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On excitation of isomeric nuclear states in a crystal by synchrotron radiation

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Abstract. The time dependence of delayed radiation is investigated when a pulse of synchrotron radiation undergoes resonance Bragg scattering by the nuclei in a crystal that have an isomeric level with a Mössbauer transition. Instead of the usual exponential law $\exp(-t/\tau_0)$ characteristic of an isolated nucleus, a time dependence in the form $(\tau_0/t)^2 \exp(-t/\tau_0)$ was discovered at small deviations α from the Bragg angle. The acceleration of the decay is connected with the collective nature of the excitation of the nuclei in the crystal—the nuclear exciton formation. The exponential law remains at large α but the intensity decreases as $1/\alpha^2$. In the case of large divergence of the incident beam the law $(\tau_0/t) \exp(-t/\tau_0)$ is obeyed. The frequency distribution of the reflected pulse and the possibility of formation of a resonance structure with sufficient resolution are analysed in detail. The problem of time evolution of the synchrotron radiation pulse in transmission through two crystals—the Bragg reflector and resonance absorber—is solved. It is found that the requirements for the maximum intensity of the delayed radiation and the necessary frequency distribution contradict one another. For example, when α or the delay time are small the resonance structure vanishes. Therefore, for Mössbauer-type experiments it is expedient to measure the intensity integrated over time with the exception of an appreciable initial time interval after the pulse, and with effective use of the reflection corresponding to a deviation from the Bragg angle by the value characteristic for the angle interval of the Bragg scattering.

1. Introduction

Recently, there has been a great interest in direct excitation of low-lying isomeric nuclear levels in the synchrotron radiation field with realisation of the Mössbauer effect (Ruby 1974, Kulipanov and Skrinskii 1977). Paradoxical as it might seem at first sight, the problem of selecting a resonance line of about 10^{-8} eV in width from the continuous radiation spectrum of about tens of kilovolts in width may actually be put forward in view of the unique possibility of combining the features of synchrotron radiation produced by the existing devices and specific properties of the isomeric levels in question as well as the resonance scattering by the nuclei in the crystal.

This relates first of all to the time characteristics. The duration T of the synchrotron radiation pulse is at least two orders of magnitude shorter than the time interval T_1

between the pulses. Therefore, using isomeric states having the lifetime τ_0 for which the condition

$$T \ll \tau_0 < T_1 \tag{1.1}$$

is fulfilled, we have the possibility of separating in time the pulses of the potentially scattered radiation (actually with the same duration T) from the resonance Mössbauer radiation travelling with a time delay of about τ_0.

Another important feature of the synchrotron radiation is a small angular divergence in a plane normal to the electron orbit which, as a rule, does not exceed a few tens of seconds of arc. This enables us to use efficiently the Bragg resonance scattering by the single crystal containing the appropriate isotope with the isomeric level. As has been shown (Afanas'ev and Kagan 1965a, Kagan *et al* 1968, Kagan and Afanas'ev 1973) the coherence is completely preserved in the resonance scattering by the system of nuclei with long-lived excited levels and the suppression effect of nuclear reaction inelastic channels (conversion) arises in the single crystal. In particular, a complete reflection (Kagan *et al* 1968) can be realised in resonance scattering even by a very strongly absorbing crystal within the range of angles of tens of seconds or seconds (depending on the values of the medium nucleus parameters). The existence of only one polarisation of the synchrotron radiation makes easier the choice of appropriate experimental conditions.

From the purely experimental point of view, the time delay of radiation within the resonance range of frequencies can be effectively used only if there is a simultaneous sharp decrease in the total number of scattered quanta reaching the detector. Use of the Bragg diffraction reflection by the magnetically ordered single crystal in which a purely nuclear resonance scattering is allowed and electron scattering is forbidden seems to be the most expedient in this case (Belyakov and Aivazyan 1968, Smirnov *et al* 1969, Stepanov *et al* 1974). Note that this version is to be used in the joint experiment of Kurchatov Institute of Atomic Energy and Novosibirsk Institute of Nuclear Physics, which is under preparation now (Artem'ev *et al* 1978). The theoretical analysis of the problem of direct excitation of isomeric nuclear states by synchrotron radiation must include at least the following questions:

(i) Investigation of the time dependence of the Bragg-reflected radiation.

(ii) Determination of the optimal conditions for maintaining intensity within the resonance range of frequencies and simultaneously suppressing the far wings of the frequency distribution.

(iii) Study of the time and frequency dependences of the intensity of the radiation passing through the Mössbauer absorber after the reflector.

(iv) Analysis of the possibility of forming scattered radiation of a definite frequency dependence in the resonance range of frequencies.

(v) Study of the possibility of direct detection of collective excitations in the nuclear subsystem of the crystal, in particular, the nuclear exciton.

All these questions are discussed in the present paper. It will be assumed that the standard procedure of preliminary scattering by ordinary single crystals narrows the energy distribution of the synchrotron radiation near the resonance frequency to about 1 eV (Artem'ev *et al* 1978) and this radiation by the basic crystal undergoes the purely nuclear Bragg diffraction.

We shall also assume that the hyperfine structure (HFS) of the resonance transition is well resolved and the measurement procedure suggests integration over a time interval greatly exceeding the inverse of the distance between the HFS components. In this case the time beats due to scattering interference in different HFS components, which are of no importance for us, can be neglected and considered as an independent resonance scattering corresponding to the individual HFS components.

2. Reflected intensity

Let radiation with a frequency $\tilde{\omega}$ fall on the crystal for a time T at a definite angle. The amplitude of the electric field can then be written as

$$E_{\tilde{\omega}}(t) = \mathscr{E}_{\tilde{\omega}} e^{-i\tilde{\omega}t} \psi(t) \tag{2.1}$$

where $\psi(t)$ is a function representing the time form of the radiation pulse, which we assume to have the form of a rectangle of unit height and length T.

Introduce the amplitude $R(\omega)$ of crystal reflection of the monochromatic wave having a frequency ω. Then

$$E'(\omega) = R(\omega)E(\omega) \tag{2.2}$$

where the primed symbol indicates the amplitude of the scattered wavefield. Then for the time dependence of the reflected wavefield we find

$$\begin{aligned} E'_{\tilde{\omega}}(t) &= \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} E_{\tilde{\omega}}(\omega) R(\omega) e^{-i\omega t} \\ &= \mathscr{E}_{\tilde{\omega}} \int_{-\infty}^{\infty} dt' \, G(t - t') e^{-i\tilde{\omega}t'} \psi(t'). \end{aligned} \tag{2.3}$$

Here

$$G(t) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} R(\omega) e^{-i\omega t}. \tag{2.4}$$

The reflection amplitude $R(\omega)$ being extended to the complex plane, ω is an analytical function in the upper half-plane. As a consequence $G(t)$ is different from zero only for $t > 0$ and actually is the delay function of the crystal response to the instantaneous pulse.

Suppose the quanta are incident on the crystal within the frequency range $\Delta\tilde{\omega}$ near the resonance frequency ω_0 corresponding to isomeric level energy and

$$\Delta\tilde{\omega} T \gg 1. \tag{2.5}$$

Proceeding from the amplitude (2.3) to the intensity and summing over the whole frequency range of incident radiation $\Delta\tilde{\omega}$ we find for the intensity of the reflected radiation, taking into account (2.5),

$$I_\alpha(t) = 2\pi \frac{N_\alpha}{\Delta\tilde{\omega}} \frac{1}{T} \int_0^T dt' |G_\alpha(t - t')|^2. \tag{2.6}$$

Here we have introduced explicitly the subscript α characterising the angle of radiation incidence on the crystal. Keeping in mind that only the case of Bragg diffraction will be considered below, α must be understood as the deviation from the angle satisfying the Bragg condition in the scattering plane:

$$\alpha = \boldsymbol{K}(\boldsymbol{K} + 2\boldsymbol{\kappa})/\kappa^2 \tag{2.7}$$

where $\boldsymbol{\kappa}$ is the wavevector of incident photons, and $\boldsymbol{K}$ is the reciprocal lattice vector corresponding to the chosen reflection.

Let us assume that the geometry is chosen so that the distribution of incident quanta with respect to α is determined by a comparatively small vertical divergence of the synchrotron radiation, with the scattering depending very weakly on the angle in the plane of the electron orbit. As a result R and G depend practically only on α. The quantity N_α in (2.6) is defined so that

$$\int N_\alpha \, \mathrm{d}\alpha = N_0 \tag{2.8}$$

where N_0 is the total number of quanta incident on the crystal per pulse of synchrotron radiation.

Equation (2.6) represents the differential time distribution of the reflected radiation. For $t \gg T$ we have roughly instead of (2.6):

$$I_\alpha(t) \approx 2\pi \frac{N_\alpha}{\Delta\tilde{\omega}} |G_\alpha(t)|^2. \tag{2.9}$$

3. Calculation of the response function

As follows from the above relations, it is sufficient to know the response function $G(t)$ for the analysis of the time dependence of the scattering. In a number of cases this function can be found in an analytical form.

3.1. Passage through the crystal

Suppose the radiation passes through the crystal of thickness l at an arbitrary angle (far from the angle meeting the Bragg condition). Let us consider the frequency range in the vicinity of a separate resonance transition having frequency ω_0 and width Γ. Then for the radiation at the crystal outlet we have (see, for example Afanas'ev and Kagan 1965a, Kagan *et al* 1968):

$$R^{\mathrm{A}}(\omega) = \exp(\mathrm{i}\kappa g_{00} l / 2\gamma_0), \tag{3.1}$$

$$g_{00} = \chi_0 - \frac{g_0}{v + i}, \qquad v = \frac{2\hbar(\omega - \omega_0)}{\Gamma}, \qquad \gamma_0 = \cos\theta_0. \tag{3.2}$$

Here θ_0 is the angle between $\boldsymbol{\kappa}$ and the vector of the inner normal to the entrance surface of the crystal, g_0 is a dimensionless quantity, related to the total cross section σ_{res} of the resonance nuclear absorption for $\omega = \omega_0$ by the rigidly fixed nucleus by the relationship:

$$g_0 = (1/\kappa)\sigma_{\mathrm{res}} n' \exp\{-Z(\boldsymbol{\kappa})\} \tag{3.3}$$

where $\exp\{-Z(\boldsymbol{\kappa})\}$ is the probability of the Mössbauer effect, $\boldsymbol{\kappa}$ is the wavevector of the incident radiation, n' is the density of the nuclei with the transition considered. The value χ_0 is coupled with the amplitude f_{e} of elastic scattering by electrons corresponding to one cell with the volume Ω_0 by the relationship

$$\chi_0 = (4\pi/\kappa^2\Omega_0) f_{\mathrm{e}}. \tag{3.4}$$

Substituting (3.1) into (2.4) and separating the resonance part, we obtain:

$$R^{\mathrm{r}}(\omega) = R^{\mathrm{A}}(\omega) - R^{\mathrm{A}}(\infty). \tag{3.5}$$

Then the integration of $R^A(\infty)$ gives the time δ-function and corresponds to the instantaneous reflection of the non-resonance part of the incident pulse.

Considering the integral of $R^r(\omega)$ we proceed to the complex plane ω and close the integration contour with a semicircle of an infinite radius in the lower half-plane, where $R^r(\infty) \to 0$.

Replacing the variables

$$z = \omega - \omega_0 + i/2\tau_0$$

which is equivalent to displacing the horizontal part of the contour by $i/2\tau_0$ to the lower half-plane (rounding the point $z = 0$ from above) we have:

$$\int_{-\infty}^{\infty} \frac{d\omega}{2\pi} R^r(\omega) e^{-i\omega t} = \exp(-i\omega_0 t + i\varphi - t/2\tau_0) \int_C \frac{dz}{2\pi} e^{-izt}[\exp(-i\xi/\tau_0 z) - 1] \tag{3.6}$$

where

$$\varphi = \chi_0 \kappa l/2\gamma_0, \qquad \xi = g_0 \kappa l/4\gamma_0, \qquad \tau_0 = \hbar/\Gamma. \tag{3.7}$$

The integral in (3.6) is determined by the singularity of the integrand at the point $z = 0$. Expanding the exponent within the square brackets in powers of $1/z$ and calculating each term of the series by means of the residue theorem we obtain a power series equivalent to the Bessel function J_1 (cf Lynch *et al* 1960). The final expression of the response function $G^A(t)$ is

$$G^A(t) = e^{i\varphi}\left(\delta(t) - \exp(-i\omega_0 t - \tau/2)\frac{\xi}{\tau_0}\frac{J_1(2\sqrt{(\xi\tau)})}{\sqrt{(\xi\tau)}}\theta(t)\right) \tag{3.8}$$

where

$$\theta(t) = \begin{cases} 1 & t > 0 \\ 0 & t < 0 \end{cases}, \qquad \tau = \frac{t}{\tau_0}.$$

As follows from (3.8), in the thin crystal for which the condition $\xi \ll 1$ is valid, the relation $J_1(2\sqrt{(\xi\tau)})/\sqrt{(\xi\tau)}$ is close to unity when $0 < t \lesssim \tau_0$, and the time dependence of the delayed radiation is equivalent to the decay of the isomeric state with lifetime τ_0. In the case of the thick crystal when the condition $\xi\tau \gg 1$ is met we have

$$G^A(t) \approx \exp(i\varphi - i\omega_0 t - \tau/2)\frac{1}{\sqrt{\pi}}\frac{\xi^{1/4}}{\tau_0 \tau^{3/4}}\cos(2\sqrt{(\xi\tau)} - \tfrac{3}{4}\pi). \tag{3.9}$$

The increase of the response amplitude with increasing crystal thickness is connected with the role of the frequency wings of the resonance in the thick crystal case. This is also the cause of the decrease in the effective delay time. Note that it is assumed implicitly that the interval $\Delta\tilde{\omega}$ is large enough so that

$$\xi \ll \Delta\tilde{\omega}\tau_0. \tag{3.10}$$

(Here it is reasonable to note that the expression δ $(t = 0)$ appearing in calculating the intensity should be understood as $\Delta\tilde{\omega}/2\pi$.)

On the contrary, the total number of quanta accounted for by the energy range $\sim\Gamma$ decreases because of a strong absorption.

3.2. Bragg reflection

Let us consider a magnetically ordered single crystal with a structure permitting the purely nuclear Bragg reflection (the electron scattering amplitude is equal to zero). Assume again that HFS of the resonance line is well resolved and the scattering in each individual transition can be considered independently (see Introduction).

Suppose the radiation is incident on the crystal at an angle close to such a reflection. Generally speaking the field arising inside the crystal has a complicated structure which is a superposition of four waves (Kagan and Afanas'ev 1973). In most cases, however, this superposition is divided into two independent subsystems for two-wave polarisation (for more details see Kagan and Afanas'ev 1973). Then the reflection amplitude corresponding to the individual HFS component and definite polarisation can be represented as follows:

$$R_\alpha^{\mathrm{B}}(\omega) = g_{10}\frac{1 - \exp(\mathrm{i}z_{21}l)}{(2\epsilon_0^{(1)} - g_{00}) - (2\epsilon_0^{(2)} - g_{00})\exp(\mathrm{i}z_{21}l)}, \tag{3.11}$$

$$\epsilon_0^{(1,2)} = \tfrac{1}{4}\{\alpha + g_{00} - g_{11} \pm [(\alpha - g_{00} - g_{11})^2 - 4g_{01}g_{10}]^{1/2}\}, \tag{3.12}$$

$$z_{21} = \kappa(\epsilon_0^{(2)} - \epsilon_0^{(1)})/\gamma_0.$$

Here g_{00} and g_{11} have the same form as (3.2). In the second case g_0 is replaced by g_1 which is determined by (3.3) with $\boldsymbol{\kappa} \to \boldsymbol{\kappa}_1$, where $\boldsymbol{\kappa}_1 = \boldsymbol{\kappa} + \boldsymbol{K}$ is the wavevector of the reflected radiation,

$$g_{10} = -\frac{\tilde{g}}{v + \mathrm{i}}, \qquad g_{01} = -\frac{\tilde{g}^*}{v + \mathrm{i}} \tag{3.13}$$

and the value of $\tilde{g}$ is complex in the general case.

In the case of the thick crystal when the condition $\mathrm{Im}\, z_{21}l \gg 1$ is satisfied, expression (3.11) is simplified. Making use of the explicit expressions for ϵ_0 and $g_{\alpha\beta}$ we have after simple transformations:

$$R_\alpha^{\mathrm{B}}(\omega) = -\frac{\tilde{g}}{g}\frac{\eta}{\tau_0}\frac{1}{\omega - z_0 + [(\omega - z_0)^2 - (\eta/\tau_0)^2]^{1/2}} \tag{3.14}$$

where

$$\eta = \frac{g}{\alpha - 2\chi_0}, \qquad z_0 = \omega_0 - \frac{1}{\tau_0}\left(\frac{\mathrm{i}}{2} + \frac{\eta}{p}\right), \tag{3.15}$$

$$p = \frac{2g}{g_0 + g_1}, \qquad g = |\tilde{g}|.$$

Note that $p \leqslant 1$ with $p = 1$ corresponding to the condition of the complete suppression effect.

Substitute (3.14) into (2.4) and again close the integration contour with a semicircle of infinite radius in the lower half-plane, with the integral along the semicircle being equal to zero (contour C_0). Transforming the expression (3.14) to the form with the square root in the numerator we have

$$G^{\mathrm{B}}(t) = \frac{\tilde{g}}{g}\frac{\tau_0}{\eta}\int_{\mathrm{C}_0}\frac{\mathrm{d}\omega}{2\pi}[(\omega - z_1)(\omega - z_2)]^{1/2}\,\mathrm{e}^{-\mathrm{i}\omega t} \tag{3.16}$$

where $z_{1,2} = z_0 \mp \eta/\tau_0$ (the integral along the closed contour of the expression containing no square root is equal to zero). The analytical continuation of the integrand suggests the existence of a cut along the segment $[z_1, z_2]$, so that we can reduce the integration contour to that in figure 1. The integrals over small circles around z_1, and z_2 make zero contribution while the integrand in (3.16) on both sides of the cut has the opposite sign. As a result the integral in (3.16) is reduced to the tabulated one (see, for

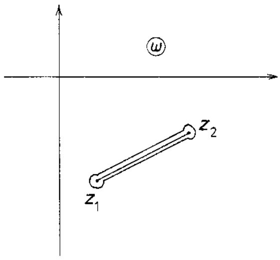

Figure 1. The contour in a complex plane ω along which the integral in the formula (3.16) is carried.

example, Gradshtein and Ryzhik 1971) and we have finally

$$G_\alpha^{\mathrm{B}}(t) = \mathrm{i}\frac{\tilde{g}}{g}\frac{\eta}{\tau_0}\mathrm{e}^{-\mathrm{i}z_0 t}\int_{-1}^{1}\frac{\mathrm{d}x}{\pi}\sqrt{(1-x^2)}\exp(-\mathrm{i}x\eta\tau)$$
$$= \frac{\mathrm{i}}{\tau_0}\frac{\tilde{g}}{g}\exp(-\mathrm{i}\omega_0 t - \tau/2)\frac{J_1(\eta\tau)}{\tau}\exp\left(\mathrm{i}\frac{\eta\tau}{p}\right)\theta(t). \qquad (3.17)$$

Here J_1 is again the Bessel function and τ is the dimensionless time defined in (3.8).

4. Time dependence of the Bragg reflection intensity

4.1. Fixed incident beam

The expression for $G^{\mathrm{B}}(t)$ obtained in the previous section allows us to determine the time evolution of the radiation delayed in the Bragg reflection. Considering the time t long compared with T we can use the expression (2.9). Then, according to (3.17), we have:

$$I_\alpha^{\mathrm{B}}(t) = \frac{2\pi}{\tau_0}\frac{N_\alpha}{(\Delta\tilde{\omega}\tau_0)}\mathrm{e}^{-\tau}\left|\frac{J_1(\eta\tau)}{\tau}\right|^2\exp\left(-\frac{2\eta''\tau}{p}\right). \qquad (4.1)$$

Here and below $\eta = \eta' + \mathrm{i}\eta''$ and similarly for other complex values.

It is seen from this expression that two new characteristic times appear in the problem, which satisfy the conditions

$$|\eta|\tau_1 = 1, \qquad \eta''\tau_2 = 1.$$

Taking into account (3.15) we have

$$\tau_1 = \frac{t_1}{\tau_0} = \sqrt{(y^2 + y_0^2)}, \qquad \tau_2 = \frac{t_2}{\tau_0} = \frac{y^2 + y_0^2}{y_0} \qquad (4.2)$$

where

$$y = (\alpha - 2\chi_0')/g, \qquad y_0 = 2\chi_0''/g. \qquad (4.3)$$

Usually the relation $y_0 \ll 1$ is valid for the crystal enriched in a resonance isotope. In this case at finite y the values of t_1 and t_2 ($> t_1$) are found to be very different except for a very narrow angle range $|y| \lesssim 2y$.

For $t < t_1$ the time dependence of the intensity of the delayed radiation in reflection remains close to the ordinary exponential law with the lifetime of the excited nucleus. However, when $t > t_1$ the time dependence changes drastically and we have the following expression for intensity within the range $t_2 > t > t_1$

$$I_\alpha^{\mathrm{B}}(t) \approx \frac{4}{\tau_0} \frac{N_\alpha}{(\Delta\tilde{\omega}\tau_0)} |y| \frac{\mathrm{e}^{-\tau}}{\tau^3} \cos^2\left(\frac{\tau}{|y|} - \frac{3\pi}{4}\right). \tag{4.4}$$

The principal change in the decay law is connected with the collective nature of absorption of each photon by the system of resonance nuclei in the reflector, with the nuclear exciton formed in the intermediate state. Such a collective excited state, as has been noted in the analysis of the nature of the suppression effect of nuclear reaction in the crystal, should decay through the elastic channel, i.e. with γ-quantum emission, essentially faster than in the case of the isolated nucleus. In this sense the result (4.4) is very instructive.

If $|y| \lesssim 1$, i.e. it lies in the angle range which is the most characteristic of the Bragg reflection, then $\tau_2 \gg \tau_0$ and the time dependence (4.4) remains practically valid for all times $t > t_1$. Absorption by electrons is of no importance since reflection goes efficiently on a small thickness of the crystal, determined by the characteristic length of the resonance scattering of γ-quanta by nuclei.

With decreasing $|y|$ the value of t_1 is continuously decreasing. Due to this the major part of the reflected intensity corresponds to short times. It is seen from (4.4) that the scale of the corresponding characteristic time is determined by the same value t_1. For $y_0 \ll |y| \ll 1$ the time-integrated flux of reflected quanta is:

$$Q_\alpha^{\mathrm{B}} \approx \frac{N_\alpha}{\Delta\tilde{\omega}\tau_0} \frac{1}{|y|}. \tag{4.5}$$

The number of quanta in the incident radiation corresponding to the resonance range of the energy Γ is close to $N_\alpha/(\Delta\tilde{\omega}\tau_0)$. Increase in Q_α^{B} with decreasing $|y|$ is due to the increasing role of the scattering in comparatively distant energy wings of the resonance. In this case the decrease in the scattering amplitude when deviating from the resonance is compensated to a considerable extent by an increase in the thickness of the crystal in which the effective reflection occurs. The increase in the relative contribution of reflection in the wings is accompanied by a decrease in the characteristic time of quanta delay in the scattering crystal for an essential part of the reflected intensity; this time becomes shorter as $|y|$ is smaller.

With decreasing $|y|$, not only t_1, but also t_2 decreases, and both parameters become closer to each other. When $|y| < \sqrt{y_0}$, the parameter $t_2 < \tau_0$ and knowledge of the time dependence for the reflected intensity for $t > t_2$ becomes of interest. In this time region the imaginary part η can not be neglected in the Bessel function argument in (4.1), and we obtain an asymptote other than (4.4) for the reflection intensity:

$$I_\alpha^{\mathrm{B}}(t) \approx \frac{1}{\tau_0} \frac{N_\alpha}{(\Delta\tilde{\omega}\tau_0)} (y^2 + y_0^2)^{1/2} \frac{\mathrm{e}^{-\tau}}{\tau^3} \exp\left[-2\left(\frac{1}{p} - 1\right)\frac{\tau}{\tau_2}\right]. \tag{4.6}$$

Comparing (4.6) and (4.4) one can draw the conclusion that when $p = 1$, i.e. in realisation of the complete suppression effect, both asymptotes are close and the picture given by (4.4) actually covers the whole time range $t > t_1$. In the absence of the suppression effect, when $p < 1$, the reflected intensity decreases sharply for $t > t_2$, which is due to an additional absorption by electrons in the crystal in the scattering. This circumstance appears to be very important when $|y|$ tends to zero ($|y| < y_0$). In this case τ_1 and τ_2 are close and (4.6) becomes valid for all $\tau > \tau_1$.

Let us consider now the case of strict satisfaction of the Bragg condition $y = 0$. Then $|\eta| = \eta'' = 1/y_0$ and the transition from (4.6) to the integrated intensity at $p = 1$ gives (4.5) with y replaced by y_0. The major part of the intensity is now concentrated in a very narrow time interval

$$\tau \leqslant \tau_{1\min} = y_0 = 2\chi_0''/g. \tag{4.7}$$

If we make χ_0'' formally tend to zero, then the dependence of I^B on t becomes of δ-functional character. Indeed, at any finite t, (4.6) or (4.1) vanishes and when $t = 0$ (4.1) becomes ∞. Thus, *if the Bragg condition is accurately fulfilled and the electron absorption is neglected, the delay effect is completely lacking and instantaneous reflection of the incident pulse occurs.* This is because the decrease in the amplitude of scattering by an individual nucleus deviating from the resonance for $y = 0$ is fully compensated by the increasing thickness of the layer where reflection occurs and, as a consequence, $R(\omega)$ in the thick crystal does not depend on frequency (see (3.14)). This is found to occur both in the presence of the suppression effect ($R = 1$) and for $p < 1$ ($R < 1$, cf. Kagan *et al* 1968).

Note that when $y = \chi_0'' = 0$ divergence also occurs in the time-integrated intensity. This divergence, however, is of fictitious character and can be easily eliminated if one takes into account the finiteness of the frequency range $\Delta\tilde{\omega}$ of the incident radiation. At the same time in accordance with the condition $R =$ constant the whole spectrum of incident radiation is uniformly reflected. The resonance region is not chosen and practically all radiation comes to the detector.

Finally, when considering large $|y|$ we have $\tau_1 \gg 1$ and for the entire region of

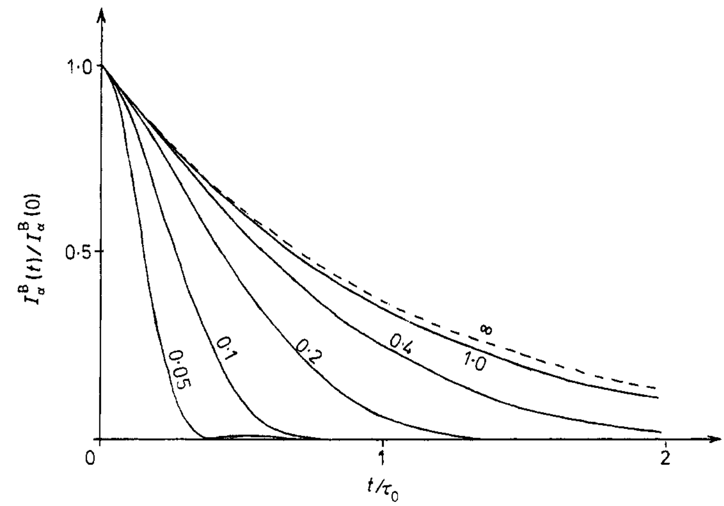

Figure 2. The time dependence of the intensity of the radiation Bragg-reflected from the thick crystal with the dependence on the deviation parameter y from the Bragg condition. The values of y are stated next to curves.

interest $\tau < \tau_1$ we find from (4.1),

$$I_\alpha^B(t) = \frac{\pi}{2\tau_0} \frac{N_\alpha}{(\Delta\tilde{\omega}\tau_0)} \frac{1}{y^2} e^{-\tau}. \tag{4.8}$$

The time delay is of classical character since the collective effects play a minor role in excitation of the resonance level in this region.

The described picture of change in the time dependence of the reflected intensity depending on the deflection angle y from the Bragg direction is readily seen in figure 2. A sharp reduction of the time delay occurs already for $|y| < 0{\cdot}3$. For $|y| \gtrsim 1$, at least for $t < 2\tau_0$, the time dependence begins to follow the time dependence of decay of an isolated excited nucleus (broken curve).

4.2. Divergent incident beam

If, nevertheless, the specifically collimated synchrotron radiation has an angular divergence $\Delta\alpha$ large compared with g (as it seems actually to be), then in the absence of an additional collimation we have to average (4.1) to obtain the actual time dependence of the reflection intensity. The same averaging also arises effectively in the case of a mosaic crystal if the crystallites are appropriately aligned only in the plane of the crystal specimen remaining perfect in thickness.

We shall confine ourselves to considering times $\tau > \tau_{1\min}$ (see (4.7)). Then we can neglect the imaginary part η in integration of (4.1) over α. As a result we find

$$\overline{I^B(t)} = g \int_{-\Delta y/2}^{\Delta y/2} I_\alpha^B(t) = \frac{4\pi}{\tau_0} \frac{N_{res}}{\Delta y} \frac{e^{-\tau}}{\tau} \left(\frac{4}{3\pi} - \int_0^{x_0} dx \frac{J_1^2(x)}{x^2} \right). \tag{4.9}$$

Here

$$\Delta y = \frac{\Delta\alpha}{g} \qquad N_{res} = \frac{\int N_\alpha \, d\alpha}{(\Delta\tilde{\omega}\tau_0)} \tag{4.10}$$

is the number of incident quanta corresponding to the resonance energy range $x_0 = 2\tau/\Delta y$. If $\Delta y \gg 1$, then the second term within the round brackets may be neglected.

It follows from (4.9) that in the case of a wide beam the effective time of delay in the reflector is also appreciably reduced compared with the lifetime of the excited nuclear state. A very peculiar law of decay was found, with increase in the intensity at small τ as $1/\tau$. Acceleration of the decay is again due to the collective effects in excitation of the resonance level by quanta incident within the angle range $|y| < 1$. The radiation incident at angles $|y| > 1$ is reflected very weakly. The effect of the resonance wings is here essentially weakened since the range of angles of the effective reflection is narrowed when deviating from the resonance. However, the logarithmic rise of the time-integrated intensity still remains as t decreases (but $t > t_{1\min}$) which is a consequence of the law

$$|R(\omega)|^2 \sim 1/|\omega - \omega_0|, \qquad |\omega - \omega_0| \gg 1/2\tau_0$$

found in Kagan *et al* (1968) for the case of the Bragg reflection of the wide beam.

The appearance of the factor $1/\Delta y$ in (4.9), which actually can decrease the intensity of the delayed radiation very sharply, is also indicative of non-effectiveness in the reflection of angles $|y| > 1$.

5. Spectral properties of the Bragg-reflected radiation

Now we proceed to the analysis of the spectral properties of the delayed pulse accompanying the Bragg resonance reflection by the nuclei in the crystal. As follows from the results discussed in the previous section, the collective nature of excitation of the system of nuclei in the reflector makes the intermediate state decay more rapidly and decreases the delay time, which should result in widening of the frequency distribution compared with the case of decay of the isolated excited nuclei. Using this radiation in experiments of the Mössbauer type we are faced with an extremely serious problem of loss of resolution. Therefore the question of the type of spectral distribution, the possibility of its formation and attainment of resolution of the scale of the excited level width of the individual nucleus is of great importance for the whole problem considered.

To analyse the problem let us consider directly the case when the synchrotron radiation pulse subjected to the Bragg nuclear reflection by the first crystal then passes through the crystal absorber containing the same nuclei but with a shift of $\Delta\omega_0$ in the resonance frequency. The time dependence of the intensity of the radiation passed through the absorber will be determined by (2.4) and (2.9) with the function R which is the product of (3.1) and (3.11):

$$G_\alpha^{AB}(t, \Delta\omega_0) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} R^{A}_{\Delta\omega_0}(\omega) R^{B}_{\alpha}(\omega) e^{-i\omega t}. \tag{5.1}$$

Here and below we count ω off the resonance frequency value of the reflector. (The formula (5.1) suggests that all coherently scattered radiation passes through the absorber.)

Proceeding from the reflection amplitudes to the response functions we have

$$G_\alpha^{AB}(t, \Delta\omega_0) = \int dt'\, G^{A}_{\Delta\omega_0}(t - t') G^{B}_{\alpha}(t'). \tag{5.2}$$

Let us consider for simplicity the case of a thin absorber and an infinitely thick reflector. Then, using (3.8) within the limit $\xi\tau_0 \ll 1$ and (3.18) we have from (5.2)

$$G_\alpha^{AB}(t) = [G_\alpha^{B}(t) - \xi\phi_\alpha(t)] e^{i\varphi} \tag{5.3}$$

where

$$\phi_\alpha(t) = \exp(-i\Delta\omega_0 t) \frac{i}{\tau_0} \frac{\tilde{g}}{g} \exp(-\tau/2) \int_0^{\tau} d\tau' \frac{J_1(\eta\tau')}{\tau'} \exp[i(\Delta\omega_0\tau_0 + \eta/p)\tau']. \tag{5.4}$$

Consider the case of a small deviation from the Bragg condition but when the electron absorption can be still neglected $y_0 \ll |y| \ll 1$. Then the integral in (5.4) can be reduced to

$$\int_0^z dz' \frac{J_1(z')}{z'} e^{i\beta z'}$$

where

$$\beta = [1 + \Delta\omega_0\tau_0 y p]/p, \qquad z = \tau/y. \tag{5.5}$$

In the analysis of the times t comparable with τ_0 the parameter $z \gg 1$ and replacing the upper limit of integration by ∞ we find (Gradshtein and Ryzhik 1971):

$$\phi_\alpha(t) = -\frac{\tilde{g}}{g} \frac{1}{\tau_0} \frac{1}{\beta + \sqrt{(\beta^2 - 1)}} \exp(-\tau/2) \exp(-i\Delta\omega_0 t). \tag{5.6}$$

It should be noted that the problem for the case of an absorber of arbitrary thickness is also solved directly within this y limit. Then

$$\phi_\alpha(t) = -\frac{\tilde{g}}{g}\frac{1}{\tau_0}\frac{1}{\beta + \sqrt{(\beta^2 - 1)}}\exp(-\tau/2)\frac{J_1(2\sqrt{(\xi\tau)})}{\sqrt{(\xi\tau)}}\exp(-i\Delta\omega_0 t). \quad (5.6a)$$

It follows from (5.5) and (5.6) that within a comparably wide range

$$|\Delta\omega_0\tau| < 1/|yp| \quad (5.7)$$

and dependence on $\Delta\omega_0$ appears to be very weak, i.e. the resonance structure is actually lacking in the reflected pulse. In addition ϕ_α depends very weakly on y and for $y \to 0$ remains finite.

If we compare this result with the behaviour of $G_\alpha^B(t)$ in (3.17) then we can easily see that when

$$\tau = t/\tau_0 \gg y^{1/3} \quad (5.8)$$

the relationship take place:

$$|\phi_\alpha(t)| \gg |G_\alpha^B(t)|. \quad (5.9)$$

Thus, at the times considered and for $\xi \sim 1$ *the intensity is really connected only with the radiation delayed in the absorber*; that is very important for many possible experiments (see § 6). It should be pointed out that an anomalous increase in the reflection intensity, including the integrated one (see (4.5)) at small y does not practically affect the scale ϕ_α, i.e. the major part of the radiation passes through the absorber almost without interaction.

When $|y| \gg 1$

$$\phi_\alpha(t) = \frac{1}{4\tau_0}\frac{\tilde{g}}{g}e^{-\tau/2}\left(\frac{1 - \exp(-i\beta\tau/y)}{\beta}\right)\exp(i\tau/py). \quad (5.10)$$

Substituting (5.10) into (5.3) and proceeding to the intensity we find

$$I_\alpha^{AB}(t) \approx I_\alpha^B(t) - \xi\Delta I_\alpha(t), \quad (5.11)$$

$$\Delta I_\alpha(t) = \frac{\pi}{\tau_0}\frac{N_\alpha}{(\Delta\tilde{\omega}\tau_0)}\frac{1}{2y^2}e^{-\tau}\frac{\sin(\beta\tau/y)}{(\beta/y)}. \quad (5.12)$$

It follows from (5.5) and (5.12) that in this case with t comparable to τ_0, the absorber distinctly shows the resonance structure of the reflected radiation, which become continuously worse with decreasing t. However ΔI_α is small, proportional to $1/y^2$ (just as I_α—see (4.8)).

In the case when the divergence of the incident beam is sufficiently large or the crystal is noticeably mosaic, the expression (5.11) must be averaged over the angle range $\Delta y \gg 1$ near the Bragg value $y = 0$ (cf. the previous section). We shall again neglect the imaginary part of χ_0 in (3.15) and pass to the new variable $x = \tau/y$ in the integral over y. The integral of the first term in (5.11) is given by the formula (4.9) so that only the second term must be integrated. Rearranging the integrals over τ' and x and proceeding to the new variable $u = 1 - \tau'/\tau$ in the integral over τ' we have

$$\overline{\Delta I(t)} = g\int_{-\infty}^{\infty} dy\, \Delta I_\alpha(t) = \frac{8\pi}{\tau_0}\frac{N_{res}}{\Delta y}e^{-\tau}\int_0^1 du \cos(z\tau u)\chi(u) \quad (5.13)$$

where we introduced a new variable $z = \Delta\omega_0\tau_0$ while

$$\chi(u) = \int_0^\infty \mathrm{d}x \cos(xu/p)\frac{J_1(x)}{x}\frac{J_1[x(1-u)]}{x(1-u)}. \tag{5.14}$$

Using the integral representation of the Bessel function (cf. (3.18))

$$\frac{J_1(x)}{x} = \frac{1}{\pi}\int_{-1}^{1} \mathrm{d}t\sqrt{(1-t^2)}\exp(\pm itx)$$

we may transform (5.14) for $\chi(u)$ to a form more suitable for calculations: the expression

$$\chi(u) = \frac{1}{\pi}\int_{-1}^{1} \mathrm{d}x\sqrt{(1-x^2)}\sqrt{[1-\psi^2(u,x)]}\theta(1-\psi^2(u,x)) \tag{5.15}$$

where

$$\psi(u,x) = u/p + (1-u)x$$

and $\theta(t)$ is the step function (see (3.8)). Note that the function $\chi(u)$ is different from zero only for $u < u_0 = 2p/(1+p) \leqslant 1$ due to the existence of the theta function in (5.15).

The spectral structure of radiation is defined in the general case by two parameters $z = \Delta\omega_0\tau_0$ and $\tau = t/\tau_0$. As follows from (5.13), at small τ, the dependence on $\Delta\omega_0$ practically vanishes, i.e. at the times which correspond to the maximum intensity the resonance structure in the reflected radiation is weakly manifested. However it manifests itself distinctly with increasing τ. This is readily seen in figure 3 where the relative intensity

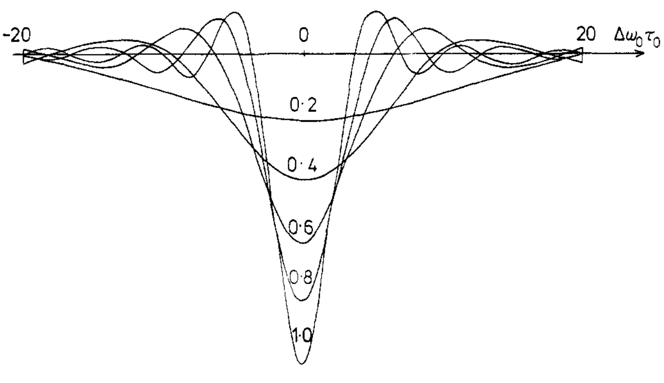

Figure 3. Relative intensity of radiation $\overline{\Delta I}/I^{\mathrm{B}}$ which is transmitted after reflection through the thin absorber with the resonance frequency displaced by $\Delta\omega_0$ in various time moments t after the pulse transmission. The values of t/τ_0 are stated next to curves. The angular divergence of radiation $\Delta y \gg 1, p = 1$.

of absorbed radiation $\overline{\Delta I}/I^{\mathrm{B}}$ is given as a function of z for various values of τ. As can be seen from this picture, the spectral distribution for $\tau < 0{\cdot}25$ still remains very wide, but for $\tau \gtrsim 0{\cdot}5$ the spectral line appears to be sufficiently narrowed, oscillating noticeably, however, in the tails.

The results obtained allow us to draw the conclusion that the reflected intensity, integrated over time excluding the finite interval $[0, t_1]$ just after the pulse of synchrotron radiation, must be used as the source of the resonance radiation. Considering again the same system—a thick reflector and thin absorber—we readily find:

$$\bar{Q}(\tau_1, z) = \int_{t_1}^{\infty} dt\, I(t) = \overline{Q^{B}}(\tau_1) - \xi\overline{\Delta Q}(\tau_1, z) = \frac{16}{3}\frac{N_{res}}{\Delta y}\left(-\mathrm{Ei}(-\tau_1) - \tfrac{3}{2}\pi\xi e^{-\tau_1}\right.$$
$$\left.\times \int_0^1 du \frac{\chi(u)}{1+(zu)^2}[\cos(z\tau_1 u) - zu\sin(z\tau_1 u)]\right) \tag{5.16}$$

where $\tau_1 = t_1/\tau_0$ and $\mathrm{Ei}(x)$ is the integral exponential function. Figure 4 shows the dependences of $\overline{\Delta Q}/\overline{Q^B}$ on $z = \Delta\omega_0\tau_0$ for various values of τ_1 and $p = 1$. It is seen that the weak frequency dependence of the intensity integral over time at small τ_1 is replaced by a striking resonance structure with increasing τ_1. Naturally with an increase in τ_1,

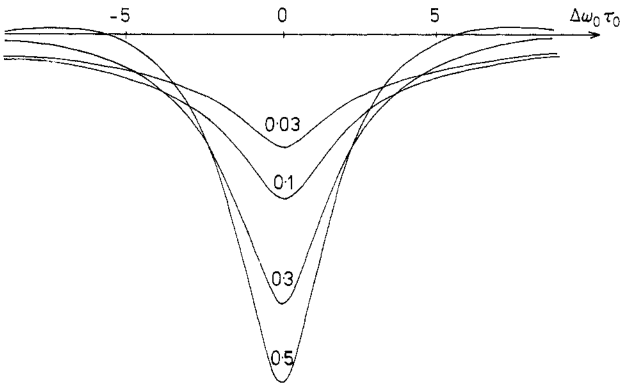

Figure 4. Integral over time of the relative intensity $\overline{\Delta Q}/\overline{Q^B}$ with the exception of the finite time interval $[0, t_1]$ for the system of a reflector and a thin absorber with the resonance frequency displaced by $\Delta\omega_0$. The values of t_1/τ_0 are stated next to curves. The angular divergence of radiation $\Delta y \gg 1$, $p = 1$.

the integral reflected intensity decreases. However, when $\tau_1 = 0{\cdot}5$, for example, the function $-\mathrm{Ei}(-\tau_1) \approx 0{\cdot}56$, and moreover, as follows from (5.16), the total number of reflected quanta exceeds the number of quanta in the synchrotron pulse corresponding to the resonance energy interval (with allowance for y) for this value of τ. Actually, the decrease in the intensity with increasing τ_1 is due, first of all, to 'illegal' quanta reflected in comparatively far resonance wings and is, therefore, inefficient for the resonance experiments.

In figure 5 the same curves are given but for $p = 0{\cdot}7$. It is seen that violation of the suppression effect in the reflector ($p < 1$) results in a sharp widening of the frequency distribution of reflected quanta. With further decrease in p the resonance structure is widened still more. Thus, it is evident that the optimum effect occurs close to the condition of full realisation of the suppression effect.

Comparison of figure 3 with figures 4 and 5 shows that intensity oscillations in the frequency tails, observed in the first case, are significantly averaged and noticeably

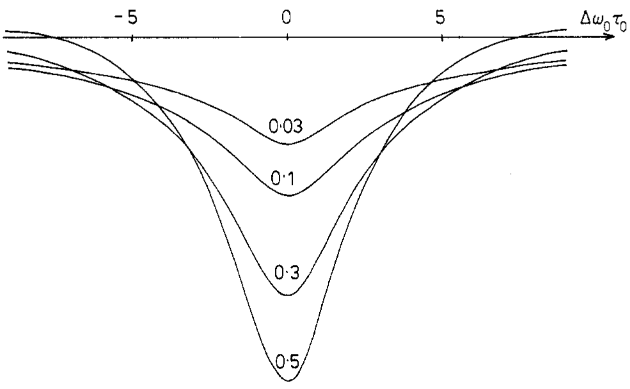

Figure 5. The same curves as in the figure 4 but for the case of incomplete suppression effect in the reflector, $p = 0{\cdot}7$.

suppressed, which, of course, improves the spectral properties. The fact of appearance of the frequency and time regions, where the intensity of radiation having passed through the absorber exceeds the incident one, results from the change of the spectral characteristics of the reflected radiation when passing through the absorber and is actually of the same nature as the known result described in Lynch *et al* (1960). Note that with increase in the thickness of the crystal-absorber the amplitude of the oscillations increase. Moreover there arise oscillations with the change of the thickness l. Use of the intensity integral over time, excluding the noticeable initial time interval $[0, t_1]$, become still more important for formation of the spectral structure.

Since we use the 'thin absorber' approximation in this section, it is necessary to make a note. The real parameter of expansion in (5.3) and (5.4) is $\xi\tau$ and not ξ. Therefore, with increase in τ the thickness range narrows in principle, when the absorber can be considered as thin. However, the existence of the exponential factor makes large values of τ inefficient and the condition $\xi \ll 1$ remains actually sufficient.

6. Possibility of detection of the nuclear exciton

As has been noted, the unusual character of the time dependence of the quanta delayed in the Bragg reflection from the crystal is accounted for by the character of decay of the collective nuclear excitation arising in the crystal (see §4). In this sense, finding the time dependence (4.6) or (4.9) would be indirect evidence of formation of such a state.

At the present time the possibility of direct detection of such a nuclear exciton in the Mössbauer-type experiments with breaking of the initial beam coming from the source is being intensively discussed. The use of synchrotron radiation opens up, in principle, interesting possibilities for the realisation of such an experiment.

Indeed, considering the two-crystal geometry described in the previous section and selecting a small angle range $|y| \ll 1$ near the Bragg value $y = 0$, we have the relationship (5.9) for the delay times (5.8). At times one can experimentally verify that the intensity of the pulse reflected by the first crystal is small compared with that due to decay in the second crystal, but this is just the condition (which is equivalent to breaking off the radiation) that is required for experiments which permit the behaviour of excited states to be observed.

Now measurement of the angular dependence of the gamma quanta emitted with the delay time (5.8) should show a sharp angular asymmetry when scattered forward as well as by the Bragg angle (when the crystal is appropriately oriented) if in absorption of the gamma quanta nuclear excitons are formed in the crystal (Afanas'ev and Kagan 1965b), or, otherwise, the uniform angular distribution.

In the time range where (5.9) is valid

$$I_\alpha(t) \approx 2\pi \frac{N_\alpha}{(\Delta\tilde{\omega}\tau_0)} \xi^2 \frac{1}{\tau_0} \frac{1}{|\beta + \sqrt{(\beta^2 - 1)}|^2} e^{-\tau}. \tag{6.1}$$

Note, first of all, that the delayed intensity (6.1) is found to be proportional to the square of the absorber thickness l^2 (see (3.7)). This is a typically coherent effect which is due to the collective nature of the nuclear excitation in the crystal. The most interesting circumstance is that *at any small ratio of the elastic width of the resonance level*, Γ_1, *to the inelastic conversion width* Γ_2 (in (3.7) $g_0 \sim \Gamma_1/(\Gamma_1 + \Gamma_2)$) condition $\xi \sim 1$ can be always reached by increasing l ((6.1) is valid only at $\xi \leqslant 1$), *the gamma decay intensity will be proportional to the total width* Γ *and not to* Γ_1, with the probability of the conversion decay remaining standard. Measuring (6.1) and the yield of the conversion electrons, one can find that the ratio of probabilities of gamma decay and conversion decay becomes comparable though $\Gamma_1 \ll \Gamma_2$. This effect, depending just on the character of the nuclear exciton decay, was predicted in Afanas'ev and Kagan (1965b) and Kagan and Afanas'ev (1966).

Thus, combination of the pulse nature of the synchrotron radiation with the time behaviour of the pulse delayed in the Bragg reflection permits us to solve, in principle, the problem of 'shutter' and investigate the appearance of the nuclear excitons in the crystal. The difficulty of this experiment is that a very high collimation and a sufficiently perfect reflecting crystal are required.

7. Conclusions

The results obtained in the previous sections answer actually all the questions stated in the Introduction and enable us to make some remarks about the experimental aspects of the problem.

Measurement of the anomalous time dependence for the delayed Bragg-reflected pulse, i.e. the law $(t/\tau_0)^{-3} \exp(-t/\tau_0)$ in small deviation from the Bragg angle or $(t/\tau_0) \exp(-t/\tau_0)$ in the case of a wide incident beam or mosaic crystal in the specimen plane (see § 4), seems to be most effective for the detection of the direct excitation of the isomeric nuclear states.

Also important is the circumstance that the delayed flash-light contains, due to the effect of the frequency wings, an essentially larger number of quanta than that corresponding to the resonance energy range N_{res} (or $N_{res}/\Delta y$ in the case of a wide beam, see (4.10)). These 'excess' quanta can not be used if, as in the case of the Mössbauer experiments, a distinctly manifested resonance structure in the spectrum incident to the specimen is required; on the contrary, they must be suppressed.

Formation of the effective resonance structure, with the number of the quanta corresponding to the resonance range saved, may be achieved, as it seems, in two ways. One way is time 'mask' excluding the noticeable time range and measurement of the intensity, integrated over the remaining part of the time, after transmission through the reflector–specimen system. As is seen from the results given in § 5, a significant part of the quanta from N_{res} and $N_{res}/\Delta y$ can be used. The other way is to reduce the thickness of the crystal intended for the Bragg reflection. This problem will be treated elsewhere.

Finally, it should be noted that the direct excitation of nuclei by the pulse synchrotron radiation enables to realise experiments on the detection and study of collective nuclear excitations in the crystal—nuclear excitons. To the results given in the previous section we would add that for the first time the possibility opens for studying the anomalous time delay of the gamma-quanta transmitted through the crystal under conditions close to realisation of the suppression effect.

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