CONTRIBUTION TO THE THEORY OF DIFFRACTION OF GAMMA RADIATION BY CRYSTALS CONTAINING MOSSBAUER NUCLEI IN SITES WITH INHOMOGENEOUS ELECTRIC FIELDS

Yu. M. AĬVAZYAN and V. A. BELYAKOV

All-Union Institute of Physico-technical and Radiotechnical Measurements

Submitted July 19, 1968

Zh. Eksp. Teor. Fiz. **56**, 346–354 (January, 1969)

A kinematic theory of diffraction in nuclear resonant scattering of γ quanta by crystals containing Mossbauer nuclei in sites with inhomogeneous electric fields is developed. Expressions are obtained for the amplitude of the scattering by individual nuclei, for the cross sections of the coherent scattering by the crystal, and for the polarization of the radiation in the Bragg maxima for the case of quadrupole splitting of the nuclear levels of the Mossbauer nuclei. The conditions under which the obtained formulas are applicable to the description of the diffraction by real crystals are discussed. It is shown that Mossbauer diffraction may turn out to be an effective method of investigating ferroelectric structures in which the Mossbauer nuclei are in several crystallographic almost-equivalent positions.

1. INTRODUCTION

THE first experimental investigations of the diffraction of Mossbauer radiation by crystal structures have by now been performed (cf., e.g.,[1-3]). The theory of Mossbauer diffraction is being developed (see[4]), and the possibilities are discussed of its utilization for crystallographic investigation[2,5] and also for the investigation of magnetic ordering in crystals[6]. The possibility of a diffraction investigation of the magnetic structures is connected with the dependence of the Mossbauer-scattering amplitude on the magnitude and direction of the magnetic field at the scattering nucleus. If the scattering nucleus is situated in an inhomogeneous electric field, then the amplitude of the Mossbauer scattering turns out to depend on the character of the inhomogeneity of the field. In this connection, Mossbauer scattering by structures in which the Mossbauer nuclei are in sites with an inhomogeneous electric field have definite distinguishing features. Thus, the polarization and intensity of the scattered radiation at the Bragg maxima contain information concerning the gradient of the electric field at the Mossbauer nuclei and concerning their orientation relative to the crystallographic direction.

In the present paper we develop a kinematic theory of diffraction in resonant scattering of γ radiation by crystals containing Mossbauer nuclei in positions with different electric field gradients (EFG), neglecting the Rayleigh scattering. We discuss the conditions of applicability of the obtained formulas. We show that the Mossbauer diffraction by ferroelectric crystals of a certain type may turn out to be an effective method of investigating their structure. This pertains to structures in which atoms of the same chemical (Mossbauer) element are in several crystallographically non-equivalent positions, and this non-equivalence is "small" (i.e., it can be regarded as the result of small displacements of the atoms from the crystallographically equivalent positions). As is well known, investigations of such structures by usual methods entail considerable difficulties[7].

2. AMPLITUDE OF MOSSBAUER SCATTERING FOR THE CASE OF QUADRUPOLE SPLITTING OF NUCLEAR LEVELS

Let us consider the amplitude of the Mossbauer scattering by a nuclei situated in an inhomogeneous electric field. We assume that the Mossbauer nucleus has a nonzero quadrupole moment in the ground and in the excited states, and that the inhomogeneity of the electric field is not large enough to cause quadrupole splitting of the nuclear levels. Let the line width of the scattered Mossbauer radiation be smaller than the quadrupole splitting of the levels. In this case the scattering process proceeds via definite energy sublevels of the quadrupole splitting of the ground and excited states of the nucleus. Therefore the scattering amplitude can be represented in the form

$$f_{\tau\tau'} = B\sum_{\varkappa} f_{\tau\tau'}^{\varkappa} = B\sum_{\varkappa} \langle\psi_0{}^{\alpha\tau}|H|\psi_1{}^{\beta\varkappa}\rangle\langle\psi_1{}^{\beta\varkappa}|H|\psi_0^{\alpha\tau'}\rangle. \qquad (1)$$

In formula (1), H is the Hamiltonian of the interaction with the electromagnetic field, B is a factor which is of no importance to us. The explicit dependence of the amplitudes on the polarizations and the wave vectors of the γ quanta will not be written out for the time being. In the expression for the amplitude, the wave functions of the nucleus in the initial, intermediate, and final states are labeled by three indices, inasmuch as in the general case the energy levels of the quadrupole splitting are degenerate. For example, for $\psi_i^{\gamma\delta}$ the index i(0, 1) determines the state of the nucleus (ground or excited), γ determines the energies of the sublevels of the quadrupole splitting of the i-th state, and δ determines the number of the degenerate state of the sublevel corresponding to the energy E_i^{γ}.

The summation in (1) is over the energy-degenerate intermediate states, and thus the total amplitude of the scattering can be represented in the form of a sum of amplitudes $f^{\kappa}_{\tau\tau'}$, each of which corresponds to a process proceeding via a definite intermediate state κ.

In order to obtain a more detailed expression for the scattering amplitude, we represent the wave functions of the nucleus in the inhomogeneous electric field, $\psi_i^{\gamma\delta}$, in the form of an expansion in $|j, \mu_k\rangle$—functions with specified values of the angular momentum j and its projection μ_k on the direction of the largest value of the electric field gradient (the z axis)

$$\psi_i^{\gamma\delta} = \sum_k C_{ik}^{\gamma\delta} |j_i \mu_{ik}\rangle. \qquad (2)$$

In (2), $C_{ik}^{\gamma\delta}$ are the expansion coefficients.

Taking (1) and (2) into account, we obtain for the amplitude $f_{\tau\tau'}$ the expression

$$f_{\tau\tau'}(\mathbf{k}, \mathbf{n}; \mathbf{k}', \mathbf{n}') = B \sum_{\varkappa} f^{\varkappa}_{\tau\tau'}$$

$$= B \sum_{\varkappa} \sum_{kspq} C_{0k}^{*\alpha\tau} C_{0q}^{\alpha\tau'} C_{1s}^{*\beta\varkappa} C_{1p}^{\beta\varkappa} \langle j_0, \mu_{0k} | n^* e^{-j\mathbf{kr}} | j_1, \mu_{1s}\rangle$$

$$\cdot \langle j_1, \mu_{1p} | \mathbf{n}' e^{i\mathbf{k}'\mathbf{r}} \mathbf{j} | j_0, \mu_{0q}\rangle. \qquad (3)$$

In formula (3), $\mathbf{k}$ and $\mathbf{k}'$ are respectively the wave vectors of the initial and scattered γ quanta, $\mathbf{n}$ is the polarization vector of the initial γ quanta, the polarization vector $\mathbf{n}'$ describes the polarization in which we are interested after the scattering, and $\mathbf{j}$ is the nuclear current. In deriving expression (3) we have used the explicit form of the Hamiltonian of the interaction between the nucleus and the electromagnetic field.

Using the results of [6,8], in which we considered resonant scattering of γ quanta by nuclei placed in a magnetic field, we transform (3) into

$$f_{\tau\tau'} = B \sum_{\varkappa, kqsp} C_{0k}^{*\alpha\tau} C_{0q}^{\alpha\tau'} C_{1s}^{*\beta\varkappa} C_{1p}^{\beta\varkappa} (\mathbf{n}^* \mathbf{n}_{sh}) (\mathbf{n}'\mathbf{n}'^{*}_{pq}) \sqrt{I_{sh} I'_{pq}}, \qquad (4)$$

where $I_{sk}(I'_{pk})$ is intensity of the radiation in the transition $j_1, \mu_{1s} \to j_0, \mu_{0k}$ ($j_1, \mu_1 \to j_0, \mu_{0q}$) in the direction $\mathbf{k}$ ($\mathbf{k}'$), and $\mathbf{n}_{sk}$ ($\mathbf{n}'_{pq}$) is the polarization vector of the γ quantum with wave vector $\mathbf{k}$ ($\mathbf{k}'$), emitted in the transition $j_1, \mu_{1s} \to j_0, \mu_{0k}$ ($j_1, \mu_{1p} \to j_0, \mu_{0q}$). In expression (4), the quantities $\mathbf{n}_{sk}$ and $\mathbf{n}'_{pq}$ differ in the general case from the corresponding quantities of [8] by phase factors. This difference is connected with the fact that here in the calculation of the polarization vector we use wave functions written in a coordinate system whose unit vectors coincide with the principal directions of the EFG. In [8], the physical conditions singled out only the direction of the magnetic field (the z axis), and the choice of the direction of the other axis was determined only by considerations of convenience. We note that the factor B which enters in (1), (3), and (4) contains the dependence of the amplitude of the Mossbauer scattering on the γ-radiation energy. The detailed form of this dependence is of no importance to us here. We shall therefore assume that B differs from zero only when the resonance-scattering conditions are satisfied.

With the aid of expression (4) we obtain for the polarization vector $\mathbf{n}'$ of the γ quantum scattered in the direction $\mathbf{k}'$

$$\mathbf{n}' = \mathbf{N}_1 / |\mathbf{N}_1|, \qquad (5)$$

where

$$\mathbf{N}_1 = \sum_{\varkappa, kqsp} B^* C_{0k}^{\alpha\tau} C_{0q}^{*\alpha\tau'} C_{1s}^{\beta\varkappa} C_{1p}^{*\beta\varkappa} (\mathbf{n}\mathbf{n}_{sh}^*)\mathbf{n}_{pq}' \sqrt{I_{sh} I'_{pq}}.$$

It follows from (5) that the polarization of the scattering radiation contains information concerning the inhomogeneity of the electromagnetic field at the scattering nucleus, the dependence of the polarization vector on the EFG enters in (5) via C_{ik} and also via the primed and unprimed quantities $\mathbf{n}_{sk}$ and I_{sh}.

3. GENERAL CASE OF DIFFRACTION

We consider the scattering of Mossbauer radiation by a crystal containing Mossbauer nuclei. The scattering of the γ quanta occurs both from the nuclei (Mossbauer scattering) and from the electrons (Rayleigh scattering). For reasons discussed later, we shall take into account only the nuclear scattering.

We assume that the Mossbauer nuclei are in crystal sites with different values of the EFG, and that the scattering crystal is sufficiently thin, so that extinction can be neglected. Using the expression for the scattering amplitude (4), we can represent the coherent-scattering cross section in the form

$$d\sigma_{\text{coh}}(\mathbf{k}, \mathbf{n}; \mathbf{k}', \mathbf{n}') = AC^2\eta^2 \Big| \sum_{l, \tau_l} f^{(l)}_{\tau_l\tau_l}(\mathbf{k}, \mathbf{n}; \mathbf{k}', \mathbf{n}') e^{i(\mathbf{k}-\mathbf{k}')\mathbf{r}_l} \Big|^2 \sum_{\mathbf{b}} \delta(\mathbf{k} - \mathbf{k}' - 2\pi\mathbf{b})\, d\Omega_{\mathbf{k}'}. \qquad (6)$$

Here A is a factor whose detailed form [9] is of no interest to us here, $\mathbf{b}$ is the reciprocal lattice vector, $\mathbf{r}$ is a vector determining the position of the Mossbauer spectrum in the unit cell of the crystal, C is the concentration of the Mossbauer isotope, and $\eta = 1/(2j_0 + 1)$ is a factor connected with the spin incoherence. The remaining symbols are the same as in the preceding section. The index l designates quantities pertaining to the sites with the l-th value of the EFG.

If the EFG at the Mossbauer nuclei are such that different quadrupole splittings of the nuclear levels take place, then the amplitudes $f^{(l)}_{\tau\tau}$ for scattering by nuclei for which the condition of resonant scattering is not satisfied vanish in expression (6). In analogy with (5) we obtain for the radiation polarization vector at the Bragg maximum, from expressions (4) and (6),

$$\mathbf{n}' = \mathbf{N}_2 / |\mathbf{N}_2|,$$

$$\mathbf{N}_2 = \sum_{\varkappa l}{}' \sum_{kqsp} B_l^* C^{\alpha\tau}_{0k, l} C^{*\alpha\tau'}_{0q, l} C^{\beta\varkappa}_{1s, l} C^{*\beta\varkappa}_{1p, l} (\mathbf{n} \cdot \mathbf{n}^*_{sk, l})\, \mathbf{n}'_{pq, l} \sqrt{I^l_{sk} I'^l_{pq}}\, e^{-i(\mathbf{k}-\mathbf{k}')\mathbf{r}_l}. \qquad (7)$$

Just as in (6), the summation over l in (7) is carried out over sites l for which the resonance-scattering condition is satisfied. The expressions for the cross section (6) and for the radiation polarization (7) were derived by us under the assumption that the initial beam of γ quanta is completely polarized.

We shall need subsequently quantities obtained from (6) by averaging over the initial and summing over the final polarizations. We shall denote by $d\sigma(\mathbf{k}; \mathbf{k}', \mathbf{n}')$ the result of averaging (6) over the initial polarizations, by $d\sigma(\mathbf{k}, \mathbf{n}; \mathbf{k}')$ the result of summing (6) over the final polarizations, and by $d\sigma(\mathbf{k}; \mathbf{k}')$ the result of averaging over the initial and summing over the final polarization. For an unpolarized initial radiation, the scattered radiation will in the general case be partly polarized. In this case, the polarization density matrix ρ_{unp} of the scattered radiation can be represented in the form

$$\rho_{\text{unp}} = \sum_{i=1,2} d\sigma(\mathbf{k}, \mathbf{n}_i; \mathbf{k}')\rho(\mathbf{n}_{0i}) \Big/ \Big(\sum_{i=1,2} d\sigma(\mathbf{k}, \mathbf{n}_i; \mathbf{k}') \Big), \tag{8}$$

where $\mathbf{n}_i (i = 1, 2)$ are two orthgonal unit vectors of the initial polarization (for example the vectors of right-hand and left-hand circular polarizations), $\mathbf{n}'_{0i}$ is the polarization vector of the scattered radiation when the initial-polarization vector is $\mathbf{n}_i$, and $\rho(\mathbf{n})$ is the polarization density matrix corresponding to the polarization vector $\mathbf{n}$[10].

If the initial beam is partly polarized, then we obtain for the scattering cross section $d\sigma_P$ and for the polarization density matrix ρ_P

$$d\sigma_P(\mathbf{k}; \mathbf{k}', \mathbf{n}') = (1-P)\, d\sigma(\mathbf{k}; \mathbf{k}', \mathbf{n}') + P\, d\sigma(\mathbf{k}, \mathbf{n}_P; \mathbf{k}', \mathbf{n}'), \tag{9}$$

$$\rho_P = \frac{(1-P)\,\rho_{\text{unp}} d\sigma(\mathbf{k}; \mathbf{k}') + P\rho(\mathbf{n}_0')\, d\sigma(\mathbf{k}, \mathbf{n}_P; \mathbf{k}')}{(1-P)\, d\sigma(\mathbf{k}; \mathbf{k}') + P\, d\sigma(\mathbf{k}, \mathbf{n}_P; \mathbf{k}')}, \tag{10}$$

where P is the degree of polarization of the initial γ quanta, the vector $\mathbf{n}_P$ describes the polarization that is partially represented in the initial beam, and $\mathbf{n}'_0$ is the polarization vector of the scattered radiation for the initial polarization, described by the vector $\mathbf{n}_P$.

From the general formulas (4), (6) and (7) we see that the dependence of the amplitude of the Mossbauer scattering on the EFG turns out to be appreciable for the intensity and polarization of the radiation at the Bragg maxima. More detailed discussions of these dependences will be presented using a concrete example.

4. DIFFRACTION FOR THE CASE OF TWO VALUES OF AN AXIALLY SYMMETRICAL EFG

Let us analyze the obtained general expressions using a concrete example. Let $l = 2$, $j_0 = {}^1\!/_2$, and $j_1 = {}^3\!/_2$ in the preceding formulas, and let the EFG be axially symmetrical. We also assume that the Mossbauer transition is a dipole transition. Under the foregoing assumptions, there is no quadrupole splitting of the ground state. The excited level is split into two sublevels. All the energy levels are doubly degenerate. Let the EFG differ only in the orientation of the axes. This means that for both EFG the energy splittings of the levels are the same, and therefore the resonance-scattering conditions are satisfied simultaneously.

Under the foregoing assumptions, the wave functions of the excited state of the nucleus (2), corresponding to the energies $E_1^{3/2}$ and $E_1^{1/2}$, are given by

$$\psi_1^{{}^3\!/_2, \pm {}^3\!/_2} = \frac{1}{\sqrt{2}}\Big(\Big| \frac{3}{2}, \frac{3}{2} \Big\rangle \pm \Big| \frac{3}{2}, -\frac{3}{2} \Big\rangle \Big),$$

$$\psi_1^{{}^1\!/_2, \pm {}^1\!/_2} = \frac{1}{\sqrt{2}}\Big(\Big| \frac{3}{2}, \frac{1}{2} \Big\rangle \pm \Big| \frac{3}{2}, -\frac{1}{2} \Big\rangle \Big) \tag{11}$$

and the wave functions of the ground state with energy $E_0^{1/2}$ by

$$\psi_0^{{}^1\!/_2, \pm {}^1\!/_2} = \Big| \frac{1}{2}, \pm \frac{1}{2} \Big\rangle. \tag{12}$$

Let the resonant scattering proceed via the transition $E_0^{1/2} \to E_1^{3/2}$. Then the expression for the amplitude (1) takes the form

$$f_{{}^1\!/_2, {}^1\!/_2} = B' \langle {}^1\!/_2, {}^1\!/_2 | H | {}^3\!/_2, {}^3\!/_2 \rangle \langle {}^3\!/_2, {}^3\!/_2 | H | {}^1\!/_2, {}^1\!/_2 \rangle. \tag{13}$$

In expression (13) there is no sum over the intermediate states, since the transition ${}^1\!/_2 \to -{}^3\!/_2$ is forbidden by the selection rules for dipole radiation. Just as in[8], the expression for the amplitude (13) can be reduced to the form

$$f_{{}^1\!/_2, {}^1\!/_2} = B' (\mathbf{n}^* \mathbf{n}_{{}^3\!/_2, {}^1\!/_2}) (\mathbf{n}' \mathbf{n}'^*_{{}^3\!/_2, {}^1\!/_2}) \sqrt{I_{{}^3\!/_2, {}^1\!/_2} I'_{{}^3\!/_2, {}^1\!/_2}}, \tag{14}$$

where

$$\mathbf{n}_{{}^3\!/_2, {}^1\!/_2} = \frac{[\mathbf{k}[\mathbf{kh}]]}{|\mathbf{k}|\,|[\mathbf{kh}]|} \cos\alpha + i \frac{[\mathbf{kh}]}{|[\mathbf{kh}]|} \sin\alpha, \quad \operatorname{tg}\alpha = \frac{1}{\cos\theta}, \tag{15*}$$

where $\mathbf{h}$ is the unit vector in the z direction, and θ is the angle between $\mathbf{k}$ and $\mathbf{h}$,

$$I_{{}^3\!/_2, {}^1\!/_2} = a(1 + \cos^2\theta). \tag{16}$$

Here a is a factor that depends on the characteristics of the nuclear transition (see, for example,[8]). The primed quantities in (14) are obtained from (15) and (16) by replacing $\mathbf{k}$ with $\mathbf{k}'$. The expression for $f_{-1/2,-1/2}$ is obtained from (14) by replacing $\mathbf{n}_{3/2,1/2}$ and $\mathbf{n}'_{3/2,1/2}$ with their complex conjugates.

We have considered the amplitudes of processes that do not change the state of the scattering nucleus, for only such processes contribute to the coherent scattering.

For formula (6) we obtain for the coherent scattering cross section

$$d\sigma_{\text{coh}}(\mathbf{k}, \mathbf{n}; \mathbf{k}'\mathbf{n}') = A' \big| (f^{(1)}_{{}^1\!/_2, {}^1\!/_2} + f^{(1)}_{-{}^1\!/_2, -{}^1\!/_2}) + e^{i\delta} (f^{(2)}_{{}^1\!/_2, {}^1\!/_2} + f^{(2)}_{-{}^1\!/_2, -{}^1\!/_2}) \big|^2 \sum_{\mathbf{b}} \delta(\mathbf{k} - \mathbf{k}' - 2\pi\mathbf{b})\, d\Omega_{\mathbf{k}'}, \tag{17}$$

where $\delta = (\mathbf{k} - \mathbf{k}')(\mathbf{r}_2 - \mathbf{r}_1)$.

For the polarization vector in the Bragg maximum we get from (5)

$$\mathbf{n}' = \mathbf{N}_3 / |\mathbf{N}_3|,$$

$$\mathbf{N}_3 = \sum_{l=1,2} [(\mathbf{n}\mathbf{n}^{*l}_{{}^3\!/_2, {}^1\!/_2}) \mathbf{n}'^{l}_{{}^3\!/_2, {}^1\!/_2} + (\mathbf{n}\mathbf{n}^{*l}_{-{}^3\!/_2, -{}^1\!/_2}) \mathbf{n}_{-{}^3\!/_2, -{}^1\!/_2}] e^{i\delta_l} \sqrt{I^l(\mathbf{k}) I'^l(\mathbf{k}')},$$

where

$$\delta_1 = 0, \quad \delta_2 = (\mathbf{k}' - \mathbf{k})(\mathbf{r}_2 - \mathbf{r}_1). \tag{18}$$

We have considered above the case for which the EFG at the Mossbauer nuclei differed only in the orientation of the symmetry axes, and therefore the quadrupole splitting and the conditions of the resonant (Mossbauer) scattering for both EFG were the same. We now proceed to the case when the differences into EFG lead to different quadrupole splittings and to different resonance-scattering conditions. Now in expression (14) the amplitudes $f^{(1)}$ and $f^{(2)}$ cannot be simultaneously different from zero. For the i-th resonant energy, expression (14) takes the form

$$d\sigma_{\text{coh}}(\mathbf{k}, \mathbf{n}; \mathbf{k}', \mathbf{n}') = A' \big| f^{(i)}_{{}^1\!/_2, {}^1\!/_2} + f^{(i)}_{-{}^1\!/_2, -{}^1\!/_2} \big|^2 \sum_{\mathbf{h}} \delta(\mathbf{k} - \mathbf{k}' - 2\pi\mathbf{b})\, d\Omega_{\mathbf{k}'}, \tag{19}$$

where i = 1 and 2, and for the radiation polarization vector in the Bragg maximum we obtain

$$\mathbf{n}' = \mathbf{N}_4 / |\mathbf{N}_4|,$$

$$\mathbf{N}_4 = (\mathbf{n}\mathbf{n}^{*i}_{{}^3\!/_2, {}^1\!/_2}) \mathbf{n}'^{i}_{{}^3\!/_2, {}^1\!/_2} + (\mathbf{n}\mathbf{n}^{i}_{{}^3\!/_2, {}^1\!/_2}) \mathbf{n}'^{i*}_{{}^3\!/_2, {}^1\!/_2}. \tag{20}$$

From (17) we obtain for the scattering cross section of the polarized beam, summed over the final polariza-

---

*$[\mathbf{kh}] \equiv \mathbf{k} \times \mathbf{h}$

tion,

$$d\sigma_{\text{coh}}(\mathbf{k};\mathbf{k}') = A'\{2 + \cos^2\theta_1\cos^2\theta_1' + \cos^2\theta_2\cos^2\theta_2' + 2\cos\delta[\cos\varphi\cos\varphi'(\cos\theta_1\cos\theta_2\cos\theta_1'\cos\theta_2' + 1) + \cos\varphi_1\cos\varphi_1'(\cos\theta_2\cos\theta_2' + \cos\theta_1\cos\theta_1')]\}\cdot\sum_{\mathbf{b}}\delta(\mathbf{k}-\mathbf{k}'-2\pi\mathbf{b})\,d\Omega_{\mathbf{k}'}, \tag{21}$$

where θ_1 and θ_2 are the angles between $\mathbf{k}$ and $\mathbf{h}_1$ and between $\mathbf{k}$ and $\mathbf{h}_2$, respectively, and φ and φ_1 are respectively the angles between the vector $\mathbf{k}\times\mathbf{h}_1$ and the vectors $\mathbf{k}\times\mathbf{h}_2$ and $\mathbf{k}\times[\mathbf{k}\times\mathbf{h}_2]$ (θ' and φ' differ from θ and φ in that $\mathbf{k}$ is replaced with $\mathbf{k}'$ in the corresponding expressions).

For the case when the conditions for resonant scattering are satisfied only for one value of the EFG, we obtain from (19)

$$d\sigma_{\text{coh}}(\mathbf{k};\mathbf{k}') = A'(1+\cos^2\theta_i\cos^2\theta_i')\sum_{\mathbf{b}}\delta(\mathbf{k}-\mathbf{k}'-2\pi\mathbf{b})\,d\Omega_{\mathbf{k}'}. \tag{22}$$

We have considered resonant scattering in the transition $E_0^{1/2}\to E_1^{3/2}$. Scattering in the transition $E_0^{1/2}\to E_0^{1/2}$ can be considered in similar fashion.

The formulas (17) and (19) for the cross section contain the Bragg maxima whose positions coincide with the positions of the maxima in x-ray scattering (at the same x-ray energy). However, the relative intensities in the Bragg maxima differ for Mossbauer spectrum from the corresponding values for x-ray scattering. This is a consequence of two factors. First, the angular dependence of the Mossbauer amplitude of the scattering differs from the angular dependence of the x-ray amplitude. Second, the amplitude of the Mossbauer scattering by an individual nucleus depends on the EFG at this nucleus (i.e., on the position of the nucleus in the crystal cell). On the other hand, the amplitude of the x-ray scattering does not depend on the position of the atom in the cell. For example, in the previously analyzed case of different quadrupole splittings at a fixed energy of the primary γ quanta, the only nonvanishing scattering amplitudes are from those nuclei, for which the resonance condition is satisfied. Whereas the first factor leads, in general, to a small difference between the relative intensities, the second factor may lead to a qualitative difference in the character of the Mossbauer diffraction from the character of the x-ray diffraction, for example to an appearance in the Mossbauer scattering of Bragg maxima that are strictly forbidden for the Rayleigh scattering, or to an appreciable difference between the relative intensities in either case. We shall illustrate the latter statement by means of an example.

5. DIFFRACTION BY FERROELECTRIC STRUCTURES

Let us consider the Mossbauer diffraction by a complex ferroelectric structure, in which the Mossbauer atoms are in crystallographically non-equivalent positions, but this non-equivalence is "small," and let us compare it with the diffraction of x-rays by the same structure. As is well known, for such a structure, it is difficult to establish the existence of non-equivalent positions of atoms of the same chemical element by the usual methods[7]. For such structures, the Bragg diffraction maxima in the scattering can be subdivided into two types: a) maxima that exist for a structure obtained from the considered structure by neglecting the aforementioned small non-equivalence of the atom positions (structure maxima); b) maxima connected with the presence of non-equivalent positions of the same chemical element in the unit cell of the crystal and vanishing in the absence of this non-equivalence (superstructure maxima).

As is well known, in the scattering of x-rays and neutrons, the ratio of the intensity of the superstructure maximum to the intensity of the structure maximum is small and is of the order of $(\delta a/a)^2$, where δa are the atomic displacements causing the non-equivalence, and a is the period of the lattice.

In the case of diffraction of Mossbauer radiation, the situation may change radically. Namely, the intensity of the superstructure maxima may not contain the small quantities $(\delta a/a)^2$ relative to the intensity of the structure maxima. This pertains to structures in which the element located at the non-equivalent positions is a Mossbauer element, and the discussed non-equivalence leads to the existence of different values of the EFG at the nuclei of the Mossbauer isotopes. Examples of such structures may be certain ferroelectrics in which the ferroelectric phase transition is connected with a change in the number of atoms per unit cell.

The physical cause of the noted difference in the character of the diffraction in Mossbauer scattering from the diffraction occurring in the scattering of x-rays and neutrons lies in the dependence of the amplitude of the Mossbauer scattering on the EFG at the scattering nucleus. Indeed, whereas for neutrons and x-rays the discussed displacements do not change the atomic scattering amplitudes, and therefore the intensity of the superstructure maxima are determined only by small displacements of the atoms, in the case of Mossbauer scattering the same displacements, by virtue of the dependence of the nuclear scattering amplitude on the EFG, lead to changes of the amplitudes of the Mossbauer scattering. Therefore in the latter case the intensity of the superstructure maxima is determined not so much by the small displacement as by the difference in the amplitudes of the Mossbauer scattering spectrum by nuclei situated in crystallographically non-equivalent positions. Since the differences of the amplitudes in the general case are of the same order as the amplitudes themselves, the intensity of the superstructure maxima turns out to be of the order of the intensity of the structure.

For two non-equivalent crystallographic positions of the Mossbauer isotope, the scattering cross section is described by the formulas of Sec. 4. In expression (17) for the scattering cross section we have $e^{i\delta}\approx 1$ in the structure maxima, and $e^{i\delta}\approx -1$ in the superstructure maxima. Therefore the ratio of the intensity of the scattered radiation in the superstructure maximum to the same quantity for the nearest structure maximum is approximately equal to

$$\frac{|(f^{(1)}_{1/2,\,1/2}+f^{(1)}_{-1/2,\,-1/2})-(f^{(2)}_{1/2,\,1/2}+f^{(2)}_{-1/2,\,-1/2})|^2}{|(f^{(1)}_{1/2,\,1/2}+f^{(1)}_{-1/2,\,-1/2})+(f^{(2)}_{1/2,\,1/2}+f^{(2)}_{-1/2,\,-1/2})|^2}. \tag{23}$$

In this formula we have neglected the difference between the Bragg angles for the structure and superstructure maxima. For unequal quadrupole splittings, one of the amplitudes in (17) vanishes and the ratio (23) is equal to unity. If the quadrupole splittings are equal, this ratio is also of the order of unity.

For x-ray diffraction, the intensity ratio, neglecting the difference between $\cos\delta$ and ± 1, is determined by a formula analogous to (23), and is equal to zero since the corresponding difference of the x-ray scattering amplitude vanishes. Allowance for the deviation of $\cos\delta$ from ± 1 yields the estimate presented above for the ratio $(\delta a/a)^2$.

6. CONCLUSION

In the foregoing analysis we have neglected the Rayleigh scattering. In many cases the Rayleigh scattering and its interference with the nuclear scattering exert an appreciable influence on the γ-quantum diffraction[3,4]. In certain cases, however, the Rayleigh scattering can be neglected. Neglect of the Rayleigh scattering is always justified for pure Mossbauer structures and, as demonstrated above, may be justified for superstructure maxima in the case of certain ferroelectric structures. In addition, diffraction conditions can be realized, in which the amplitude of the Rayleigh scattering vanishes (for example[2]), and it is therefore sufficient to take into account only the nuclear scattering.

We did not discuss the angular dimensions of the diffraction maxima and the temperature dependence of the intensity of the diffraction maxima. Just as in[6], we obtain for the width of the maxima the estimate $\theta \sim \lambda/l$, where λ is the wavelength of the γ quantum and l is the extinction length. The temperature dependence of the intensity of the diffraction maxima under the assumptions made in this paper is determined by the temperature dependence of the Lamb-Mossbauer factor, the square of which is proportional to the factor A in (6).

[1] P. J. Black and P. B. Moon, Nature **188**, 481 (1960).

[2] P. J. Black and J. P. Duerdoth, Proc. Phys. Soc. **84**, 169 (1964).

[3] V. K. Voĭtovetskiĭ, I. L. Korsunskiĭ, A. I. Novikov, and Yu. R. Pazhin, Zh. Eksp. Teor. Fiz. **54**, 1361 (1968) [Sov. Phys.-JETP **27**, 729 (1968)].

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

[5] R. N. Kuz'min, A. V. Kolpakov, and G. S. Zhdanov, Kristallografiya **11**, 511 (1966) [Sov. Phys. Crystallogr. **11**, (196).

[6] V. A. Belyakov and Yu. M. Aĭvazyan, ZhETF Pis. Red. **7**, 477 (1968) [JETP Lett. **7**, 368 (1968)]; Preprint VNIIFTRI No. 16 (1968).

[7] F. Jona and G. Shirane, Ferroelectric Crystals, Pergamon, 1962.

[8] V. A. Belyakov and V. P. Orlov, Zh. Eksp. Teor. Fiz., in print, [Sov. Phys.-JETP in print).

[9] G. T. Trammel, Phys. Rev. **126**, 1045 (1962).

[10] A. I. Akhiezer and V. B. Berestetskiĭ, Kvantovaya elektrodinamika (Quantum electrodynamics) Fizmatgiz, 1959. [Interscience, 1965].

Translated by J. G. Adashko
44