

Theory of coherent phenomena and fundamentals in nuclear resonant scattering

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We discuss the general theory of coherent phenomena in nuclear resonant interaction of γ -quanta with crystals. The coherence is realized in collective excitation of the ensemble of nuclei (nuclear exciton) with the conservation of phase memory and in the transformation of a γ -quantum into a quasi-particle of Bloch type in a crystal. The collective character of excitations causes a change in the resonant nuclear parameters and in the lifetime of the excited state. This manifests itself in a speed-up of the decay in the forward direction in a thin crystal and, on the contrary, a strong reduction of elastic scattering in a thick crystal. The reconstruction of the wavefunction of an individual γ -quantum in scattering under Laue or Bragg conditions leads to the suppression effect of inelastic incoherent channels. This effect is discussed in detail. The analysis is based on a derived general system of equations describing the resonant diffraction of γ -quanta in a crystal with an arbitrary relation between the coherent and incoherent channels. This system is used to deduce the equations describing the time-dependent nuclear resonant scattering of synchrotron radiation. We discuss the most instructive experiments with revealing coherent phenomena.

Keywords: coherence, resonant diffraction, suppression effect, nuclear exciton, time-dependent NRS, synchrotron radiation

1. Introduction

The Mössbauer effect [1–3] emerged as a phenomenon inherent in an individual nucleus imbedded in a crystal. The ensemble of nuclei manifested itself simply as a set of independent resonant centers. The discovery of this effect led to the appearance of γ -quantum sources with an extremely sharp linewidth ($\sim 10^{-9}$ – 10^{-6} eV) and simultaneously of absorbers with the same sharpness in energy. Such parameters are characteristic for low-lying nuclear isomer states and they are displayed during the decay or excitation of a nucleus in a solid without emitting or absorbing phonons (“recoilless transition”). It is essential that this energy scale is much smaller than any other energetic parameter in a solid.

From the very beginning the unique small ratio of linewidth Γ to the energy E_γ of γ -rays attracted attention to the possibility of realizing coherent effects in the ensemble of Mössbauer nuclei.

At first sight it seems that collective coherent phenomena for γ -quanta with a quantum energy above 10 keV should play a minor role under these conditions.

A number of reasons could be responsible. First of all, it is well known that the resonant interaction of a γ -quantum with a nucleus occurs via the formation of an excited state. The typical magnitude for the lifetime τ_0 of the Mössbauer excited states is 10^{-5} – 10^{-9} s. This is enormously long compared with the intrinsic nuclear times. In addition, τ_0 is many orders of magnitude larger than the reciprocal Debye frequency ω_D^{-1} . This means that a nucleus experiences a great number of oscillations between excitation and decay. In addition, most of the Mössbauer nuclei decay via the inelastic (internal conversion) channel which dominates over the elastic one. It is rather natural to assume that the decay of the excited nucleus under these conditions should not correlate with the excitation process. In other words, loss of phase memory should take place, at least, to an essential extent. Other crucial questions arise if we consider the phase relation for different nuclei and its conservation in the process of scattering γ -quanta. The problem is especially puzzling if one takes into account that the wavelength of the γ -radiation is less than the interatomic distance and that a direct interaction between the nuclei is practically absent.

However, in spite of all such circumstances one can find, at least, necessary conditions for the preservation of the phase memory. First of all, it is necessary that the environment and nuclear subsystem itself revert exactly to the initial quantum state after an interaction event. In addition, an excited nucleus during its long lifetime should not be subjected to a chaotic external perturbation. Under these conditions it is very important that the absorption of a γ -quantum in a system of identical nuclei is connected not with the excitation of one or another nucleus but with the excitation of the system as a whole. In this case the coherence is realized both in the conservation of the phase memory at an individual nucleus and in the strong correlation of the phases between different nuclei. The excited state turns out to be delocalized and extended over the system. As a result, we arrive at the notion of a collective nuclear excited state or nuclear exciton. In the simple case the wavefunction of such a collective state can be represented in the form

$$\psi_{\mathbf{k}} = \frac{1}{\sqrt{N}} \sum \varphi_n e^{i\mathbf{k}\mathbf{r}_n}, \quad (1.1)$$

where \mathbf{k} is the wave vector of the incident quantum and φ_n is the wavefunction of the excited state of the n th nucleus. It was predicted that the character of the elastic decay and the lifetime of such a nuclear exciton can differ noticeably from the case of an isolated nucleus [4–6] (see also [7]). Since the resonant scattering by the nuclear system takes place via the formation of the collective excited state, the position and elastic width Γ_γ of the resonance level can change significantly. It is interesting that Γ_γ and τ^{-1} can increase as well as decrease (see [6]). The most instructive examples are as follows: a noticeable enhancement of the γ -decay probability Γ_γ of a nuclear exciton produced inside a thin slab [4,5] and, on the contrary, a reduction of the total width when a γ -quantum travels through a thick crystal due to the disappearance of the elastic width Γ_γ [6].

The previous remarks imply the purely coherent γ -decay of a collective excited state. If the ground state of the nuclei has spin $I_0 \neq 0$, the γ -decay can be accompanied by the return of one nucleus from the ensemble to the ground state with another spin projection with respect to the initial state. This is a typical incoherent scattering because the site remains tagged in the system after the event. (We have the same when the decay of the collective excited state occurs in the electronic conversion channel.) Another violation of coherence is connected with the excitation of the phonon subsystem in the process of γ -decay. Now the tag of the event is the change of the state of the environment. These incoherent channels reduce the coherent reconstruction of the nuclear parameters (see [8]) and the amplitude of coherent scattering. However, as long as the Mössbauer effect remains, this amplitude keeps a finite value.

The conservation of coherence and its significant role in the general picture of nuclear resonant scattering have already been revealed indirectly or directly soon after the discovery by Mössbauer. Black and Moon [9] observed the interference between nuclear resonant and electronic potential scatterings by an individual atom (see also [10]). Clearly, it is indirect evidence for the conservation of the long-term phase memory since the electronic Rayleigh scattering is actually prompt. On the other hand, Black and Duerdorth [11] have observed Bragg scattering by a crystal where the nuclear resonant scattering is dominant. This is unambiguous evidence for the strong correlation between the phases of individual nuclei in a relatively large bulk. An additional proof of the temporal coherence can be extracted from the experiments on the time-delayed transmission of γ -quanta through a thin foil with Mössbauer resonant nuclei [12,13].

Later, a remarkable experiment was performed to reveal the consequences of the violation of coherence [14]. For this purpose, the authors measured the energy distribution of Bragg-scattered γ -quanta. They proved that, in spite of an absolutely negligible difference in energy, the γ -quanta, which were related to returning a nucleus to other hyperfine (HF) sublevels of the ground state, did not experience Bragg scattering at all. This is a clear manifestation of destroying space coherence.

The most intriguing aspects of the coherent phenomena in this field are connected with the motion of γ -quanta in a thick crystal with strong resonant nuclear interaction. The problem in this case cannot be reduced to a single act of absorption or emission. Actually, the continuous transformation of the plane wave of an incident particle into a superposition of Bloch-type states takes place. Owing to the strong collective coherent scattering this occurs at a limited depth before absorption starts to play a serious role.

The further evolution of γ -quanta in this superposed state occurs at the background of the strong inelastic channels of conversion and the incoherent channels of scattering. The description of the γ -quantum motion in a thick crystal for such conditions, involving the specific features of the interaction with nuclei in the presence of HF structure, requires the development of a general dynamical theory of resonant diffraction. Such a general theory was developed in the work of Afanas'ev and Kagan [15–18] (see also reviews [7,19]). An alternative derivation of the dynamical equations was made in the work of Hannon and Trammell [20,21]. The central place

in [15–18] was the prediction and detailed analysis of the suppression effect for nuclear reactions in a crystal. The essence of the effect is associated with the fact that the amplitude of any process after the appearance of the Bloch state is a sum of amplitudes referring to each component of such a state. It is found that in Bragg and Laue diffraction such states are realized in which the total amplitude of the formation of an excited nucleus vanishes. As a result, the internal conversion channel and incoherent scattering with the formation of an excited nucleus are suppressed. Thus, the strongly absorbing crystal becomes transparent. It is remarkable that the temperature and the oscillations of atoms do not break the effect.

The theoretical predictions induced an extensive development of the experimental research. The suppression effect was discovered by Voitovetskii et al. [22] for Laue diffraction in a single crystal of metallic tin and by Smirnov et al. [23] in a single crystal of iron enriched in ^{57}Fe . A detailed study of the effect with a purely nuclear reflection in a single crystal of $^{57}\text{FeBO}_3$ was performed by van Bürck et al. [24]. The anomalous resonant nuclear Bragg diffraction [18], predicted within the framework of the dynamical theory (see also [21]), was observed for the first time in the work of van Bürck et al. [25]. References to numerous investigations performed before and after the above work can be found in reviews [26,27]. Note that principal ideas such as the acceleration of the forward γ -decay of the nuclear exciton and the suppression of the elastic scattering channel in a thick single crystal have obtained experimental evidence at this time [28,29].

A new stage of studying coherent phenomena by nuclear resonant scattering (NRS) in crystals started after realizing the direct excitation of the Mössbauer transitions by synchrotron radiation (SR). The fruitful idea was to measure the delayed emission after the pulsed SR irradiation of samples. The delayed emission, associated with the resonant excitation of nuclei, forms in fact a source of Mössbauer radiation (see also discussion in [30]). The unique merit of such a source is the possibility to measure the temporal picture of scattering. An attempt of observing retarded emission with purely nuclear reflection from a crystal was published by Chechin et al. [31]. The comprehensive experiment which became the starting point for the development of this field has been performed by Gerdau et al. [32]. The time-dependent dynamical theory of the nuclear resonant scattering of SR by crystals was developed in [33]. To a great extent it uses the results [15,17–19] obtained for the stationary case. The analysis of the coherent collective phenomena under nonstationary conditions led, in particular, to the prediction of the anomalous time behaviour for the Bragg diffraction changing drastically with the deviation from the exact Bragg angle and for the radiation transmitted through the crystal under resonant nuclear scattering. The experiments studying Bragg [34] and forward [35,36] (also [37]) scattering of the SR radiation have revealed all the qualitative features of the coherent temporal picture. The specific feature for the temporal evolution of the delayed emission is the manifestation of the interference between components related to scattering with the excitation of various HF transitions. The theory of the quantum beats originating from the interference was considered first by Trammell and Hannon [39] (see also [40]).

The quantum beats have been observed experimentally for the first time by Gerdau et al. [41]. In the wake of displaying the main coherent phenomena of the SR-crystal interaction a great number of papers, where various aspects of coherence are revealed, have been published. References to these papers can be found in reviews, e.g., [42,43].

2. Change of resonant nuclear parameters

In order to demonstrate how the collective character of excitations in the nuclear system leads to a variation of the resonant nuclear parameters, we consider the scattering for a one-dimensional rigid regular chain of identical atoms. Without loss of generality and for simplification we consider the resonant scattering of particles experiencing s -scattering by an individual nucleus of zero spin in the ground state. In this case the general expression for the wavefunction of a particle can be written as

$$\psi(\mathbf{r}) = e^{i\mathbf{k}\mathbf{r}} + \sum_m A_m \frac{\exp(ik|\mathbf{r} - \mathbf{r}_m|)}{|\mathbf{r} - \mathbf{r}_m|}. \quad (2.1)$$

The coefficients A_m may be determined by using the fact that the logarithmic derivative of the function $\chi = |\mathbf{r} - \mathbf{r}_m|\psi(\mathbf{r})$ is

$$\gamma_0 = \frac{1 + ikf}{f} \quad (2.2)$$

for $|\mathbf{r} - \mathbf{r}_m| \rightarrow 0$ and arbitrary m . Here f is the scattering amplitude of an individual atom. This results in a set of algebraic equations

$$\begin{aligned} \sum_m g_{pm} A_m &= -e^{-i\mathbf{k}\mathbf{r}_p}, \\ g_{pm} &= \frac{\exp(ik|\mathbf{r}_p - \mathbf{r}_m|)}{|\mathbf{r}_p - \mathbf{r}_m|}, \quad m \neq p, \quad g_{mm} = ik - \gamma_0. \end{aligned} \quad (2.3)$$

Neglecting edge effects in a sufficiently long chain the solution of eq. (2.3) can be written

$$A_m = A e^{ik_z z_m}, \quad z_m = am. \quad (2.4)$$

The z axis lies along the chain.

In the case of pure resonant scattering

$$f = -\frac{1}{2k} \frac{\Gamma_{\text{el}}}{E - E_0 + (i\Gamma)/2},$$

where Γ_{el} and Γ are the elastic and total width, respectively. Then, substituting eq. (2.4) into eq. (2.3), we find

$$A = -\frac{1}{2k} \frac{\Gamma_{\text{el}}}{E - E_0 + (i\Gamma)/2 + R}, \quad R = \frac{\Gamma_{\text{el}}}{2k} \sum_{m \neq p} g_{mp} e^{ik_z(z_p - z_m)}. \quad (2.5)$$

The series in eq. (2.5) can be summed. As a result, we arrive at the following expressions for the variation $\Delta\Gamma$ of the elastic width and shift ΔE of the resonant level:

$$\begin{aligned}\Delta\Gamma &= 2\text{Im}R = \frac{\pi\Gamma_{\text{el}}}{ak} \left(1 - \left\{ \frac{a(k+k_z)}{2\pi} \right\} - \left\{ \frac{a(k-k_z)}{2\pi} \right\} \right), \\ \Delta E &= -\text{Re}R = \frac{\Gamma_{\text{el}}}{ak} \ln \left[2 \left| \sin \left(\frac{a(k+k_z)}{2} \right) \sin \left(\frac{a(k-k_z)}{2\pi} \right) \right| \right].\end{aligned}\quad (2.6)$$

Here $\{x\}$ is the fractional part of x defined as the difference between x and the entier of x . It follows from eq. (2.6) that the elastic width can increase as well as decrease, depending on the magnitude of k and on the direction of the incident beam with respect to the chain. This interesting circumstance is a direct consequence of the collective character of the excited state appearing in the scattering at the stage of the primary absorption of a particle. To verify it, let us consider the probability of the elastic decay of a collective excitation described by the wavefunction eq. (1.1):

$$W_{\text{el}} = \frac{2\pi}{\hbar N} \int |M|^2 \left| \sum_m e^{i(\mathbf{k}-\mathbf{k}')\mathbf{r}_m} \right|^2 \delta(E_0 - E_{\mathbf{k}'}) \frac{d^3k'}{(2\pi)^3}. \quad (2.7)$$

Here M is the matrix element corresponding to the transition of an individual nucleus from the excited state to the ground state with the emission of a primary particle.

In the case of the one-dimensional chain it is convenient to rewrite the phase volume d^3k' as

$$d^3k' = 2\pi k' \frac{dk'}{dE_{k'}} dE_{k'} dk'_z.$$

Then the integration in eq. (2.7) is straightforward. As a result, we find

$$\begin{aligned}W_{\text{el}} &= W_{\text{el}}^{(0)} F, \\ F &= \frac{1}{2kN} \int dk'_z \frac{\sin^2((k_z - k'_z)aN/2)}{\sin^2((k_z - k'_z)a/2)} \\ &= 1 + \frac{\pi}{ak} \left(1 - \left\{ \frac{a(k+k_z)}{2\pi} \right\} - \left\{ \frac{a(k-k_z)}{2\pi} \right\} \right),\end{aligned}\quad (2.8)$$

where $W_{\text{el}}^{(0)}$ is the probability of the elastic decay of an individual nucleus.

Comparing eq. (2.8) with eq. (2.6), we see that the variation of the elastic width is connected unambiguously with the decay of the collective excitation extended over the chain.

One can readily check that the inelastic decay of the collective excitation, which is accompanied inevitably by a change of the ground state of one of the atoms, is

realized with the same probability as for an individual atom. This also leads to a variation of the relation between elastic and inelastic scattering cross-sections.

Let us consider a chain of finite size. For a distance large compared with the size of the chain, the wavefunction eq. (2.1) can be rewritten as

$$\Psi(\mathbf{r}) \approx e^{i\mathbf{k}\mathbf{r}} + A \left(\sum_m e^{i(k_z - k'_z)z_m} \right) \frac{e^{ikr}}{r}.$$

Here k'_z is the component of the wave vector of the scattered particles in the direction of the chain.

This expression allows us to write down the differential scattering cross-section per atom. Using eq. (2.5), we have

$$d\sigma_{\text{el}} = \frac{1}{4k^2} \frac{\Gamma_{\text{el}}^2}{(E - E'_0)^2 + \Gamma'^2/4} \frac{1}{N} \left| \sum_m e^{i(k_z - k'_z)z_m} \right|^2 d\Omega_{\mathbf{k}'}, \quad (2.9)$$

$$E'_0 = E_0 + \Delta E \quad \text{and} \quad \Gamma' = \Gamma + \Delta\Gamma.$$

Taking into account that $d\Omega_{\mathbf{k}'} = 2\pi k^{-1} dk'_z$, we can reduce the calculation of the total cross-section of elastic scattering to the same integral as for deriving eq. (2.7). As a result, taking into account eq. (2.6) we find

$$\sigma_{\text{el}} = \frac{\pi}{k^2} \frac{\Gamma_{\text{el}}\Gamma'_{\text{el}}}{(E - E'_0)^2 + \Gamma'^2/4} \quad \text{with} \quad \Gamma'_{\text{el}} = \Gamma_{\text{el}} + \Delta\Gamma. \quad (2.10)$$

The cross-section σ_{in} , related to the inelastic decay channel, is determined by the same formula eq. (2.10), replacing Γ'_{el} by the inelastic part Γ_{in} of the width. The ratio of both cross-sections

$$\frac{\sigma_{\text{el}}}{\sigma_{\text{in}}} = \frac{\Gamma'_{\text{el}}}{\Gamma_{\text{in}}} \quad (2.11)$$

can be either larger or smaller than the corresponding value of an individual atom.

The collective character of excitations results in the shift ΔE of the resonant level. From eq. (2.6) it follows that the shift may have a large value and both signs.

It is worthwhile to note that more drastic changes both for the variation of the resonant nuclear parameters and for the role of the collective character of the excitations are disclosed in the course of analyzing the scattering in a two-dimensional lattice (see [6]).

3. Change of resonant nuclear parameters in a thick crystal. Suppression of elastic scattering

The picture of resonant interaction in a three-dimensional crystal differs essentially from the case considered above. This is connected mainly with the fact that the amplitude of the primary wave will have different magnitudes at nuclei of different

crystal planes, decreasing along k . This gives rise to difficulties in determining the explicit form of the solution, especially if the crystal size l is comparable with the thickness of the transition layer, that is, if $l|f|/v_0k < 1$, v_0 being the volume of the unit cell.

In the opposite limiting case $l|f|/v_0k \gg 1$, however, the problem is simplified because we can ignore the transition layer in the thick crystal and take into account the incident wave effectively only in the boundary condition. We will consider the latter case below.

Let us return to the general expression eq. (2.1). We seek the coefficients A_m in the form

$$A_m = Ae^{i\boldsymbol{\kappa}\mathbf{r}_m}, \quad (3.1)$$

where $\boldsymbol{\kappa}$ is a complex vector. Then we obtain after identical transformations

$$\psi(\mathbf{r}) = e^{i\mathbf{k}\mathbf{r}} + 2iA \sum_m e^{i\boldsymbol{\kappa}\mathbf{r}_m} \frac{\sin k|\mathbf{r} - \mathbf{r}_m|}{|\mathbf{r} - \mathbf{r}_m|} + A \sum_m e^{i\boldsymbol{\kappa}\mathbf{r}_m} \frac{\exp(-ik|\mathbf{r} - \mathbf{r}_m|)}{|\mathbf{r} - \mathbf{r}_m|}. \quad (3.2)$$

With the relation

$$\frac{\sin k|\mathbf{r} - \mathbf{r}_m|}{|\mathbf{r} - \mathbf{r}_m|} = \frac{k}{4\pi} \int \exp[i\mathbf{k}'(\mathbf{r} - \mathbf{r}_m)] d\Omega_{k'}$$

the second term in eq. (3.2) is then written as

$$A \frac{ik}{2\pi} \int e^{i\mathbf{k}'\mathbf{r}} \left(\sum_m \exp[i(\boldsymbol{\kappa} - \mathbf{k}')\mathbf{r}_m] \right) d\Omega_{k'}. \quad (3.3)$$

Here we represent the vector \mathbf{k} in the form

$$\boldsymbol{\kappa} = \mathbf{k} + \mathbf{q} \quad (3.4)$$

and consider for simplicity the case when the flux is normally incident on the surface of the crystal, that is, $\mathbf{q} \parallel \mathbf{k}$. Since the interaction with each crystal plane is assumed to be weak ($|f| \ll a$), we get

$$|\mathbf{q}| \ll \frac{2\pi}{a}.$$

In this case the sum in (3.3) can be replaced by an integral in the standard fashion. As a result, we find, assuming henceforth that there is no Bragg scattering,

$$\sum_m \exp[i(\boldsymbol{\kappa} - \mathbf{k}')\mathbf{r}_m] \approx \frac{(2\pi)^2}{v_0} \delta(k'_x) \delta(k'_y) \frac{i}{k_z - k'_z + q_z}. \quad (3.5)$$

Here the z axis is directed along \mathbf{k} .

The presence of two δ -functions in eq. (3.5) and the involvement of the condition $k' = k$ lead to the equalities $k'_z = k_z$ and $\mathbf{k}' = \mathbf{k}$. Then, substituting eq. (3.5) into (3.3) and involving the result of eq. (3.2), we readily obtain

$$\psi(r) = e^{i\mathbf{k}\mathbf{r}} - A \frac{2\pi}{v_0 k q} e^{i\mathbf{k}\mathbf{r}} + A e^{i\mathbf{k}\mathbf{r}} \left(\sum_m e^{i\mathbf{k}\mathbf{r}_m - \mathbf{r}} \frac{\exp(-ik|\mathbf{r} - \mathbf{r}_m|)}{|\mathbf{r} - \mathbf{r}_m|} \right). \quad (3.6)$$

The expression in brackets is a periodic function. Owing to the factor $\exp(i\mathbf{k}\mathbf{r})$ the last term in eq. (3.6) as a whole decreases continuously as the crystal thickness ($\text{Im } q > 0$) increases. It is clear that the second term should cancel the first in a sufficiently thick crystal, that is,

$$A \frac{2\pi}{v_0 k q} = 1, \quad (3.7)$$

and

$$\psi(\mathbf{r}) = A e^{i\mathbf{k}\mathbf{r}} \left(\sum_m e^{i\mathbf{k}\mathbf{r}_m - \mathbf{r}} \frac{\exp(-ik|\mathbf{r} - \mathbf{r}_m|)}{|\mathbf{r} - \mathbf{r}_m|} \right) \equiv A e^{i\mathbf{k}\mathbf{r}} \Phi(\mathbf{r}). \quad (3.8)$$

The relation (3.7) determines in fact the constant A of the solution eq. (3.1). The condition (2.2) on each nucleus makes it possible to determine the equation for q by using eq. (3.8).

Omitting the details of calculation [6], we give the final result for q :

$$q = -\frac{\pi}{v_0} \frac{1}{k^2} \frac{\Gamma_{\text{el}}}{E - E'_0 + i\Gamma_{\text{in}}/2} \quad \text{with} \quad E'_0 = E_0 - \frac{\Gamma_{\text{el}}}{2k} D, \quad (3.9)$$

$$D = \frac{4\pi}{v_0} \left[\sum_{\mathbf{K} \neq 0} \frac{1}{(\mathbf{k} + \mathbf{K})^2 - k^2} - \frac{v_0}{(2\pi)^3} \int \frac{d^3 q_1}{q_1^2} \right]. \quad (3.10)$$

Here \mathbf{K} is the reciprocal lattice vector. The second term in the brackets cancels the divergence of the first term.

The imaginary part of q is connected with the total cross-section σ_t per nucleus via the relation $\text{Im } q = \frac{1}{2} n \sigma_t = (1/2v_0) \sigma_t$. Hence,

$$\sigma_t = \frac{\pi}{k^2} \frac{\Gamma_{\text{el}} \Gamma_{\text{in}}}{(E - E'_0)^2 + \Gamma_{\text{in}}^2/4}. \quad (3.11)$$

Expressions (3.11) and (3.9) show a remarkable result. In a thick crystal the width of the resonant level is completely independent of the elastic width Γ_{el} . This is a typical coherent phenomenon. From the physical point of view the phenomenon is due to the fact that the particle in a thick crystal becomes a quasiparticle as a result of the collective interaction. However, in a regular crystal the momentum (more accurately, quasimomentum) of a quasiparticle is conserved. This means that the particle is not subjected to scattering and the elastic decay of the collective excited state takes place

strictly in the direction of incidence of a particle. In the course of deriving Γ_{el} one assumes implicitly that the decay may occur in any direction.

Analyzing the result of eqs. (3.7)–(3.11), we see again how the coherent collective effects change the nuclear parameters in thick crystals.

So far, considering the role of the coherent effects, we have ignored the incoherent channels of scattering. The presence of the latter reduces the scale of the variation of the nuclear parameters. The most pronounced channels are spin incoherence and incoherence due to excitation of the phonon system with resonant scattering. A detailed analysis of the problem is given in [8]. It is found for all cases that the variation of the resonant width in a thick crystal with involvement of incoherent channels can be represented as

$$\Delta\Gamma = -\Gamma_{el}\xi\varphi(T)\eta. \quad (3.12)$$

Here ξ is the spin incoherence factor. In the case of an unsplit HF structure

$$\xi = \frac{(2I + 1)}{(2I_0 + 1)(2l + 1)}, \quad (3.13)$$

where I_0 and I are the spins of the ground and excited states of a nucleus and l is the multipolarity of the γ -transition. (For resonant scattering of particles of spin $s = 1/2$, the quantity l in eq. (3.13) should be replaced by $1/2$.) In the case of a resolved HF structure the spin factor ξ takes a different value for each transition. So, for ^{57}Fe with $I_0 = 1/2$ and $I = 3/2$, magnetic dipole ($\pm 1/2 \rightarrow \pm 3/2$) transitions have a spin factor of $\xi = 1$ since during the decay of the excited state the nucleus can return only to the initial state. For a transition of the ($1/2 \rightarrow \pm 1/2$) type, the spin factor is $\xi < 1$ because the excited nucleus may return to any of two levels of the ground state.

The temperature factor $\varphi(T)$ is due to excitation of phonons. For $T \ll \Theta_D$, Θ_D being the Debye temperature, the temperature factor is close to the probability amplitude of the Mössbauer effect, i.e., to the recoilless transition probability at the excitation of a nucleus. The isotopic incoherence factor η is equal to the relative concentration of the resonant isotope.

Provided all these incoherent factors are close to unity, we have $\Delta\Gamma \approx -\Gamma_{el}$, corresponding exactly to the above-mentioned effect of suppressing the elastic part of scattering in a thick perfect crystal.

The effect of suppression of the elastic scattering channel has been observed experimentally for the first time by Smirnov and Shvyd'ko [29]. The authors observed the scattered emission in the direction normal to the beam of resonant quanta travelling across a nearly ideal single crystal $^{57}\text{FeBO}_3$. The well-resolved HF structure gives an opportunity to measure the intensity of scattering with the energy of the incident radiation corresponding to the ($\pm 1/2 \rightarrow \pm 3/2$) and ($\pm 1/2 \rightarrow \mp 1/2$) transitions. The authors have found that the scattering is significantly smaller in the first case compared with the second transitions whereas the inverse is true in absorption. The drastic reduction of resonant scattering into off Bragg directions in the first case, where the spin-incoherent scattering is forbidden, is striking evidence for the suppression of the

elastic scattering channel when the proper scattering is associated with the incoherent processes alone.

4. Speed-up of nuclear de-excitation in a thin crystal

A specific feature of the resonant interaction with nuclei having anomalously long-lived isomeric states is the possibility to measure the time behaviour of the decay of the excited states. Due to this there is also a unique possibility for separation from the primary beam and measuring forward scattering.

Unlike the preceding section, let us consider a thin crystal of thickness l satisfying the condition

$$\frac{1}{v_0} \sigma_t l < 1. \quad (4.1)$$

Here σ_t is the total scattering cross-section for a single nucleus. In this case the absorption of a single γ -quantum gives rise to a collective state with practically the same excitation amplitudes of all atoms. Thus, a bulk nuclear exciton appears with a wavefunction of the same structure as eq. (1.1). Note that we do not have such a situation in a thick crystal since the wavefunction falls off far from the entrance surface.

Consider the decay of such an exciton, taking again for simplicity a rigid crystal lattice and zero ground state spin. Then we can employ the general expression (2.7) for determining the reemission probability of γ -quanta. For an arbitrary direction of \mathbf{k} with respect to the lattice, except those permitting Bragg scattering, the lattice sum in eq. (2.7) allows only forward decay. If eq. (4.1) is supplemented by the condition $2\pi\hbar c/l \gg \Gamma$, which is usually weaker than eq. (4.1), the smearing of the integrand eq. (2.7) in the momentum space proves to be larger than the real smearing of the δ -function over energy. Taking this into account, we find [5]

$$W_{\text{el}} \approx W_{\text{el}}^{(0)} \frac{\pi l}{k^2 v_0}, \quad l \gg a. \quad (4.2)$$

Thus, we have an enhancement for the rate of the exciton γ -decay accompanied by a sharp peak of the radiation pattern in the direction of the incident vector \mathbf{k} . The large enhancement, limited by eq. (4.1), is possible only for $\Gamma_{\text{in}} \gg \Gamma_{\text{el}}$. Note that the requirement for the quality of a crystal is more moderate for the forward decay.

Provided the direction of the vector \mathbf{k} satisfies the Bragg condition $|\mathbf{k} + \mathbf{K}| \approx k$, there arises an additional decay channel in the direction of the Bragg angle. In this case the decay probability has the same magnitude (4.2) with the conservation of condition (4.1) [4,5].

It should be emphasized that the decay speed-up effect is associated with the excitation of the crystal as a whole. In a thick crystal, provided the condition opposite to eq. (4.1) is fulfilled, such a situation does not exist. In reality, multiple rescattering takes place inside a crystal. It is interesting, as we have seen in the preceding sec-

tion, that the elastic width of a single nucleus reduces or even vanishes under these conditions.

The speed-up of the forward decay after fast chopping of the γ -quantum beam has been observed in the remarkable work of Shvyd'ko et al. [28]. The original shutter, designed by this group and based on the re-alignment of the spin system with the aid of a magnetic field in a $^{57}\text{FeBO}_3$ crystal, allowed them to interrupt the primary beam from a stationary Mössbauer source within 10 ns. (The lifetime of the excited state of a ^{57}Fe nucleus is $\tau_0 = 140$ ns.) After chopping the forward emission from a crystal occurs effectively during a time significantly shorter than τ_0 . The observation gives evidence for the collective character of exciting the nuclear system with the conservation of coherent coupling between phases at individual nuclei. In parallel this phenomenon was discovered and studied in experiments with SR [36]. The predicted speed-up of the collective decay in the Bragg channel was observed in [55]. We will return to the problem treated in this section when analyzing the interaction of the SR with crystals.

5. Dynamical equations of the resonant diffraction

In this section we will derive the general dynamical equations describing the propagation of γ -quanta inside a thick crystal under the condition of resonant diffraction for an arbitrary scale of the conversion and incoherent channels.

Diffraction scattering is a purely elastic coherent process in which the quantum mechanical state of a crystal remains the same. This allows us to use the classical Maxwell equations for describing the electromagnetic field inside the crystal. The role of inelastic and incoherent channels finds its reflection in the determination of the current induced by the electromagnetic radiation in the crystal.

The Maxwell equation for the Fourier component $\mathbf{E}(\mathbf{k}, \omega)$ of the electromagnetic field can be written as

$$\left(k^2 - \frac{\omega^2}{c^2}\right)\mathbf{E}(\mathbf{k}, \omega) - \mathbf{k}(\mathbf{k}\mathbf{E}(\mathbf{k}, \omega)) = \frac{4\pi}{c^2}i\omega\mathbf{j}(\mathbf{k}, \omega). \quad (5.1)$$

The large times of the nucleus–radiation interaction correspond to purely elastic scattering. Under these conditions the current $\mathbf{j}(\mathbf{k}, \omega)$ on the right-hand side of eq. (5.1) represents the quantum mechanical average for the Fourier component of the nuclear current density operator $\hat{\mathbf{j}}(\mathbf{k}, \omega)$ over the state of a crystal in the presence of the field $\mathbf{E}(\mathbf{k}, \omega)$. First, we ignore the interaction with electrons. In the crystal

$$\hat{\mathbf{j}}(\mathbf{k}, \omega) = \sum_n e^{-i\mathbf{k}\mathbf{r}_n} \hat{\mathbf{j}}_n(\mathbf{k}, \omega), \quad (5.2)$$

where $\hat{\mathbf{j}}$ is the current density operator for a nucleus at site n , and \mathbf{r}_n is the coordinate of its center of gravity.

To simplify, we assume $\Gamma_{\text{in}} \gg \Gamma_{\text{el}}$ and neglect the variation of the resonant parameters. As a gauge condition, let us set the scalar potential equal to zero. Then,

within the framework of standard perturbation theory we have for the average value of the current density in linear approximation for the field

$$j^i(\mathbf{k}, \omega) = \int \frac{d^3 k'}{(2\pi)^3} \sigma_\omega^{il}(\mathbf{k}, \mathbf{k}') E^l(\mathbf{k}', \omega), \quad (5.3)$$

$$\sigma_\omega^{il} = \frac{i}{\omega} \sum_n \sum_s \frac{\overline{(\hat{j}_n^{i*}(\mathbf{k}) e^{-i\mathbf{k}\mathbf{r}_n})_{s_0 s} (\hat{j}_n^l(\mathbf{k}') e^{i\mathbf{k}'\mathbf{r}_n})_{s s_0}}}{\hbar\omega - E_n(s_0, s) + i\Gamma/2}. \quad (5.4)$$

Here s_0 and s characterize the states of the phonon spectrum and the nuclear quantum numbers ζ_0 and ζ are related to the initial and excited states, respectively. Thus,

$$E_n(s_0, s) = E_n(\zeta_0, \zeta) + \sum_\beta \hbar\omega_\beta (n_\beta^s - n_\beta^{s_0}), \quad (5.5)$$

where $E_n(\zeta_0, \zeta)$ is the energy of the corresponding nuclear transition. The upper bar in eq. (5.4) means averaging over the initial state.

Retaining index n for the numbers of the elementary cell, introducing index p for the position of a nucleus in the elementary cell, and denoting the vibrational displacement of a nucleus as \mathbf{u}_p , we represent \mathbf{r}_n in the following form:

$$\mathbf{r}_n = \mathbf{R}_n + \boldsymbol{\rho}_p + \mathbf{u}_p.$$

The current density operator and the energy of the transition have the index p in this notation. Then, the summation over \mathbf{n} is reduced to a lattice sum

$$\sum_{\mathbf{n}} e^{i(\mathbf{k}' - \mathbf{k})\mathbf{R}_n} = \eta \frac{(2\pi)^3}{v_0} \sum_{\mathbf{k}} \delta(\mathbf{k}' - \mathbf{k} - \mathbf{K}), \quad (5.6)$$

η being the relative concentration of the resonant isotope. The sum over the phonon states in eq. (5.4) is given by

$$\sum_{\{n_s\}} \frac{(e^{-i\mathbf{k}\mathbf{u}_p})_{\{n_s\}\{n_{s_0}\}} (e^{i\mathbf{k}'\mathbf{u}_p})_{\{n_{s_0}\}\{n_s\}}}{\hbar\omega - E_p(\zeta_0, \zeta) - \sum_\beta (n_\beta^s - n_\beta^{s_0}) \hbar\omega_\beta + i\Gamma/2}. \quad (5.7)$$

The specific feature of the Mössbauer transition is the smallness of the width Γ of excited states compared with the typical energies Θ_{ph} of the phonon spectrum. The condition $\Gamma \ll \Theta_{\text{ph}}$ means that the term with $\{n_s\} = \{n_{s_0}\}$ is predominant in the sum (5.7). The diagonal element $(e^{-i\mathbf{k}\mathbf{u}_p})_{\{n_{s_0}\}\{n_{s_0}\}}$ is simply the probability amplitude of the Mössbauer effect of the nucleus at site p :

$$f_p(\mathbf{k}) = \exp\left\{-\frac{1}{2} Z_p(\mathbf{k})\right\}, \quad Z_p = \frac{\hbar}{2M} \sum_\beta \frac{[\mathbf{k}\mathbf{e}_p(\beta)]^2}{\omega_\beta} (2\bar{n}_\beta + 1). \quad (5.8)$$

Here $\mathbf{e}_p(\beta)$ is the polarization vector of the β th phonon mode. Eventually, we arrive at an interesting result. In the case of narrow excited isomer states the amplitude σ_ω^{il} or the elastic resonant scattering amplitude contains a product $f_p(\mathbf{k})f_p(\mathbf{k}')$. The physical

meaning lies in the following. Due to its long lifetime the nucleus is subjected to an impact at absorption and to another absolutely independent one at reemission. For the inverse relation $\Gamma \gg \Theta_{\text{ph}}$, we arrive at the single factor $f_p(\mathbf{k} - \mathbf{k}')$, omitting the phonon contribution in the denominator of eq. (5.7) and performing direct summation. In this case the nucleus experiences only one impact determined by the difference $\mathbf{k} - \mathbf{k}'$. This is the standard Debye–Waller factor typical for the resonant scattering of neutrons and the usual potential scattering of X-rays.

Taking into account the results obtained, we find

$$\begin{aligned} \sigma_{\omega}^{il}(\mathbf{k}, \mathbf{k}') &= (2\pi)^3 \sum_{\mathbf{K}} \delta(\mathbf{k}' - \mathbf{k} - \mathbf{K}) \sigma_{\omega}^{il}(\mathbf{k}, \mathbf{k} + \mathbf{K}), \quad (5.9) \\ \sigma_{\omega}^{il}(\mathbf{k}, \mathbf{k} + \mathbf{K}) &= \frac{i\eta}{\omega v_0} \frac{1}{2I_0 + 1} \sum_{p, \zeta_0, \zeta} e^{i\mathbf{K}\rho_p} \frac{(\hat{j}_p^{i*}(\mathbf{k}))_{\zeta_0\zeta} (\hat{j}_p^l(\mathbf{k} + \mathbf{K}))_{\zeta\zeta_0}}{\hbar\omega - E_p(\zeta_0, \zeta) + i\Gamma/2} \\ &\quad \times f_p(\mathbf{k}) f_p(\mathbf{k} + \mathbf{K}). \quad (5.10) \end{aligned}$$

In this expression the factor $(2I_0 + 1)^{-1}$ originates from averaging over the initial states of a nucleus.

Let us substitute eqs. (5.9) and (5.10) into the general relation for the current density eq. (5.3). Inserting the expression derived on the right-hand side of eq. (5.1), we obtain the complete set of equations for the electric field components. Then we introduce the notation

$$\mathbf{k}_h = \mathbf{k}_0 + \mathbf{K}_h, \quad \kappa = \frac{\omega}{c} \quad (5.11)$$

and represent a set of equations in the compact form

$$(\mathbf{k}_h^2 - \kappa^2) E^i(\mathbf{k}_h) - k_h^i (\mathbf{k}_h \mathbf{E}(\mathbf{k}_h)) = \kappa^2 \sum_{h'} g_{hh'}^{il} E^l(\mathbf{k}_{h'}), \quad (5.12)$$

$$g_{hh'}^{il} = -\frac{4\pi\eta}{\omega^2 v_0} \frac{1}{2I_0 + 1} \sum_{p, \zeta_0, \zeta} \frac{A_h^{i*}(p, \zeta_0, \zeta) A_{h'}^l(p, \zeta_0, \zeta)}{\hbar\omega - E_p(\zeta_0, \zeta) + i\Gamma/2}. \quad (5.13)$$

In expression (5.13)

$$\mathbf{A}_h(p, \zeta_0, \zeta) = f_p(\mathbf{k}_h) e^{i\mathbf{k}_h \rho_p} (\hat{\mathbf{j}}_p(\mathbf{k}_h))_{\zeta_0\zeta}. \quad (5.14)$$

For simplicity the index ω is omitted after this.

Let us restrict ourselves to considering only E1 and M1 transitions. In these cases the current density operator reduces to

$$\hat{\mathbf{j}}(\mathbf{k}) = i\omega \hat{\mathbf{d}} \quad (\text{E1}); \quad \hat{\mathbf{j}}(\mathbf{k}) = ic [\mathbf{k} \hat{\boldsymbol{\mu}}] \quad (\text{M1}). \quad (5.15)$$

Here $\hat{\mathbf{d}}$ and $\hat{\boldsymbol{\mu}}$ are the operators of the electric and magnetic dipole moments, respectively. Introducing m_0 and m for the spin projections of a nucleus in the ground

and excited states onto the quantization axis \mathbf{n}_0 , we find for the matrix element of operator $\hat{\mathbf{d}}$:

$$(\hat{\mathbf{d}})_{m_0 m} = -i \left(\frac{3(2I+1)\Gamma_{\text{el}}}{4\kappa^3} \right)^{1/2} (-1)^q \begin{pmatrix} I_0 & 1 & I \\ m_0 & q & m \end{pmatrix} \mathbf{n}_q. \quad (5.16)$$

The same formula is valid for the operator $\hat{\boldsymbol{\mu}}$. In eq. (5.16) the matrix

$$\begin{pmatrix} I_0 & 1 & I \\ m_0 & q & m \end{pmatrix}$$

is the standard $3j$ -symbol, $q = m_0 - m$, and $\mathbf{n}_{\pm 1} = \mp(\mathbf{n}_x \pm i\mathbf{n}_y)$, where \mathbf{n}_x and \mathbf{n}_y are two vectors orthogonal to each other and to \mathbf{n}_0 .

Equations (5.12)–(5.14) represent a full set in the dynamical theory of resonant nuclear diffraction, describing the coherent coupling of the diffracted field components $\mathbf{E}(\mathbf{k}_h)$ with the field $\mathbf{E}(\mathbf{k}_0)$ of the incident wave. The solution of this set governs the field inside the crystal. We emphasize that the superposed field describes the state of a single photon.

The coefficients (5.13) are in fact the amplitudes of the coherent elastic scattering by a crystal. It is essential that these amplitudes always remain finite regardless of the scale of the incoherent inelastic processes. However, the coherent coupling between the field components becomes weaker. Usually $\Gamma_{\text{in}} \gg \Gamma_{\text{el}}$ and the availability of the conversion decay channel diminishes the coherent amplitude in the resonance by a factor of $\Gamma_{\text{el}}/\Gamma_{\text{in}}$. The incoherence due to exciting the phonon system is associated with the appearance of factors $f_p(\mathbf{k}_h) < 1$ decreasing with temperature. The spin incoherence is reflected in the factor $(2I_0 + 1)$.

Despite the resonant interaction the dimensionless coefficients $g_{hh'}^{il}$ prove to be much smaller than unity. For a typical case, $|g_{hh'}^{il}| \leq 10^{-3}$, i.e., the amplitudes only for which

$$\frac{1}{\kappa^2} (\mathbf{k}_h^2 - \kappa^2) \leq |g_{0h}| \quad (5.17)$$

will be noticeably different from zero.

In essence, this reduces the infinite set of equations (5.12) to a finite set for the fields satisfying condition eq. (5.17). In principle, the other field components can be determined in the framework of perturbation theory, resulting in a certain renormalization of the coefficients $g_{hh'}^{il}$. However, these corrections turn out to be small and can be neglected henceforth.

The smallness of the coefficients $g_{hh'}^{il}$ also entails that the longitudinal field originating from scattering in a medium proves to be small. From eq. (5.12) we conclude easily that the longitudinal component is $|\mathbf{k}\mathbf{E}_h| \sim |g_{00}||\mathbf{k}_h \times \mathbf{E}_h|$. This allows us to omit the second term on the left-hand side of eq. (5.1) and consider the field in a crystal as completely transverse.

For the experimental realization, the case when the Bragg condition (5.6) is fulfilled only for a single wave of $h = 1$ is of highest interest. Thus, eq. (5.12) simplifies strongly and is reduced, involving the above, to

$$\begin{aligned} \left(\frac{k_0^2}{\kappa^2} - 1\right) E_0^i &= g_{00}^{il} E_0^l + g_{01}^{il} E_1^l, \\ \left(\frac{k_1^2}{\kappa^2} - 1\right) E_1^i &= g_{10}^{il} E_0^l + g_{11}^{il} E_1^l. \end{aligned} \quad (5.18)$$

Taking into account the transverse character of the field, we can choose two polarization vectors orthogonal to \mathbf{k}_h and to each other for each wave:

$$\mathbf{E}_h = E_h^{(1)} \mathbf{e}_h^{(1)} + E_h^{(2)} \mathbf{e}_h^{(2)}. \quad (5.19)$$

So, using this relation, we rewrite eqs. (5.18) in a form convenient for further analysis:

$$\begin{aligned} \left(\frac{k_0^2}{\kappa^2} - 1\right) E_0^{(s)} &= \sum_{s'=1,2} g_{00}^{ss'} E_0^{s'} + \sum_{s'=1,2} g_{01}^{ss'} E_1^{(s')}, \\ \left(\frac{k_1^2}{\kappa^2} - 1\right) E_1^{(s)} &= \sum_{s'=1,2} g_{10}^{ss'} E_0^{s'} + \sum_{s'=1,2} g_{11}^{ss'} E_1^{(s')}, \end{aligned} \quad (5.20)$$

where

$$g_{hh'}^{ss'} = \sum_{i,l} (\mathbf{e}_h^{(s)})_i^* g_{hh'}^{il} (\mathbf{e}_{h'}^{(s')})_l. \quad (5.21)$$

As a result, we arrive at a set of four equations for the scalar amplitudes $E_h^{(s)}$.

6. Solution of the diffraction problem. The suppression effect of inelastic channels

Let a γ -quantum flux with wave vector $\boldsymbol{\kappa}$ be incident on a lamina-shaped crystal. Refracted slightly due to $|g_{00}^{ss'}| \ll 1$ at the boundary, the flux propagates along the crystal with a wave vector

$$\mathbf{k}_0 = \boldsymbol{\kappa} + \delta \boldsymbol{\kappa} \mathbf{n}, \quad (6.1)$$

where \mathbf{n} is normal to the crystal surface

$$k_0 = \kappa(1 + \varepsilon_0), \quad |\varepsilon_0| \ll 1. \quad (6.2)$$

Then

$$\delta \approx \varepsilon_0 / \gamma_0, \quad \gamma_0 = \cos \Theta_0; \quad \Theta_0 = \widehat{\boldsymbol{\kappa} \mathbf{n}}. \quad (6.3)$$

The factors from the left-hand side of eq. (5.18) are (see eq. (5.11))

$$\begin{aligned} \frac{k_0^2}{\kappa^2} - 1 &= 2\varepsilon_0, & \frac{k_1^2}{\kappa^2} - 1 &= 2\varepsilon_0 \frac{\gamma_1}{\gamma_0} + \alpha, \\ \alpha &= \frac{\mathbf{K}_1(\mathbf{K}_1 + 2\boldsymbol{\kappa})}{\kappa^2}, & \gamma_1 &= \cos \Theta_1, & \Theta_1 &= \widehat{\mathbf{k}_1 \mathbf{n}}. \end{aligned} \quad (6.4)$$

The quantity α , characterizing the deviation from the exact Bragg condition, is connected with the deviation $\Delta\Theta$ from the Bragg angle Θ_B via the relation

$$\alpha = 2 \sin(2\Theta_B) \Delta\Theta. \quad (6.5)$$

In order to make the consideration more transparent, we first treat a relatively frequent situation when the tensor $g_{hh'}^{ss'}$ in (5.21) can be reduced by choosing polarization vectors to the following form:

$$g_{hh'}^{ss'} = g_{hh'}^{(s)} \delta^{ss'}. \quad (6.6)$$

In this case the set of equations (5.20) splits into two independent pairs of equations for $s = 1$ and 2

$$\begin{aligned} 2\varepsilon_0 E_0^{(s)} &= g_{00}^{(s)} E_0^{(s)} + g_{01}^{(s)} E_1^{(s)}, \\ (2\varepsilon_0/\beta + \alpha) E_1^{(s)} &= g_{10}^{(s)} E_0^{(s)} + g_{11}^{(s)} E_1^{(s)}, \\ \beta &= \gamma_0/\gamma_1. \end{aligned} \quad (6.7)$$

The condition for the existence of a nontrivial solution for this set results in a quadratic equation for ε_0 , the roots being

$$\varepsilon_{0s}^{(m)} = \frac{1}{4} (g_{00}^{(s)} + \beta g_{11}^{(s)} - \beta\alpha) \pm \frac{1}{4} [(g_{00}^{(s)} + \beta g_{11}^{(s)} - \beta\alpha)^2 + 4\beta (g_{00}^{(s)}\alpha - \Delta^{(s)})]^{1/2},$$

$$m = 1, 2, \quad (6.8)$$

$$\Delta^{(s)} = g_{00}^{(s)} g_{11}^{(s)} - g_{01}^{(s)} g_{10}^{(s)}. \quad (6.9)$$

The fields related to root m are denoted by $E_h^{(s)}(m)$.

Treating diffraction in Laue geometry, we have for the boundary conditions

$$E_0^{(s)}(1) + E_0^{(s)}(2) = \mathcal{E}_0^{(s)}, \quad E_1^{(s)}(1) + E_1^{(s)}(2) = 0. \quad (6.10)$$

Here $\mathcal{E}_0^{(s)}$ is the amplitude of the electric field incident on the crystal.

Defining the relation between the field components in eq. (6.7), we find for the wave field in the crystal, involving boundary conditions eq. (6.10) and eqs. (5.19), (6.2), and (6.3)

$$\begin{aligned} \mathbf{E}^{(s)}(\mathbf{r}) &= \mathcal{E}_0^{(s)} e^{i\boldsymbol{\kappa}\mathbf{r}} \frac{1}{2(\varepsilon_{0s}^{(2)} - \varepsilon_{0s}^{(1)})} \left\{ e^{i\boldsymbol{\kappa}\varepsilon_{0s}^{(1)}x/\gamma_0} [\mathbf{e}_0^{(s)} (2\varepsilon_{0s}^{(2)} - g_{00}^{(s)}) - \mathbf{e}_1^{(s)} \beta g_{10}^{(s)} e^{i\mathbf{K}\mathbf{r}}] \right. \\ &\quad \left. - e^{i\boldsymbol{\kappa}\varepsilon_{0s}^{(2)}x/\gamma_0} [\mathbf{e}_0^{(s)} (2\varepsilon_{0s}^{(1)} - g_{00}^{(s)}) - \mathbf{e}_1^{(s)} \beta g_{10}^{(s)} e^{i\mathbf{K}\mathbf{r}}] \right\} \end{aligned} \quad (6.11)$$

with $x = \mathbf{n}\mathbf{r}$.

When the deviation from the Bragg angle is large, i.e., $|\alpha| \gg g_{hh'}^{(s)}$, it follows from eq. (6.9) that

$$\varepsilon_{0s}^{(1)} \approx \frac{1}{2}g_{00}^{(s)}, \quad \varepsilon_{0s}^{(2)} \approx -\frac{1}{2}\beta\alpha.$$

In this case the amplitude of the reflected wave is about $g_{10}^{(s)}/\alpha$ and its role is insignificant, and the expression for the electric field can be simplified to

$$E^{(s)}(\mathbf{r}) = \mathbf{e}_0^{(s)} \mathcal{E}_0^{(s)} \exp\left(\mathbf{i}\kappa\mathbf{r} + \frac{\mathbf{i}\kappa g_{00}^{(s)}}{2\gamma_0}x\right). \quad (6.12)$$

Obviously, the decay of the field is associated with the imaginary part of $g_{00}^{(s)}$. On the other hand, the reduction of the intensity is connected with the total scattering cross-section σ_t via the standard exponential factor $\exp(-n\sigma_t x/\gamma_0)$, where n is the density of the scatterers. At last, according to the optical theorem, the imaginary part of the scattering amplitude f_{sc} for zero angle is related to σ_t via the relation

$$\text{Im } f_{sc}(0) = \frac{\kappa}{4\pi}\sigma_t.$$

As a result, all three characteristics are coupled together by

$$\text{Im } g_{00}^{(s)} = \frac{1}{\kappa}n\sigma_t^{(0)}f^2(\mathbf{k}_0) = \frac{4\pi n}{\kappa^2}\text{Im } f_{sc}(0). \quad (6.13)$$

Here $\sigma_t^{(0)}$ is the total scattering cross-section for a fixed nucleus. Since $f_{sc}(0)$ and g_{00} are associated with purely elastic scattering, the probability of a recoilless transition $f^2(\mathbf{k}_0)$ appears in σ_t . For simplification, this probability is assumed to be the same for ν resonant nuclei in the elementary cell. Treating the isolated resonant transition of energy E_0 and arbitrary degeneration, we conclude easily from eqs. (5.13), (5.14) and (5.21) that the imaginary part of $g_{00}^{(s)}$ is connected with the resonant denominator alone. Then, using the known expression for the total cross-section

$$\sigma_t^{(0)} = \frac{\pi}{k^2} \frac{\Gamma'_{\text{el}}\Gamma}{(\hbar\omega - E_0)^2 + \Gamma^2/4}, \quad \Gamma'_{\text{el}} = \Gamma_{\text{el}}\zeta, \quad (6.14)$$

we find for $g_{00}^{(s)}$ in the general case ($n = \eta\nu/v_0$)

$$g_{00}^{(s)} = -\frac{2\pi\eta\nu}{v_0\kappa^3} \frac{\Gamma'_{\text{el}}}{\hbar\omega - E_0 + \mathbf{i}\Gamma/2} f^2(k_0). \quad (6.15)$$

Here ζ is the spin incoherence factor.

For each polarization, the field inside the crystal (eq. (6.11)) represents a superposition of a pair of waves coupled coherently. However, the evolution of the waves takes place in different ways. So, if

$$\Delta^{(s)} = 0 \quad (6.16)$$

we have for the exact Bragg condition $\alpha = 0$,

$$\varepsilon_{0s}^{(1)} = 0, \quad \varepsilon_{0s}^{(2)} = \frac{1}{2}(g_{00}^{(s)} + \beta g_{11}^{(s)}). \quad (6.17)$$

Since the quantity $\text{Im} \varepsilon_{0s}^{(m)}$ governs the absorption coefficient of a crystal, it follows from eq. (6.17) that one coherent pair is strongly absorbed in a resonant medium (for $\beta = 1$, twice as large compared with a single wave) while the other pair propagates in the crystal without absorption.

Condition (6.16) is relatively easy to realize. If a well-resolved HF structure exists and the energy of the incident quanta is close to the energy of a single nondegenerate transition $E_{p'}(\zeta'_0, \zeta')$, this corresponds to the possibility of retaining only one term in the sum of eq. (5.13). Let us choose the polarization vector $\mathbf{e}_h^{(2)}$ in a direction normal to the vectors $\mathbf{A}_h(p', \zeta'_0, \zeta)$ in (5.14):

$$\mathbf{e}_h^{(2)} \cdot \mathbf{A}_h(p', \zeta'_0, \zeta) = 0.$$

It follows from eqs. (5.21) and (5.13) that γ -quanta polarized in this way do not interact at all with the nuclei within the approximation considered. However, γ -quanta of the $\mathbf{e}_h^{(1)} = (\mathbf{k}_h \times \mathbf{e}_h^{(2)})/k_h$ polarization interact strongly with the nuclear system. The tensors $g_{hh'}^{ss'}$ from eq. (5.21) differ from zero only for $s = s' = 1$ and, correspondingly, satisfy relation (6.6). Hence, all results of eqs. (6.7)–(6.11) are valid in this case. If one comes back to eqs. (5.13) and (5.14), it is easy to see that relation (6.16) is fulfilled identically.

Let us turn now to the opposite case, when HF splitting is absent. In this case the summation over ζ_0 and ζ in eq. (5.13) reduces to the determination of

$$\sum_{\zeta_0} (\hat{j}_p^i(\mathbf{k}_h) \hat{j}_p^l(\mathbf{k}_{h'}))_{\zeta_0 \zeta_0}. \quad (6.18)$$

Let us consider the case of an E1 transition. Since the current density operator eq. (5.15) is independent of \mathbf{k} , this sum is proportional to δ^{il} with the coefficient determined straightforwardly by eq. (5.16). As a result, we find

$$g_{hh'}^{il} = g_{hh'} \delta^{il}, \quad (6.19)$$

$$g_{hh'} = -\frac{2\pi\eta}{\kappa^3 v_0} \frac{2I + 1}{2I_0 + 1} \frac{\Gamma_{\text{el}}}{\hbar\omega - E_0 + i\Gamma/2} \sum_p e^{i\mathbf{k}\rho_p} f_p(\mathbf{k}_h) f_p(\mathbf{k}_{h'}).$$

Let us introduce the polarization vectors $\mathbf{e}_h^{(\sigma)}$ normal to the scattering plane and the vectors $\mathbf{e}_h^{(\pi)} = (\mathbf{k}_h \times \mathbf{e}_h^{(\sigma)})/k_h$ lying in the scattering plane. Taking into account eq. (6.18) we find that in this case relation (6.6) is valid. For a single resonant nucleus in the elementary cell or for the condition $e^{i\mathbf{k}\rho_p} = 1$ and $f_p(\mathbf{k}_h)$ independent of h , we have $\Delta^{(\sigma)} = 0$ for the σ -polarization.

In the case of an M1 transition, involving expression (5.15) for the current density operator and calculating eq. (6.18), we find for tensor (5.13)

$$g_{hh'}^{il} = \frac{1}{\kappa^2} (k_{h'}^i k_h^l - \delta^{il} (\mathbf{k}_h \cdot \mathbf{k}_{h'})) g_{hh'}, \quad (6.20)$$

where $g_{hh'}$ has the same magnitude as in eq. (6.19). It is easy in this case to trace that condition (6.6) is satisfied and the σ - and π -polarizations prove to be independent. Again, equality (6.16) is valid but now for the π -polarization.

Thus we see that, for a number of important cases, the set of four equations (5.20) decouples into the two pairs of equations for different polarizations and that condition (6.16) is satisfied, at least, for one polarization at the same time.

Let us return to the general problem involving eqs. (6.16) and (6.17). We obtained a unique result. In fact, for the resonant nuclear Bragg scattering of γ -quanta in the crystal there appears a pair of superposed states which is not subjected to absorption in spite of the presence of strong inelastic and incoherent channels. At the input the incident plane wave is converted into a combination of two pairs of waves. One of them is strongly absorbed in the crystal and, on the contrary, the other makes the thick crystal transparent. This effect, called suppression of the inelastic channel, is apparently the most striking phenomenon in the general problem of coherence with nuclear resonant scattering (NRS).

For large thicknesses, at which there remains practically only one pair of superposed states in eq. (6.11), the electric field, taking into account the quantities $\mathcal{E}_{0s}^{(m)}$ from eq. (6.8) with $\alpha = 0$, has the following form:

$$\mathbf{E}^{(s)}(\mathbf{r}) = \mathcal{E}_0^{(s)} e^{i\kappa\mathbf{r}} \frac{\beta}{g_{00}^{(s)} + \beta g_{11}^{(s)}} [\mathbf{e}_0^{(s)} g_{11}^{(s)} - \mathbf{e}_1^{(s)} g_{10} e^{i\kappa\mathbf{r}}]. \quad (6.21)$$

In all cases considered above one has $g_{hh'}^{(s)} \sim f(\mathbf{k}_h) f(\mathbf{k}_{h'})$. In an anisotropic crystal one has $g_{11}^{(s)} \neq g_{10}^{(s)}$ for the general case. This means that, for the nuclear sites where $e^{i\kappa\mathbf{r}} = 1$, neither the electric field nor the magnetic field of the wave with $(\mathbf{k}_h \times \mathbf{e}_h^{(s)})/k_h$ polarization vectors vanishes. For many cases, the vectors $\mathbf{e}_0^{(s)}$ and $\mathbf{e}_1^{(s)}$ are not collinear at all. If, e.g., for the E1 transition and internal magnetic field normal to the scattering plane the quantity $q = M - M_0 = \pm 1$ corresponds to the nondegenerated transition line, the electric field at the nucleus is circularly polarized (see details in [19]).

Actually, the nature of the suppression effect is associated with the vanishing of the total amplitude of formation of an excited nucleus in the pair superposed state. One can see this straightforwardly by analyzing the case of the nondegenerate transition line. The amplitude of formation of the excited nucleus is proportional to the product of the field intensity at the nuclear site and the matrix element for the transition from

the ground to the excited state. In correspondence with eqs. (6.20), (5.13) and (5.14) we have the amplitude of formation of the excited state in the incident wave

$$F_0 = C \frac{g_{11}^{(s)}}{g_{00}^{(s)} + \beta g_{11}^{(s)}} (\mathbf{e}_0^{(s)} \cdot \mathbf{A}_0).$$

The corresponding amplitude in the diffracted wave is ($e^{i\mathbf{k}\mathbf{r}_n} = 1$)

$$F_1 = -C \frac{g_{10}}{g_{00}^{(s)} + \beta g_{11}^{(s)}} (\mathbf{e}_1^{(s)} \cdot \mathbf{A}_1).$$

According to eqs. (5.21) and (5.13), $g_{hh'}^{(s)} \sim (\mathbf{e}_h^{(s)} \mathbf{A}_h)(\mathbf{e}_{h'}^{(s)} \mathbf{A}_{h'})$. Hence, we have

$$F = F_0 + F_1 = 0. \quad (6.22)$$

The same result is obtained straightforwardly from the consideration of the unsplit HF structure for E1 and M1 transitions, and σ - and π -polarizations, respectively.

A nontrivial specific feature of the suppression effect is the fact that temperature and vibration of the nuclei do not break it. This result has an interesting physical origin. As noted, the width Γ of the resonant levels is extremely small compared with the typical frequencies of the phonon spectrum. When absorbing γ -quanta by a nucleus in the crystal, there occurs no simultaneous emission or absorption of phonons without effective destruction of the resonant interaction. The absorption process, completely elastic in phonons, corresponds to a very long time of \hbar/Γ for the interaction of a γ -quantum with the nucleus. Eventually, a γ -quantum effectively “sees” the nuclei at their mean positions, in other words, it “feels” a regular lattice. This results in total suppression eq. (6.22) at an arbitrary temperature. On the contrary, provided $\Gamma \gg \Theta_{\text{ph}}$ as for the resonant scattering of neutrons, the interaction time with a nucleus proves to be shorter than the typical oscillation period of nuclei. In this case a neutron “sees” the instantaneous picture of the nuclear arrangement and, therefore, a disturbed periodical symmetry. This results in breaking condition (6.22) and in conserving, at least partially, the inelastic reaction channel. From a formal point of view this results in the replacement of $f(\mathbf{k}_h)f(\mathbf{k}_{h'})$ by $f(\mathbf{k}_h - \mathbf{k}_{h'})$, and, inevitably in $\Delta^{(s)} \neq 0$, i.e., in breaking down condition (6.16).

Note that the spin incoherence does not violate the suppression effect either because the condition $F = 0$ (6.22) locks the γ -decay channel necessary for displaying spin incoherence, i.e., the return of a nucleus into the ground state with another spin projection. Also it is of no significance how the resonant isotope with $\eta \neq 1$ is distributed over the lattice sites provided the exact periodicity of the lattice is conserved. Hence, the isotopic incoherence does not break the suppression effect either.

The presence of the inelastic and incoherent channels affects the collimation conditions. Let us analyze a small deviation from the exact Bragg condition $\alpha \ll |g_{01}|, |g_{10}|$. The reduction of the intensity of the superposed state, for which

the suppression effect takes place, is determined by the imaginary part $\text{Im } \varepsilon_{0s}^{(1)}$ (see eq. (6.11)). Using eq. (6.8), we find

$$\frac{\kappa l}{\gamma_0} \text{Im } \varepsilon_{0s}^{(1)} = -\frac{\kappa l}{\gamma_0} \frac{\beta^3}{2} \text{Im} \left(\frac{g_{00}^{(s)} g_{11}^{(s)}}{(g_{00}^{(s)} + \beta g_{11}^{(s)})^3} \right) \alpha^2 \equiv -\frac{\alpha^2}{2\alpha_0^2}. \quad (6.23)$$

It is easy to show that, for all cases, the larger the inelastic conversion width, i.e., the ratio $\Gamma/\Gamma_{\text{el}}$ and the spin I_0 of the ground state, the smaller the quantity α_0 . Also, the smaller the probability of the Mössbauer effect ($f(\mathbf{k}_h)$) and the smaller the concentration of the resonant isotope, the smaller the quantity α_0 .

Provided a divergent beam is incident on the crystal, we have for the integral transmitted intensity, using eqs. (6.11), (6.23) and (6.5),

$$I_T = \frac{1}{2 \sin 2\Theta_B} \int I_T(\alpha) d\alpha = I_0^{(s)} \frac{\sqrt{\pi}}{2 \sin 2\Theta_B} \left| \frac{\beta g_{11}^{(s)}}{g_{00}^{(s)} + \beta g_{11}^{(s)}} \right|^2 \alpha_0. \quad (6.24)$$

We see that the transmission intensity in the presence of the suppression effect falls off as $1/l^{1/2}$ with the thickness l of a crystal instead of the usual exponential decay.

In the general case, if eq. (6.6) is not valid, we should return to the general system eq. (5.20) involving eq. (6.4). The uniform system of four linear equations has a nontrivial solution, provided the corresponding determinant of fourth order vanishes. The equation obtained has four roots $\varepsilon_0^{(m)}$. A coherent superposition of four waves corresponds to every root. The incident plane wave is converted at the boundary into a coherent ensemble of four such quartets related to the various roots $\varepsilon_0^{(m)}$. The field inside the crystal is determined by the general expression (cf. eq. (6.11))

$$\mathbf{E}(\mathbf{r}) = e^{i\kappa\mathbf{r}} \sum_{m=1}^4 \exp(i\kappa\varepsilon_0^{(m)}x/\gamma_0) \left\{ \sum_{s=1,2} [\mathbf{e}_0^{(s)} E_0^{(s)}(m) + e^{i\mathbf{K}\mathbf{r}} \mathbf{e}_1^{(s)} E_1^{(s)}(m)] \right\}. \quad (6.25)$$

The boundary conditions are imposed by the same eqs. (6.10) in which the left-hand sides should have a sum of four fields corresponding to different m for each polarization s .

The suppression effect can occur when the condition

$$\text{Im } \varepsilon_0^{(m)} = 0 \quad (6.26)$$

is realized at least for one of the roots. The corresponding coherent quartet will propagate in the crystal without absorption. As before, the suppression effect supposes vanishing total amplitude for the formation of excited nuclei by the waves involved in the superposed quartet:

$$F = \sum_{s,h=1,2} F_h^{(s)}(m) = 0, \quad F_h^{(s)}(m) = E_h^{(s)}(m) (\mathbf{e}_h^{(s)} \cdot \mathbf{A}_h(\zeta_0, \zeta, p)). \quad (6.27)$$

Such an equality should be fulfilled for all transitions and all nuclei in the elementary cell with energies close to the energy of the incident radiation. Let us consider

eq. (6.27) as an equation for the four field components of a quartet m . Then it is clear that, if the number of transition lines does not exceed three, eqs. (6.27) can always be satisfied and a configuration for the fields of the quartet can be found which produces the suppression effect.

It is worthwhile to note that the fields $E_h^{(s)}$ derived from the solution of eqs. (6.27) are the solution of the initial system (5.20) for $\alpha = 0$ and $\varepsilon_0 = 0$. Really, provided the condition (6.27) is fulfilled, we can check that the right-hand side of eqs. (5.20) vanishes, taking into account eq. (5.21) and the multiplicative structure of the terms forming tensor (5.13).

The quartet structure of the independent superposed states introduces a large variety of HF structures and atomic configurations for which the suppression effect takes place. A detailed analysis of a number of concrete cases is given in [19,44]. Note that in some cases the suppression effect occurs for both polarizations, i.e., for the total incident radiation. In particular, the latter takes place if two transition lines are within the range of the resonant interaction.

It is interesting to note that a phenomenon that resembles the suppression effect, the so-called Borrmann effect [45], is known from X-ray physics. The Borrmann effect is displayed by decreasing the photoelectric absorption for Laue diffraction. However, both phenomena differ intrinsically from each other. The Borrmann effect requires the electric field to vanish at the lattice sites. As has been shown, for the suppression effect it is necessary that the amplitude for the formation of the excited nucleus vanishes, neither the electric nor the magnetic field of γ -quanta at the nucleus need to be equal to zero. For the Borrmann effect, there is always only a limited decrease of absorption even in the case of a rigid lattice. The temperature and vibrations of atoms restore the X-ray absorption. Naturally, the decay of intensity with transmission through a thick crystal remains exponential. In these aspects the suppression effect, demonstrating the total suppression of γ -quantum absorption, differs drastically from the Borrmann effect.

So far, we have considered dynamical diffraction for purely nuclear scattering. In fact, γ -quanta experience electronic scattering at the same time. Though this scattering is weak compared to the resonant nuclear scattering, its role in diffraction may be significant for a number of cases.

The coherent character of elastic electronic scattering, which is displayed, in particular, in the interference of nuclear and electronic scattering, allows us to incorporate the interaction with electrons by introducing additional terms to the coefficients in (5.13) of the dynamical system of equations (5.18)

$$\chi_{hh'}^{il} = -\delta^{il} \frac{4\pi r_0}{\kappa^2 v_0} \sum_p \chi_p(\mathbf{k}_h - \mathbf{k}_{h'}) f_p(\mathbf{k}_h - \mathbf{k}_{h'}) e^{i(\mathbf{k}_h - \mathbf{k}_{h'}) \cdot \boldsymbol{\rho}_p}. \quad (6.28)$$

Here $\chi_p(\mathbf{k})$ is the atomic structure factor involving a small imaginary part determined by the photoelectric absorption process (see, e.g., [46]). In the absence of nuclear scattering, the system of equations (5.18) with the coefficients in (6.28) has a superposition of either only σ - or π -polarized waves as solutions. Provided the conditions

are chosen so that eq. (6.6) is valid and the decoupled pairs of equations are determined for the σ - and π -polarizations, the electronic scattering does not *recover* the nuclear absorption. This does take place for the E1 and M1 transitions in the general case. Moreover, provided $e^{i\mathbf{K}\rho} = 1$ and for the same values of factors $f_p(\mathbf{k})$ for all resonant nuclei in the elementary cell, the suppression effect occurs in this case for one of the polarizations for any character of the HF splitting (see details in [44]). If one neglects the imaginary correction to $\chi_p(\mathbf{K})$ and makes the same assumptions, the suppression effect is conserved completely for some finite magnitude of α , i.e., for some deviation from the exact Bragg angle (see details in [7]). The absorption will appear only at a significant thickness, the magnitude of which is determined by the electronic absorption length.

7. Suppression effect of inelastic channels. Experimental results

The experimental study of coherent phenomena in the resonant interaction of γ -quanta with the Mössbauer nucleus ensemble was concentrated for a relatively long period on the discovery and investigation of the suppression effect. Here, a significant problem is to grow high quality single crystals enriched in the resonant isotope. A large amount of work has been performed and some of it will be described here, because of its key role in the development of this field.

The suppression effect has been established for the first time in the work of Voitovetskii et al. [22], while studying resonant Laue diffraction of γ -quanta in a thick metal tin single crystal enriched in ^{119}Sn to 88% ($(\mu_{\text{nucl}}t)_{\text{res}} \approx 640$). The authors have measured the intensity of the diffracted beam as a function of the incident γ -quantum energy and compared it to the intensity of the transmitted radiation if the crystal is off the Bragg position. A giant difference in the intensities was observed. The diffracted Laue beam has incomparably smaller absorption near resonance where the absorption is at a maximum in normal conditions. These results were a striking demonstration of the suppression effect, and agreed well with the theoretical calculations.

In order to disclose the suppression effect, an original idea has been realized in the work of Smirnov et al. [23]. The radiation incident onto the crystal has a resonant energy distribution determined by the Mössbauer source. Under the usual conditions with the penetration into the depth of a crystal, the closer to the resonance center the stronger the absorption that takes place. As a result, in the thick crystal the transmitted radiation loses its resonance structure, flattening within a noticeable energy range. The idea consists of measuring the energy distribution of the diffracted Laue beam as it leaves the crystal and comparing it with the energy distribution in the incident beam. An iron single crystal, enriched to 85% by the ^{57}Fe isotope, was used. For the first time the authors found a remarkable effect. The resonance structure is conserved in the diffracted beam whereas the radiation transmitted far from the Bragg direction has practically no dispersion structure. In other words, this is a direct demonstration that γ -quanta propagate almost without absorption under the conditions chosen according

to the theoretical prediction for a vanishing amplitude of formation of the excited nucleus.

The most precise measurements for discovering the suppression effect have been undertaken by van Bürck et al. [24]. Purely nuclear Bragg reflection is used in a perfect $^{57}\text{FeBO}_3$ single crystal. The possibility of having pure nuclear reflections in a crystal for reflections forbidden for electronic Bragg scattering is based on the spin dependence of the scattering amplitude (in full analogy with neutron scattering). It was first stated in [47] and confirmed experimentally in [48].

The antiferromagnetic structure of FeBO_3 provides such a possibility. The most significant moment of the work [24] is the use of an angle divergence of the primary beam equal to $4''$. This allowed the first measurement of the dependence of the Laue transmitted beam in pure nuclear diffraction on the deviation from the exact Bragg angle and its energy distribution after traversing the crystal. The measurements demonstrate that the beam has an anomalous transmission through a strongly absorbing thick crystal within a narrow range of scattering angles. In addition, the transmitted beam conserves the resonant energy distribution of the incident beam. Such a combination gives direct evidence for the suppression effect.

In [49] an attempt was made to test one of the most interesting predictions of the theory, namely, the independence of the suppression effect from the thermal vibrations of a crystal, while the Bragg condition was fulfilled. The residual resonant absorption and line shape are measured near the resonance center with pure nuclear scattering in a $^{57}\text{FeBO}_3$ single crystal. The high sensitivity in this region to a small violation of the strict suppression allowed the authors to conclude that thermal vibrations do not disturb the suppression effect, with a relatively high accuracy.

A special study of the suppression effect in strongly anisotropic Bragg scattering, when the amplitude of the diffracted wave is small compared with that of the transmitted wave, has been performed in [50]. The authors have proved that, in the absence of any effects requiring the equality of both amplitudes, e.g., the Borrmann effect, the suppression effect takes place due to the vanishing amplitude of formation of the excited nucleus in this anomalous configuration of the electromagnetic field.

To conclude this section, let us note that the theory [16,17] for suppressing the inelastic channels of the reaction with the resonant nuclear scattering of neutrons has been developed in parallel with the theory of the suppression effect for γ -quanta. The effect has been observed experimentally in the measurements [51,52] of Shil'shtein et al. studying the Laue diffraction of neutrons with an energy close to the known $\lambda_0 \approx 0.67 \text{ \AA}$ cadmium resonance in a strongly absorbing CdS crystal.

8. Resonant Bragg scattering of γ -quanta in crystals

In the previous sections we have considered resonant nuclear diffraction, assuming Laue geometry and the corresponding picture of transmission of γ -quanta through a crystal. In this section we will treat the reflection of resonant γ -quanta in a thick

crystal for the case of a Bragg geometry, still assuming that the diffraction is practically determined by a single vector \mathbf{K} of the reciprocal lattice.

The motion of γ -quanta in a crystal is described, obviously, by the same system of dynamical equations (5.20). Only the boundary conditions change, taking the following form for a lamina-shaped crystal of thickness l :

$$\sum_m E_0^{(s)}(m) = \mathcal{E}_0^{(s)}, \quad \sum_m E_1^{(s)}(m) \exp\left(i \frac{\kappa}{\gamma_0} \varepsilon_{0s}^{(m)} l\right) = 0. \quad (8.1)$$

The second boundary condition corresponds to the fact that the diffracted radiation emerges from the crystal on the entrance side.

Let us restrict ourselves by analyzing the cases for which condition (6.6) is valid. Then, for each $s = 1, 2$ we have a root $\varepsilon_{0s}^{(1,2)}$ (eq. (6.8)) provided $\beta < 0$. For the electric field in the crystal, we have for the general case, involving eq. (8.1),

$$\mathbf{E}^{(s)}(\mathbf{r}) = \mathcal{E}_0^{(s)} [\mathbf{e}_0^{(s)} \cdot E_0^{(s)}(\mathbf{r}) + \mathbf{e}_1^{(s)} \cdot \mathbf{e}^{i\mathbf{K}\mathbf{r}} \cdot E_1^{(s)}(\mathbf{r})], \quad (8.2)$$

$$\begin{aligned} E_0^{(s)}(\mathbf{r}) &= \frac{1}{L} [(2\varepsilon_{0s}^{(1)} - g_{00}^{(s)}) \exp(iz_s^{(2)}x) \\ &\quad - (2\varepsilon_{0s}^{(2)} - g_{00}^{(s)}) \exp(iz_s^{(1)}x) e^{i(z_s^{(2)} - z_s^{(1)})l}], \\ E_1^{(s)}(\mathbf{r}) &= -\frac{1}{L} \beta g_{10}^{(s)} [\exp(iz_s^{(2)}x) - \exp(iz_s^{(1)}x) e^{i(z_s^{(2)} - z_s^{(1)})l}], \end{aligned} \quad (8.3)$$

where

$$L = (2\varepsilon_{0s}^{(1)} - g_{00}^{(s)}) - (2\varepsilon_{0s}^{(2)} - g_{00}^{(s)}) e^{i(z_s^{(2)} - z_s^{(1)})l}, \quad z_s^{(1,2)} = \frac{\kappa}{\gamma_0} \varepsilon_{0s}^{(1,2)}. \quad (8.4)$$

As before, $x = \mathbf{n} \cdot \mathbf{r}$. Expressions (8.2) and (8.3) allow us to determine readily the intensities of the reflected $P_1^{(s)}$ and transmitted $P_0^{(s)}$ γ -quantum beams:

$$\frac{P_1^{(s)}}{I_0^{(s)}} = |\beta| |g_{10}^{(s)}|^2 \frac{1}{|L|^2} |1 - e^{i(z_s^{(2)} - z_s^{(1)})l}|^2, \quad (8.5)$$

$$\frac{P_0^{(s)}}{I_0^{(s)}} = 4 \frac{1}{|L|^2} |(\varepsilon_{0s}^{(1)} - \varepsilon_{0s}^{(2)}) \exp(iz_s^{(2)}l)|^2. \quad (8.6)$$

Expressions (8.5) and (8.6) are valid for a crystal of arbitrary thickness. Let us consider the case of a thick crystal when the condition

$$\text{Im}(z_s^{(2)} - z_s^{(1)})l = \frac{\kappa l}{\gamma_0} \text{Im}(\varepsilon_{0s}^{(2)} - \varepsilon_{0s}^{(1)}) \gg 1 \quad (8.7)$$

is fulfilled.

Hereafter we suppose that the upper index 2 refers to the root with the larger imaginary part. Thus, in a sufficiently thick crystal the intensity of the transmitted beam practically vanishes. Using the values of roots $\varepsilon_{0s}^{(1,2)}$ (eq. (6.8)) found before

and taking the symmetrical reflection $\beta = -1$ with the usual relation $g_{11}^{(s)} = g_{00}^{(s)}$ for simplification of the analysis, we find for the reflected intensity

$$\frac{P_1^{(s)}}{I_0^{(s)}} = \frac{4|g_{10}^s|^2}{|(\alpha - 2g_{00}^{(s)}) \pm [(\alpha - 2g_{00}^{(s)})^2 - 4g_{01}^s g_{10}^s]^{1/2}|^2}. \quad (8.8)$$

The sign in front of the root should be chosen so that the imaginary part of the root will be negative.

If the relation between the amplitudes is such that condition (6.16) is valid, total reflection takes place for $\alpha = 0$:

$$\frac{P_1^{(s)}}{I_0^{(s)}} = 1. \quad (8.9)$$

Provided the interaction with electrons is involved and the most interesting cases of purely nuclear scattering are considered, the components of tensor (6.28) differ from zero only for $h = h'$. Thus,

$$g_{hh}^{(s)} = (g_{hh}^{(s)})_{\text{nucl}} + \chi_0, \quad (8.10)$$

where according to eq. (6.28)

$$\chi_0 = -\frac{4\pi r_0}{\kappa^2 v_0} \sum_p \chi_p(0). \quad (8.11)$$

Neglecting electronic absorption, we again obtain total reflection but for an angle

$$\alpha_0 = 2\chi_0 \quad (8.12)$$

different from $\alpha = 0$.

Let us turn to a more general case when scattering by electrons is allowed and the reflected vectors of \mathbf{K} are chosen so that the imaginary part of tensor (6.28) vanishes. In addition, let the relations

$$g_{01}^{(s)} = g_{10}^{(s)}, \quad \text{Im } g_{10}^{(s)} = \text{Im } g_{00}^{(s)} \quad (8.13)$$

be valid, which are typical for a large number of cases, especially, for the E1 and M1 transitions. The direct analysis of eq. (8.8) again results in eq. (8.9), now at the angle

$$\alpha_0 = 2(\text{Re } g_{00}^{(s)} - \text{Re } g_{10}^{(s)}). \quad (8.14)$$

Note that the result obtained occurs for an arbitrary relation between the amplitudes of electronic and nuclear scattering.

The total reflection of the resonant Bragg scattering in a strongly absorbing crystal is completely associated with the inelastic channel suppression effect. In contrast to γ -quantum transmission in a Laue geometry when a certain coherent superposition of waves conserves the field amplitudes, it follows from eq. (8.3) that the field components decrease strongly deep in the crystal. However, the relation between $E_0^{(s)}$ and

$E_1^{(s)}$ is conserved at an arbitrary depth and the amplitude of the formation of an excited nucleus vanishes (cf. eq. (8.3) for $\beta = -1$ and $g_{00} = g_{11}$ with eq. (6.20) and eq. (6.21)).

It is interesting that in the case of Laue diffraction one of the superposed states, on the contrary, is absorbed strongly. Thus, in general only a part of the incident radiation is transmitted anomalously through the thick crystal. Only for special cases can the suppression effect be realized for both pairs of superposed states. In the case of Bragg geometry, at least for a symmetrical reflection, the suppression effect can be realized for the entire radiation, resulting in total reflection. Note that the picture appearing in Bragg reflection is determined to a great extent by the coherent collective character of the excitation of the nuclear system.

Analyzing a purely nuclear reflection, we find from eqs. (8.8) and (8.12) that the intensity $P_1^{(s)}(\alpha_0)$ does not depend on the deviation from resonance, all $g_{hh'}^{(s)}$ having the same resonance denominator. From a physical point of view, this is due to the fact that the reduction of the nuclear scattering amplitude is compensated by the number of crystal planes involved in reflection. The self-consistency is provided by the reduction of the decay of the field components of eq. (8.3) with x since $\text{Im} \varepsilon_{0s}^{(r)}$ falls as $g_{hh'}^{(s)}$ decreases.

Even a relatively weak violation of the suppression effect results in a very strong reduction of the reflection intensity. A characteristic parameter of such a violation is the deviation of the following relation from unity:

$$p^{(s)} = \frac{\text{Im} g_{10}^{(s)}}{\text{Im} g_{00}^{(s)}}. \quad (8.15)$$

This can obviously be seen from figure 1, where, neglecting electronic scattering, the $P_1^{(s)}/I_0^{(s)}$ vs. α/g_0 dependence is given for three values of $p^{(s)}$ at $\omega = \omega_0$. Here ω_0 is the resonant-transition frequency and $g_0 = |g_{00}|$ ($\omega = \omega_0$). From figure 1 one can also see the nontrivial peak-like behaviour of the angular dependence, demonstrating a drastic reduction of $P_1^{(s)}$ with increasing $|\alpha|$. Such behaviour is a specific feature of resonant nuclear Bragg diffraction. The analytical dependence for $\alpha/g_0 \ll 1$ and $p^{(s)} = 1$ is

$$\frac{P_1^{(s)}}{I_0^{(s)}} = 1 - \sqrt{2|\alpha|/g_0}. \quad (8.16)$$

If $\omega \neq \omega_0$, a strong asymmetry appears in the $P_1^{(s)}(\alpha)$ -dependence.

It should be noted that the case of the symmetrical $\beta = -1$ reflection is singled out. Thus, for $g_{11}^{(s)} = g_{00}^{(s)}$ eq. (6.8) for the roots has the simple form

$$\varepsilon_{0s}^{(1,2)} = \frac{1}{4} \left\{ \alpha \pm [\alpha^2 - 4(g_{00}^{(s)}\alpha - \Delta^{(s)})]^{1/2} \right\}.$$

Provided eq. (6.16) is valid, i.e., $\Delta^{(s)} = 0$, we have for $|\alpha| \ll |g_{00}^{(s)}|$

$$\text{Im} \varepsilon_{0s}^{(1,2)} \sim \alpha^{1/2}.$$

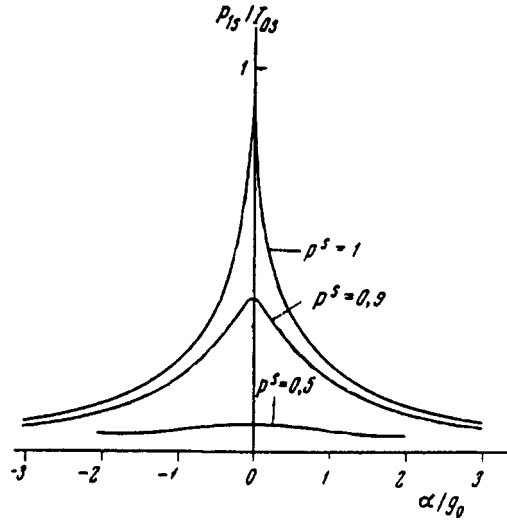


Figure 1. (From [18].)

This means that, as α decreases, the field penetrates into larger depths $x_\alpha \sim \alpha^{-1/2}$ and a larger number of nuclei scatters with conservation of phase coherence. It is easy to show that the amplitude of formation of an excited nucleus in the total field eq. (8.2) is proportional to $\alpha^{1/2}$. This compensation leads to total reflection as $\alpha \rightarrow 0$. The probability of incoherent processes, which occur independently for each of the nuclei in a layer of about x_α , will be proportional to α . Hence, the integral intensity of the incoherent processes is proportional to $x_\alpha \alpha \sim \alpha^{1/2}$, resulting in eq. (8.16).

In figure 2 the integral intensity $R^{(s)}$ over all reflection angles is plotted as a function of the dimensionless parameter $V = 2\hbar(\omega - \omega_0)/\Gamma$ for various values of $p^{(s)}$. The intensity $R^{(s)}$ is obtained by integrating distribution equation (8.8) with the assumption $g_{01}^{(s)} = g_{10}^{(s)}$ and using eq. (6.5)

$$R^{(s)} = \int \frac{P_1^{(s)}}{I_0^{(s)}} \frac{d\alpha}{2 \sin 2\Theta_B}.$$

The curves in figure 2 demonstrate the strong broadening of the $R^{(s)}(V)$ dependence. This is a specific feature of the reflection from a thick crystal where the reduction of the scattering amplitude for a single nucleus with increasing V is partially compensated by the reflection from a larger depth. For $|V| \gg 1$,

$$R^{(s)} \sim \frac{1}{|V|},$$

whereas for the thin crystal $R^{(s)} \sim |V|^{-2}$ as for a single nucleus.

The description of a series of other specific features for resonant Bragg reflection, in particular associated with the interference of nuclear and electronic scattering, can be found in [18]. Note only that all initial results of eqs. (8.2)–(8.6) are valid for crystals

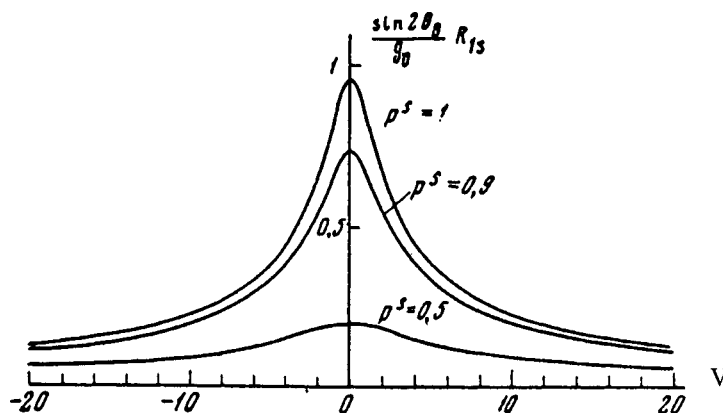


Figure 2. (From [18].)

of arbitrary thickness l , in particular for thin crystals. In the latter case we refer to the work of Hannon and Trammell [21] and also the early work of Trammell [4] devoted specifically to nuclear Bragg scattering by a thin crystal.

A great number of papers has been devoted to the experimental study of resonant nuclear scattering in Bragg geometry. The investigations have been performed over the whole period of studying coherent phenomena, using stationary resonant γ -quantum sources. However, real success in the study of the detailed features inherent in resonant nuclear reflection has been achieved only when measurements [25,53–55] started with an angular collimation of some seconds in combination with high time stability. Here it is significant that the application of nearly perfect antiferromagnetic $^{57}\text{FeBO}_3$ single crystals allows the study of purely nuclear reflections.

The dependence of the reflection intensity on the deviation from the Bragg angle at a strictly resonant energy of incident radiation was measured for the first time in [25,54]. The peculiar shape of curves observed in these studies, namely, a sharp peak at the Bragg position and broad wings, corresponds to the predictions of the theory. It is essential that the measurements have been performed under the suppression effect, i.e., when the ideal curve has a shape as $p^{(s)} = 1$ in figure 1. In these cases and in [55], an anomalously wide energy dependence of the reflection intensity has been observed for a beam of γ -quanta incident at the Bragg angle with a narrow divergence. The width is 20–30 natural linewidths.

The work in [54] demonstrates the increase of linewidth when the angular divergence of the incident beam is brought down to $1''$. If there is a certain deviation from the exact Bragg angle, the broadening is reduced [25]. All these results are in good agreement with the predictions of the dynamical theory for a thick crystal. In particular, the last result has a clear physical nature. The reduction of the scattering amplitude of a single nucleus is now compensated significantly less with the deviation from resonance, since coherent scattering in the phase for $\alpha \neq 0$ occurs at a limited depth. The larger the parameter α , the smaller the broadening should be.

Special effects were studied in order to measure the absolute value of the reflection intensity. Due to weak mosaicity of the crystal and finite divergence of the incident beam total reflection is not achieved. The measurements have demonstrated a very large reflection: the ratio $P_1^{(s)}/I_0$ reaches 35% in [53] and exceeds 40% in [54].

9. Anomalous small-angle diffusive scattering in crystals

As is known, X-ray Bragg scattering in crystals is accompanied by anomalous diffusive scattering within a narrow range of angles near the diffraction maximum. Here the differential cross-section of scattering behaves as $1/q^2$, where the vector $\mathbf{q} = \mathbf{k}' - \mathbf{k}_1 = \mathbf{k}' - (\mathbf{k} + \mathbf{K})$ corresponds to a small deviation of a diffracted X-ray quantum from \mathbf{k}_1 (see, e.g., [56]). From the physical point of view this result is a consequence of the fact that, due to the periodicity of the phonon spectrum in the reciprocal lattice space for a regular crystal, it is always possible in Bragg scattering to excite phonons of extremely small frequencies with a transfer of noticeable momentum $\hbar\mathbf{K}$.

Though the excitation of low frequency phonons accompanying small-angle scattering takes place, the proper cross-section is proportional to q^2 and anomalous scattering is absent. This is a common result for the scattering of any particles.

However, the picture changes drastically for resonant nuclear scattering of Mössbauer γ -quanta. As we have already discussed, for $\Gamma \ll \Theta_{\text{ph}}$ the nucleus acquires a momentum $\hbar\mathbf{k}$ when absorbing a γ -quantum and a recoil momentum of $(-\hbar\mathbf{k}')$, uncorrelated with $\hbar\mathbf{k}$, during γ -quantum emission. Hence, for resonant nuclear scattering a unique phenomenon, namely, anomalous small-angle diffusive scattering, appears. This phenomenon was predicted and analyzed in [57].

The amplitude of coherent resonant γ -quantum scattering, when emitting or absorbing a phonon of wave vector \mathbf{q} and number α of the branch can be represented as

$$f_{\mathbf{q}\alpha}(\mathbf{k}, \mathbf{k}') \cong -\frac{\Gamma'_{\text{el}}}{2k} \sum_m \sum_{\{n^s\}} e^{i(\mathbf{k}-\mathbf{k}')\mathbf{r}_m} \times \frac{(e^{i\mathbf{k}\mathbf{u}_m})_{\{n\}\{n^s\}}(e^{-i\mathbf{k}'\mathbf{u}_m})_{\{n^s\}\{n'\}}}{\hbar\omega_k - E_0 + \sum_{\beta} \hbar\omega_{\beta}(n_{\beta}^s - n_{\beta}) + i(\Gamma/2)}. \quad (9.1)$$

Here the state $\{n'\}$ differs from $\{n\}$ only by $n'_{\mathbf{q}\alpha} = n_{\mathbf{q}\alpha} \pm 1$ for a single phonon.

In the case of purely elastic scattering, which we have considered so far, one has $\{n'\} = \{n\}$ and due to $\Gamma \ll \Theta_{\text{ph}}$ one can retain only one term $\{n^s\} = \{n\}$ in a sum. (See transformation from eqs. (5.7)–(5.10).) Taking into account the relation between g_{00} and the amplitude of elastic scattering at zero angle (eq. (6.13)) and involving the explicit form of g_{00} (eq. (6.15)), we have

$$\Gamma'_{\text{el}} = \Gamma_{\text{el}}\xi.$$

Note that $\xi = (2I + 1)/(2(2I_0 + 1))$ for the unresolved HF structure. In eq. (9.1) we neglected the coefficient that is equal to unity when the distinction between \mathbf{k} and \mathbf{k}' is small.

In the sum over $\{n^s\}$ in eq. (9.1) the terms with $\{n^s\} = \{n\}$ and $\{n^s\} = \{n'\}$ prove to be essential. The other terms give a small contribution of about $\Gamma/\Theta_{\text{ph}}$ to the final result. The diagonal term in the nominator gives $f(\mathbf{k})$ (eq. (5.8)) and a nondiagonal term gives a factor $f(\mathbf{k})$ multiplied by the matrix element of a single-phonon transition. As a result, we find after a simple transformation

$$f_{\mathbf{q}\alpha}(\mathbf{k}, \mathbf{k}') = \frac{i\Gamma'_{\text{el}}}{2k} f(\mathbf{k}) f(\mathbf{k}') \frac{(\bar{n}_{\mathbf{q},\alpha} + 1/2 \pm 1/2)^{1/2}}{(2MN\omega(\mathbf{q},\alpha))^{1/2}} \left\{ \frac{\mathbf{k}' \cdot \mathbf{e}(\mathbf{q},\alpha)}{\hbar\omega_k - E_0 + i\Gamma/2} - \frac{\mathbf{k}\mathbf{e}(\mathbf{q},\alpha)}{\hbar\omega_k - E_0 \mp \hbar\omega(\mathbf{q},\alpha) + i\Gamma/2} \right\} \sum_m e^{i(\mathbf{k}-\mathbf{k}'\mp\mathbf{q})\mathbf{r}_m}. \quad (9.2)$$

Here $\mathbf{e}(\mathbf{q},\alpha)$ and $\bar{n}_{\mathbf{q},\alpha}$ are the phonon polarization vector and average occupation numbers, respectively. To simplify, we write expression (9.2) with the assumption of a one-atom crystal, N being the number of atoms of mass M in the crystal.

It is interesting that the first term in the brackets in eq. (9.2) remains purely resonant, and no phonon frequencies appear in the denominator. The term corresponds to the emission or absorption of a phonon only at the stage of the γ -decay of the excited nucleus.

It should be emphasized that eq. (9.2), at least within the accuracy of an insignificant factor, is valid in the general case for the excitation of arbitrary phonons.

The lattice sum in eq. (9.2) results in the relation $\mathbf{k}' = \mathbf{k} \mp \mathbf{q}$. For small-angle scattering, where the vector \mathbf{q} is small, the vector \mathbf{k}' in the first term in the brackets and in $f(\mathbf{k}')$ can be replaced by \mathbf{k} . In addition, we assume $k_{\text{B}}T \gg \hbar\omega(\mathbf{q},\alpha)$ with temperature T and Boltzmann factor k_{B} . Then, going from the scattering amplitude to the differential cross-section per nucleus, we find

$$\frac{d\sigma}{d\Omega_{\mathbf{k}'}} = \frac{\eta}{4\pi} \sigma_t(\omega_k) \frac{\Gamma'_{\text{el}}}{\Gamma} f^2(\mathbf{k}) \frac{T}{2M} \sum_{\pm} \sum_{\alpha} \frac{|\mathbf{k}\mathbf{e}(\mathbf{q},\alpha)|^2}{(\hbar\omega_k - E_0 \mp \hbar\omega(\mathbf{q},\alpha))^2 + \Gamma^2/4}, \quad (9.3)$$

where σ_t is the total cross-section of scattering by a fixed nucleus (eq. (6.14)).

For phonons with small q , we have a sound spectrum $\omega(\mathbf{q},\alpha) = c_{\alpha}(\mathbf{q}/q)q$. For really achievable small angles, it is obvious that $\hbar\omega(\mathbf{q},\alpha) \gg |\hbar\omega_k - E_0|, \Gamma$. As we see, for the resonant nuclear interaction anomalous small-angle diffusive scattering takes place and

$$\frac{d\sigma}{d\Omega_{\mathbf{k}'}} \sim \frac{1}{q^2}. \quad (9.4)$$

It follows from eq. (9.3) that the differential cross-section is large. It is interesting that coherent one-phonon scattering, which does not keep a tag in the nuclear system, leads to anomalous scattering. Coherence is displayed in the law of momentum conservation in eq. (9.2), which determines the whole picture.

Let us transform eq. (9.3) into the total cross-section, using $d\Omega_{\mathbf{k}'} = d^2q/k^2$ due to the large energy of γ -quanta. Then

$$\sigma = \sigma_t \frac{\Gamma'_{\text{el}}}{\Gamma} f^2(\mathbf{k}) \frac{T}{4M} \sum_{\alpha} \int \frac{d\varphi}{2\pi} \frac{|\mathbf{n}\mathbf{e}(\varphi, \alpha)|^2}{c_{\alpha}^2(\varphi)} \left\{ \ln \frac{\hbar^2 c_{\alpha}^2(\varphi) q_0^2}{\Delta^2 + \Gamma^2/4} + \frac{4\Delta}{\Gamma} \arctan \frac{2\Delta}{\Gamma} \right\}. \quad (9.5)$$

Here $\Delta = \hbar\omega_k - E_0$ and q_0 is the wave vector of the order of the limiting phonon vector and $\mathbf{n} = \mathbf{k}/k$. The integration in eq. (9.5) over angle φ is performed in the plane normal to \mathbf{k} . The large logarithm of about $\log(\Theta_{\text{ph}}^2/\Gamma^2)$ in eq. (9.5) strongly enhances the integral cross-section. The use of the Mössbauer effect allows us to separate inelastically scattered small-angle emission from purely resonant emission, transmitted without scattering. For this purpose, it is sufficient to measure the reduction of the integral intensity in the resonant absorber as a function of its velocity. This discloses an effective way for measuring the integral cross-section of anomalous diffusive forward scattering.

Note that all results are referred to the thin crystal and the total intensity of scattering should be determined by the product of eqs. (9.3)–(9.5) and the number of resonant nuclei. Since $\hbar\omega(\mathbf{q}, \alpha) \gg \Gamma$, the thickness of a crystal is limited by the electronic absorption. This means that the size limit is increased by at least two orders of magnitude, and so is the achievable limit of the intensity of scattering.

10. Nuclear resonant scattering of synchrotron radiation

The observation of the direct excitation of nuclei by SR has opened a completely new area of studying coherent phenomena with resonant nuclear interactions in matter. The solution of the problem which is paradoxical at first sight, namely, the isolation of the resonant line of about 10^{-8} eV width from the continuous radiation spectrum of a width of some 10 keV, is associated to a great extent with the features of Mössbauer transitions in nuclei. For these transitions, a large lifetime τ_0 of the excited states is typical. At the same time the duration T of a single SR pulse is much smaller than τ_0 and the time interval T_1 between the pulses is larger or of the same order of magnitude as τ_0 . This gives a principal opportunity to measure the delayed emission associated with the decay of the excited nuclei after the SR pulse. The potential scattering of almost all incident radiation occurs in fact instantaneously. This idea, formulated by Ruby [30], was realized experimentally in a complete form by Gerdau et al. [32] who started a wide field of research.

The isolation of retarded nuclear emission with a fixed instant of excitation enabled the unique possibility of introducing the time dimension into the study of coherent phenomena. On the other hand, the use of coherent nuclear scattering should allow us to realize the conditions for producing a pulsed source of Mössbauer radiation. It is worthwhile to emphasize that the properties of SR are very suitable for both problems, in particular the directional radiation, its small angular divergence in the plane normal to the plane of the electronic orbit, and the pure polarization state.

The solution of all the questions associated with SR-crystal interaction supposes the development of a time-dependent theory of resonant nuclear scattering. For this purpose, the results of the stationary theory of resonant nuclear diffraction, which have been given in the preceding sections, can be used effectively. A theory for the time domain was developed for the first time in [33] which we rely on in our further considerations. To make all results clear, we restrict ourselves to the case of an unsplit HF structure. As has been noted, in the presence of HF splitting, quantum beats associated with the virtual excitation of several HF transitions at the same time appear in the time structure of the scattered radiation. The theory of quantum beats has been developed in [39,40] and interpreted in other reviews.

1. Let radiation with a frequency range $\Delta\tilde{\omega}$ run through the crystal during time T . Then a single component of the field can be represented as

$$E_{\tilde{\omega}}(t) = \mathcal{E}_{\tilde{\omega}} e^{-i\tilde{\omega}t} \Psi(t), \quad (10.1)$$

where $\Psi(t)$ is a function describing the shape of the radiation pulse.

Let us introduce the amplitude $R(\omega)$, relating the amplitudes of the scattered $E'(\omega)$ and incident $E(\omega)$ waves:

$$E'(\omega) = R(\omega)E(\omega). \quad (10.2)$$

Finding the Fourier component of field (10.1) and using definition (10.2) for a time-dependent wave field, we have

$$E'_{\tilde{\omega}}(t) = \mathcal{E}_{\tilde{\omega}} \int dt' G(t-t') e^{-i\tilde{\omega}t'} \Psi(t'), \quad (10.3)$$

where

$$G(t) = \int \frac{d\omega}{2\pi} R(\omega) e^{-i\omega t}. \quad (10.4)$$

The function $G(t)$ differs from zero only for $t > 0$, corresponding to the analyticity of the function $R(\omega)$ in the upper half-plane with its continuation into the complex plane.

Let us calculate the intensity $I(t)$ of the scattered radiation, involving the inequality $\Delta\tilde{\omega}T \gg 1$:

$$I(t) = 2\pi \frac{I_0}{\Delta\tilde{\omega}} \int_0^T dt' |G(t-t')|^2 \approx 2\pi \frac{I_0 T}{\Delta\tilde{\omega}} |G(t)|^2. \quad (10.5)$$

The latter equality in eq. (10.5) assumes that the intensity is determined for $t > T$, $I_0 T$ being the total number of quanta in the pulse.

Thus, for solving time problems, it is necessary to know the function $R(\omega)$. For the cases of interest to us, this function is determined in the preceding sections.

2. Let us start by analyzing the transmission of radiation across a crystal of an arbitrary thickness l far from the Bragg condition. Let the resonant frequency of $\omega_0 = E_0/\hbar$ be within the range $\Delta\tilde{\omega}$. Then, according to eqs. (10.1), (10.2) and (6.12)

$$R^T(\omega) = \exp(i\kappa g_{00}l/2\gamma_0). \quad (10.6)$$

Hereafter we omit the index s . It is convenient to rewrite the coefficient g_{00} (eq. (6.15)) as

$$g_{00} = -\frac{g_0}{V+i} + \chi_0, \quad V = \frac{2\hbar(\omega - \omega_0)}{\Gamma}. \quad (10.7)$$

Here,

$$g_0 = \frac{1}{\kappa} \sigma_{\text{res}} n f^2(\mathbf{k}) \quad (10.8)$$

and σ_{res} is the magnitude of the total cross-section (eq. (6.14)) at $\omega = \omega_0$. In eq. (10.7) electronic scattering is involved as determined by eq. (8.11) according to eq. (8.10).

Let us substitute eq. (10.6) into eq. (10.4). The corresponding integration can be performed explicitly (see details in [33]). Eventually, we find

$$G^T(t) = e^{i\varphi} \left\{ \delta(t) - \exp\left(-i\omega_0 t - \frac{\tau}{2}\right) \frac{\xi}{\tau_0} \cdot \frac{J_1(2\sqrt{\xi\tau})}{\sqrt{\xi\tau}} \Theta(t) \right\}, \quad (10.9)$$

where

$$\xi = \frac{g_0 \kappa l}{4\gamma_0}, \quad \varphi = \frac{\chi_0 \kappa l}{2\gamma_0}, \quad \tau = \frac{t}{\tau_0}, \quad \tau_0 = \frac{\hbar}{\Gamma}. \quad (10.10)$$

Here J_1 is the Bessel function of first order.

The first term in the brackets of eq. (10.9) corresponds to a practically instant transmission of the nonresonant part of the incident radiation. In the course of the calculation the quantity $\delta(t=0)$ should be understood as $\Delta\tilde{\omega}/(2\pi)$.

The second term in the brackets of eq. (10.9) describes the retarded emission of the radiation resonantly scattered forward. Naturally, the emission is determined for $t > 0$, entailing the appearance of the usual step-like function $\Theta(t)$. In a thin crystal, for $\xi \ll 1$ the ratio $J_1(2\sqrt{\xi\tau})/\sqrt{\xi\tau}$ is close to unity for $t \lesssim \tau_0$. The intensity (eq. (10.5)) in this case has a typical $\sim l^2$ behaviour. One may estimate that there is a certain acceleration of the decay as a result of the collective excitation in the nuclear system. The picture changes significantly with the transition to a thick crystal. For $\xi\tau > 1$, the oscillations of the intensity in time, determined by the behaviour of the Bessel function, become pronounced. For $\xi\tau \gg 1$, expression (10.9) approaches the asymptotic limit

$$G^T(t) \approx \exp\left(i\varphi - i\omega_0 t - \frac{\tau}{2}\right) \frac{\xi^{1/4}}{\tau_0 \tau^{3/4} \pi^{1/2}} \cos\left(2\sqrt{\xi\tau} - \frac{3}{4}\pi\right). \quad (10.11)$$

The specific time behaviour is connected completely with the coherent collective interaction of resonant photons with the nuclear system. The conservation and even weak

growth of the intensity with the thickness is associated with the role of the frequency wings of the resonance in a thick crystal. Note that the inequality $\xi \ll \Delta\tilde{\omega}\tau_0$ is implicitly assumed to be valid. At the same time, the number of quanta within the energy range of about Γ around the resonance decreases with increasing τ due to absorption.

The time evolution of SR nuclear forward scattering has been observed for the first time in Brookhaven [36,37]. This became possible when the authors drastically reduced the energy width $\Delta\tilde{\omega}$ of the radiation incident on the sample. Practically all specific features predicted by theory for the time domain behaviour of the transmitted radiation in samples of various thicknesses have been observed by these experiments. Good agreement with theory is shown in [38] where analogous measurements have been performed very precisely. In the last study a well-resolved structure of quantum beats is observed from two transitions as a fine structure in the Bessel modulation.

3. Let us consider now the time problem for SR Bragg scattering, restricting ourselves to an isolated nuclear resonance. We again suppose purely nuclear reflection corresponding to polarization of the incident SR. In order to determine the reflection amplitude $R(\omega)$, one can employ the results of section 8. Involving eqs. (8.2)–(8.4), we readily obtain

$$R^B(\omega) = -\beta g_{10} \frac{1}{L} (1 - e^{i(z^{(2)} - z^{(1)})l}). \quad (10.12)$$

Let us introduce notation analogous to eq. (10.7):

$$g_{10} = -\frac{\tilde{g}}{V+i}, \quad g_{01} = -\frac{\tilde{g}^*}{V+i}. \quad (10.13)$$

Transforming expression (10.12) and using the explicit form of L (eq. (8.4)) and roots $\varepsilon_0^{(1,2)}$ (eq. (6.8)) for the case of symmetrical $\beta = -1$ reflection, we find for a thick crystal when the inequality $\text{Im}(z^{(2)} - z^{(1)})l \gg 1$ allows us to neglect the second term in brackets in eq. (10.12),

$$R^B(\omega) = -\frac{\tilde{g}}{g} \frac{s}{\tau_0} \frac{1}{\omega - z_0 + [(\omega - z_0)^2 - (s/\tau_0)^2]^{1/2}}. \quad (10.14)$$

Here

$$s = \frac{g}{\alpha - 2\chi_0}, \quad z_0 = \omega_0 - \frac{1}{\tau_0} \left(\frac{i}{2} + \frac{s}{p} \right), \quad p = \frac{2g}{g_0 + g_1}, \quad g = |\tilde{g}|. \quad (10.15)$$

We have used g_{11} written in the form (10.7), replacing g_0 by g_1 with the same definition (10.8) for the substitution of $\mathbf{k} \rightarrow \mathbf{k}_1 = \mathbf{k} + \mathbf{K}$. Note that in the general case χ_0 and s are complex quantities.

The factor p coincides with that of eq. (8.15) provided $g_1 = g_0$. Note that $p \leq 1$ and, as we have seen, the condition $p = 1$ is necessary to realize the inelastic channel suppression effect.

Let us substitute eq. (10.14) into eq. (10.5). Putting the integration into the complex ω plane and keeping in mind that the singularities of the function $R(\omega)$ lie in the lower half-plane, we can derive an analytical expression for $G^B(t)$ (see details in [33]):

$$G^B(t) = i \frac{\tilde{g}}{g} \frac{1}{\tau_0} \exp\left(-i\omega_0 t - \frac{\tau}{2}\right) \frac{J_1(s\tau)}{\tau} \cdot e^{is\tau/p} \Theta(t). \quad (10.16)$$

Here J_1 is again the Bessel function of first order, however, now of a complex argument. For several transitions allowed for the given HF structure and fixed polarization of incident radiation, the function $G^B(t)$ contains a superposition of field components of the scattered radiation with the same structure as eq. (10.16) but, naturally, with different frequencies ω_0 . In this case the reflection intensity exhibits time oscillations or quantum beats. To analyze the physical picture, it is sufficient to consider the behaviour of the intensity related to a single transition.

At time $t \gg T$, using (10.16), we find

$$I_\alpha^B = \frac{2\pi}{\tau_0} \frac{N_\alpha}{\Delta\tilde{\omega}\tau_0} e^{-\tau} \left| \frac{J_1(s\tau)}{\tau} \right|^2 \exp\left(-\frac{2s''\tau}{p}\right). \quad (10.17)$$

Hereafter $s = s' + is''$ and the same for the other complex quantities. In eq. (10.17) we introduced the index α characterizing the angle of incidence of the primary beam according to eq. (6.4). The quantity N_α is the total number of quanta incident on the crystal at this angle per pulse.

Note that an expression analogous to eq. (10.17) has been found later in the course of the analysis of grazing-incidence reflection [58].

From the expression obtained we can readily conclude that there are two typical times satisfying conditions

$$|s|\tau_1 = 1, \quad s''\tau_2 = 1.$$

It follows from eq. (10.15) that

$$\tau_1 = \frac{t_1}{\tau_0} = (y^2 + y_0^2)^{1/2}, \quad \tau_2 = \frac{t_2}{\tau_0} = \frac{y^2 + y_0^2}{y_0}, \quad (10.18)$$

where

$$y = \frac{\alpha - 2\chi'_0}{g}, \quad y_0 = \frac{2\chi''_0}{g}. \quad (10.19)$$

For a crystal enriched in the resonant isotope, we have $y_0 \ll 1$ and time $t_2 \gg \tau_0$ associated with electronic absorption. The time t_1 is practically always small compared with t_2 , except for an extremely small deviation of $|y| \lesssim 2y_0$ from the Bragg angle.

Near the Bragg angle, when $\alpha < g$, the time t_1 is small compared with τ_0 . However, for times $t_2 > t > t_1$ the time behaviour of the reflected intensity takes a very specific form:

$$I_\alpha^\beta(t) \approx \frac{4}{\tau_0} \frac{N_\alpha}{(\Delta\tilde{\omega}\tau_0)} |y| \frac{e^{-\tau}}{\tau^3} \cos^2 \left(\frac{\tau}{|y|} - \frac{3\pi}{4} \right). \quad (10.20)$$

The collective character of excitation in the nuclear system accompanying the absorption of a single photon results in a drastic change of the decay law. The decay process accelerates: the smaller the deviation $|y|$ and, correspondingly, the time t_1 , the earlier the process starts.

For $y_0 \ll |y| \ll 1$, the time-integral flux of reflected quanta is

$$Q_\alpha^B \approx \frac{N_\alpha}{\Delta\tilde{\omega}\tau_0} \frac{1}{|y|}. \quad (10.21)$$

The number of quanta within an energy range of about Γ at resonance is close to $N_\alpha\Gamma/\Delta\tilde{\omega} = N_\alpha/(\Delta\tilde{\omega}\tau_0)$. The growth of Q_α^B with decreasing $|y|$ is due to the enhancement of the reflection for energy wings far from resonance. As has been discussed in section 8, in a thick crystal the reduction of the scattering amplitude of a single nucleus with deviation from resonance is compensated to a noticeable extent, as $|y|$ decreases, by the growth of the thickness of the layer effectively involved in the reflection. The increased influence of the wings entails the reduction of the characteristic decay time of the excited system.

As one approaches the exact Bragg condition shifted by the angle α_0 (eq. (8.12)) when $|y|$ becomes smaller than $y_0^{1/2}$, it is necessary to include the imaginary part of the argument of the Bessel function in eq. (10.17). Now $t_2 < \tau_0$ and the involvement of electronic absorption becomes significant. In this limit the reflection intensity (eq. (10.17)) becomes

$$I_\alpha^\beta(t) \approx \frac{N_\alpha}{\tau_0(\Delta\omega\tau_0)} (y^2 + y_0^2)^{1/2} \frac{e^{-\tau}}{\tau^3} \exp \left[-2 \left(\frac{1}{p} - 1 \right) \frac{\tau}{\tau_2} \right]. \quad (10.22)$$

For $p = 1$, i.e., fulfillment of the condition necessary for realizing the suppression effect, expressions (10.22) and (10.20) are close to each other, describing the whole interval of time $t > t_1$. If $p < 1$, for $t > t_2$ the intensity is further reduced due to electronic absorption.

Let us now turn to the case of the exact Bragg condition ($y = 0$). If eq. (10.22) is integrated over time, the total flux of reflected quanta for $p = 1$ will be determined by eq. (10.21) with $y \rightarrow y_0$. A huge intensity is now reflected for short times of the order of magnitude of $\tau_{1\min} = y_0$. If one puts formally $\chi_0'' = 0$, the $I^B(t)$ -dependence becomes a δ -function. In fact, at any finite t the quantity (10.17) or (10.22) vanishes whereas (10.17) becomes infinite for $t = 0$. Thus, for the strict fulfilment of the Bragg condition and neglecting electronic absorption, the retardation effect vanishes and the reflection occurs instantaneously. This is a limiting manifestation of collective excitation in the nuclear system. In an infinitely thick crystal the above takes place

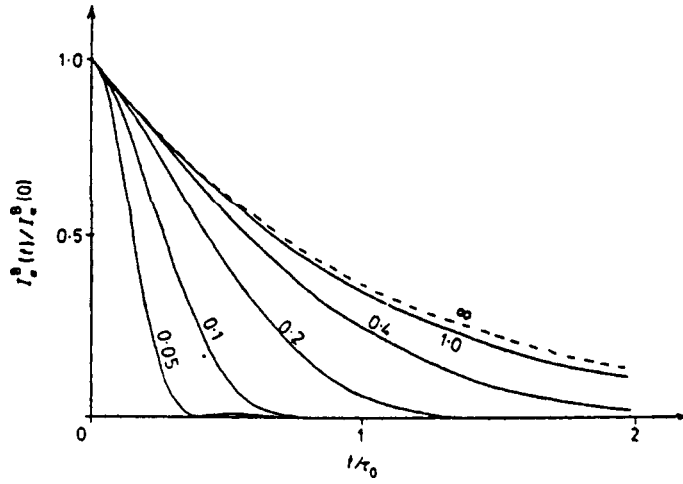


Figure 3. The time dependence of the intensity of the radiation Bragg-reflected from the thick crystal with the dependence on the deviation parameter y from the Bragg condition. The values of y are stated next to the curves. (From [33].)

for arbitrary p . For $p = 1$, when total suppression occurs at $y = 0$, the pulse is totally reflected, as we have seen in the stationary problem.

For $|y| \gg 1$, i.e., for large deviations from the Bragg angle, we have $\tau_1 \gg 1$ and find from eq. (10.17) for the whole time interval $\tau < \tau_1$

$$I_{\alpha}^{\text{B}}(t) = \frac{\pi}{2\tau_0} \frac{N_{\alpha}}{(\Delta\tilde{\omega}\tau_0)} \frac{1}{y^2} e^{-t/\tau_0}. \quad (10.23)$$

The time delay has a classical character since the collective coherent effects play an insignificant role. The change of character of the time delay of the reflected intensity at various angular deviations from the Bragg condition is seen clearly in figure 3 where the $I_{\alpha}^{\text{B}}(t)$ dependence is plotted for various values of the dimensionless parameter $|y|$.

In a real situation the incident beam has a finite angular divergence $\Delta\alpha$ around the value α_0 and, correspondingly, $\Delta y = \Delta\alpha/g$. Integrating eq. (10.17) over α with the assumption $\tau_2 \gg \tau > y_0$, we find the time behaviour of the reflection intensity at $\Delta y \gg 1$:

$$\bar{I}^{\text{B}}(t) \approx \frac{1}{3} \frac{16}{\tau_0} \frac{N_{\text{res}}}{\Delta y} \frac{e^{-\tau}}{\tau}, \quad (10.24)$$

where

$$N_{\text{res}} = \frac{\int N_{\alpha} d\alpha}{(\Delta\tilde{\omega}\tau_0)}.$$

It is seen that decay acceleration and growth of the reflection intensity with small τ are conserved for a large angular divergence.

Purely nuclear Bragg diffraction has been widely used for isolating the SR Mössbauer component, starting from the pilot experiment of Gerdau's group [32] (see,

e.g., reviews [42,43]). The study of time and angle Bragg-scattered SR dependence which is important for analyzing coherence was undertaken in [34,38,41,59]. Already in [41], performed on a ^{57}Fe -YIG single crystal, one can see the acceleration of the decay. In [34] with the Bragg scattering by a $^{57}\text{FeBO}_3$ single crystal, for the first time decay acceleration at the Bragg incident angle was studied quantitatively. A noticeable reduction of this acceleration when the incidence angle of the beam was shifted to both sides from Θ_B was observed. Later, the angular resolved time dependence was studied in [59]. A unique experiment, measuring the time dependence of the reflection intensity during the period of six (!) natural decay times was performed in Japan [38]. All these experiments, in fact, have displayed the main specific features typical for the time behaviour of diffracted radiation as well as the principal theoretical notions.

4. The possibility of using SR resonant nuclear scattering created an essential interest in designing pulse sources of Mössbauer radiation. The key feature is the isolation of the resonant part of the spectral density from the reflected radiation. Meanwhile, the collective character of exciting the nuclear system, the decay acceleration caused by this and, therefore, the broadening of the energy distribution of the reflected radiation produce essential difficulties for realizing the conditions necessary for conventional Mössbauer experiments.

To analyze this problem, we can investigate the transmission of the reflected radiation through a thin resonant absorber containing the same Mössbauer nuclei. Then the unified coherent system of Bragg scatterer and absorber is involved in the process and at the output from the latter the amplitude of the field equals

$$G(t, \Delta\omega_0) = \int \frac{d\omega}{2\pi} R_{\Delta\omega_0}^T(\omega) R^B(\omega) e^{-i\omega t}. \quad (10.25)$$

Here $R_{\Delta\omega_0}^T$ is the forward reflection amplitude (10.6) when shifting the resonance frequency in the absorber by $\Delta\omega_0$ with respect to that of the Bragg reflector. All coherently reflected radiation is assumed to be transmitted through an absorber.

For the analysis, it is convenient to transform eq. (10.25) to

$$G(t, \Delta\omega_0) = \int dt' G_{\Delta\omega_0}^T(t-t') G^B(t'). \quad (10.26)$$

A detailed consideration of the originating picture is given in [33]. We note only some qualitative results.

- (a) Provided the reflection occurs at a small deviation from the Bragg angle and $|y| \ll 1$, the dependence on $\Delta\omega_0$ proves to be weak. In other words, the resonant structure is practically lost. The time delay in this case is connected only with the absorber. This result can readily be understood if from the preceding analysis one recalls the strong involvement of the far-energy wings of the incident radiation in the reflection. Qualitatively, an improvement of the situation can be reached with

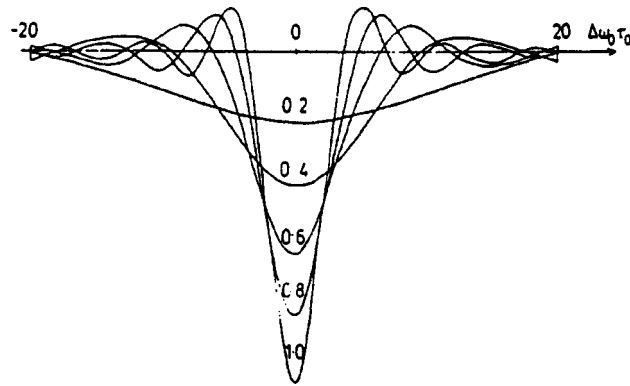


Figure 4. Relative intensity of radiation which is transmitted after reflection through the thin absorber with the resonance frequency displaced by $\Delta\omega_0$ at various time moments t after the pulse transmission. The values of t/τ_0 are stated next to the curves. The angular divergence of radiation $\Delta y \gg 1$, $p = 1$. (From [33].)

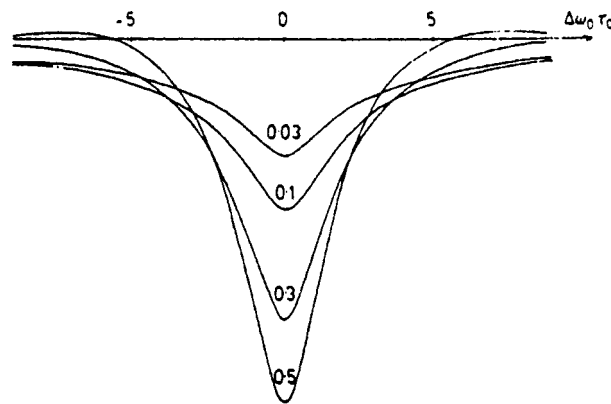


Figure 5. Integral over time of the relative intensity with the exception of the finite time interval $[0, t_1]$ for the system of a reflector and a thin absorber with the resonance frequency displaced by $\Delta\omega_0$. The values of t_1/τ_0 are stated next to the curves. The angular divergence of radiation $\Delta y \gg 1$, $p = 1$. (From [33].)

reflections from a relatively thin crystal. For $|y| \gg 1$, the field of the reflected radiation acquires resonant structure, but the intensity falls off as $1/y^2$.

- (b) If the incident beam has a wide angular distribution, the spectral properties of the retarded radiation depend significantly on the time delay. This is clearly seen in figure 4, where the dependence of the intensity at the absorber output on $\Delta\omega_0$ demonstrates the appearance of a clear resonance structure if the delay time becomes comparable with τ_0 . For a time small compared with τ_0 , the resonant structure of the radiation is practically absent.
- (c) A possibility to perform conventional Mössbauer measurements arises from using time-integral measurements excluding an initial time interval comparable to or,

maybe, somewhat smaller, than τ_0 . This can straightforwardly be traced in figure 5 where the special distributions of the integral intensity for various values of the excluded interval are given. Note that the omitted fraction of the intensity does not essentially decrease the number of quanta in a resonant interval of about Γ .

11. Concluding remarks

In the present paper we have presented the main ideas and results of the theory of coherent phenomena in resonant nuclear interaction of γ -quanta with crystals. The limited frame of the paper does not allow us to include all interesting results in this field. We only mention some of them.

- A detailed theory of γ -decay of nuclei in crystals containing the same nuclei in the ground state has been developed in [60]. The theory is based on the results given in sections 5 and 6 in accordance with the reciprocity theorem for absorbing media.
- There exists an interesting effect for the interference of the inelastic processes of photoeffect and internal conversion accompanying the resonant γ -quantum absorption. The effect has been observed experimentally by Sauer et al. [61]. The theory of the effect has been developed in [62–64].
- In connection with the analysis of γ -lasers exploiting the Mössbauer transition, a number of ideas based on the coherent dynamics of γ -quanta in crystals has appeared. One of them is based on inducing not a plane wave but a pair or multiwave Bragg states in the crystal (see, e.g., [65,66]).
- Large interest has recently been attracted by the possibility of measuring the phonon spectrum in crystals using nuclear resonant scattering of SR. In this connection we should note that the direct generalization of the results of section 9 enables the solution of the problem of finding the phonon dispersion law on the basis of inelastic coherent scattering of SR.

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