t_1 = 1.4$ ns; curve 2, $t_2 = 20.8$ ns ($\varphi_{12} \simeq 3.22$ rad); curve 3, $t_2 = 37.5$ ns ($\varphi_{12} \simeq 3.14$ rad).

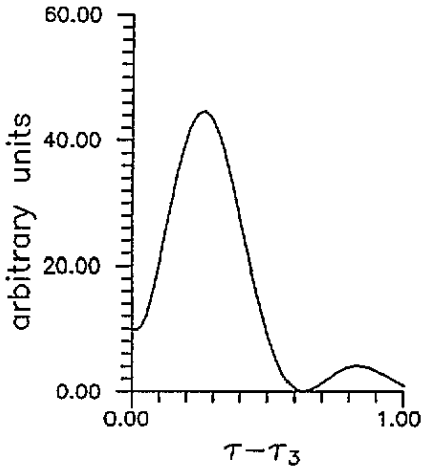

Figure 5. FID dependence for diffraction scattering (three-pulse regime) ($t_2 = 16.7$ ns; $t_3 = 37.4$ ns; $\varphi_{13} \simeq 3.37$ rad).

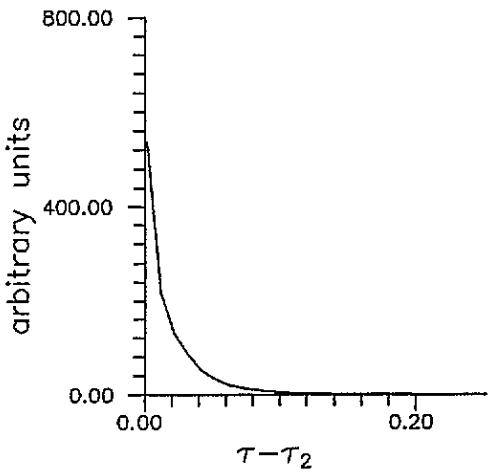

Figure 6. FID dependence for diffraction scattering averaged over a Gaussian distribution (two-pulse regime) ($\alpha_{0H} = 0.68$ μrad; $\sigma = 2.9$ μrad [2]; $t_2 = 1.4$ ns).

If $\varphi_{12} \to 2\pi$ and $\tau_2 \ll \tau_{00} = 3.8/y^s$, the first null Bessel function $J_1(y^s\tau)$; then, using (6*b*), we have

$$P^s(\alpha_H, \tau) \sim J_1^2(y^s(\tau - \tau_2))/(\tau - \tau_2)^2 \text{(figure 3, curve 1)} \tag{8a}$$

instead of

$$P^s(\alpha_H, \tau) \sim J_1^2(y^s(\tau - \tau_2))/[(\tau - \tau_2)^2/2]$$

for the incoherent sum. Coherent effects are attenuated when τ_2 increases (figure 3, curves 2 and 3) and for $\tau_2 \simeq \tau_{00}$ the following can be obtained:

$$P^s(\alpha_H, \tau) \to J_1^2(y^s(\tau - \tau_2))/4(\tau - \tau_2)^2 \tag{8b}$$

(single-pulse response) (figure 3, curve 4).

If $\varphi_{12} \to \pi$, then, using [11], for $\tau_2 \ll \tau_{00}$ we have

$$P^s(\alpha_H, \tau) \sim [-2J_1(y^s(\tau - \tau_2))/(\tau - \tau_2)^2 + y^s J_0(y^s(\tau - \tau_2))/(\tau - \tau_2)] \tag{8c}$$

(figure 4, curve 1). For small $\tau - \tau_2$, $P^s(\alpha_H, \tau) \sim y^{s^6}(\tau - \tau_2)^2/64$. $P^s(\alpha_H, \tau)$ is then given by (8*b*), when $\tau_2 \to \tau_{00}$ (figure 4, curves 2 and 3; figure 3, curve 5). If the sequence consists of three pulses, then choosing particular phases $\varphi_{12}, \varphi_{13}$ the FID curve for diffraction scattering may be strongly transformed (figure 5). Let us consider once again the two-pulse sequence for diffraction scattering when α_H varies within some interval defined by the distribution function $f(\alpha_H)$. It could be, for instance, due to the angular divergence of the incident beam, to the mosaic of the single-crystal sample, or to when the sample is polycrystalline. Then, it is necessary to average (6*b*) over $\alpha_H = -2\sin(2\theta_B)\,\mathrm{d}\theta$:

$$\langle P^s \rangle_{\alpha_H} = \int_{-\infty}^{\infty} \mathrm{d}\alpha_H\, f(\alpha_H) P^s(\alpha_H, \tau). \tag{8d}$$

Let the function $f(\alpha_H)$ be the Gaussian distribution of width σ centred at α_{0H} and $\varphi_{12} \to \pi$. If $\sigma \ll \alpha_{0H}$, then the time dependences (8*d*) are like curves 1–3 in figure 4 and curve 5 in figure 3. If $\sigma \gtrsim \alpha_{0H}$, then the phase $x\tau_2/\Gamma$ changes within a broad interval. These components give the main contribution to (8*d*) for which the phase $\varphi_{12} - (x - x_0)\tau_2/\Gamma$ becomes 2π either within the width σ or on the wings of the Gaussian distribution (figure 6).

5. Conclusion

In this paper we have analysed the main peculiarities of Mössbauer filtration of SR in the multipulse regime. It is shown that different correlations between pulses cause both the sharp amplification and the strong transformation of the FID curve. The coherent amplification of the resonant response signal can lead to a considerable increase in the count rate for Mössbauer experiments with SR for the present synchrotron facilities.

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