CERTAIN PROBLEMS OF KINEMATIC THEORY OF BRAGG SCATTERING OF RESONANT X-RAYS

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Submitted June 13, 1970

Zh. Eksp. Teor. Fiz. 59, 2083–2090 (December, 1970)

A study is made of the kinematic theory of diffraction of resonant γ rays by single crystals. The differential cross sections of Bragg scattering are calculated for the case of total degeneracy and Zeeman splitting (in ferro- and antiferromagnets of the collinear type) of the nuclear levels, and the contributions of the nuclear-resonance, Rayleigh, and interference scatterings are determined exactly. The Mossbauer spectrum of the diffraction is also calculated, with account taken of the absorption of the primary and secondary beams in the scatterer.

BRAGG scattering of resonant γ rays by single crystals has been intensely investigated recently. The results of recent experiments (see, for example,[1,2]) demonstrate convincingly that there is a real possibility of using Bragg scattering as a new method of diffraction, in addition to traditional methods of diffraction of x-rays and neutrons. A number of features of both the kinematic[3-5] and the dynamic theory of diffraction of resonant γ rays[2,6,7] give grounds for hoping that this trend will become quiet promising for the study of different problems of physics, particularly solid-state physics. The development of experimental research in this direction obviously calls, first, for an exact kinematic theory capable of determining the intensity of the Bragg diffraction by ideal single crystals. Unfortunately, there is still no fully developed theory of this kind.

We calculate in this paper the differential cross sections of Bragg elastic scattering of resonant γ rays, with an exact determination of the contributions of the nuclear-resonance, Rayleigh, and interference scattering in each Bragg peak. It should be noted that in experiments, these differential cross sections and the physical quantities associated with them are determined from the Mossbauer diffraction spectra. Owing to the presence of interference between the nuclear-resonance and Rayleigh scatterings, this spectrum has a more complicated form than the ordinary absorption spectrum, and is very sensitive to variations of the effective thickness of the scatterer. This question is also considered in the paper.

Without loss of generality, we assume henceforth that the crystal consists of identical (resonant) nuclei. Allowance for the isotopic incoherence is quite trivial (see, for example[8]). We also neglect the influence of thermal motion of the lattice. This influence can readily be taken into account by multiplying the scattering amplitudes by certain known factors. This question was considered in[4,9].

The differential cross section for elastic scattering of resonant γ rays, referred to one nucleus of the crystal, can be written in the general case in the form

$$\frac{d\sigma}{d\Omega}=\frac{1}{N}\left|\sum_n M_n \exp\{i(\mathbf{k}-\mathbf{k}')\mathbf{r}_n\}\right|^2, \qquad (1)$$

where N is the number of atoms in the crystal, $\mathbf{r}_n$ are the coordinates of the n-th atom in the lattice, $\mathbf{k}$ and $\mathbf{k}'$ are the wave vectors of the incident and scattered photons, and M_n is the amplitude of elastic scattering by the n-th atom, which consists of the amplitude of the nuclear-resonance scattering and the amplitude of the Rayleigh scattering

$$M(\mathbf{k}\mathbf{k}'pp'j_nj_n')=\frac{N(\mathbf{k}\mathbf{k}'pp'j_nj_n')}{E-E_0+i\Gamma/2}+R(\mathbf{k}\mathbf{k}'pp'). \qquad (2)$$

In this expression, E_0 is the resonant energy, Γ the width of the excited state of the nucleus, j_n and j_n' the projections of the ground-state spin of the scattering nucleus before and after scattering, and p and p′ are the polarization indices of the incident and scattered photons ($p, p' = \pm 1$).

Expression (1) produces diffraction maxima along directions satisfying the Bragg condition $\mathbf{k}' = \mathbf{k} - 2\pi\boldsymbol{\tau}$, where $\boldsymbol{\tau}$ is the reciprocal lattice vector. Then, as is well known, only the coherent part of the scattering amplitude contributes to the intensity of the diffraction maxima. For this part we have

$$\left(\frac{d\sigma}{d\Omega}\right)_{\text{coh}}=\frac{(2\pi)^3}{V_0}\left|\sum_{n'}(M_{\text{coh}})_{n'}\exp\{i(\mathbf{k}-\mathbf{k}')\mathbf{r}_{n'}\}\right|^2\delta(\mathbf{k}-\mathbf{k}'-2\pi\boldsymbol{\tau}), \qquad (3)$$

where V_0 is the volume of the unit cell, and n′ now denotes the indices of summation over the atoms within the limits of one unit cell; M_{coh} is the amplitude of the coherent elastic scattering. In the case when the scatterer consists of atoms of the same element, the quantity $(M_{\text{coh}})^2$ can be taken outside the summation sign (with the exception of the case considered in Sec. 3), i.e., we obtain

$$\left(\frac{d\sigma}{d\Omega}\right)_{\text{coh}}=\frac{(2\pi)^3}{V_0}M_{\text{coh}}^2F(\mathbf{k}-\mathbf{k}')\delta(\mathbf{k}-\mathbf{k}'-2\pi\boldsymbol{\tau}), \qquad (4)$$

where $F(\mathbf{k}-\mathbf{k}')$ is the usual structure factor.

To determine the expression for M_{coh}, we first note that the quantum-mechanical coherence condition requires that the scattering act leave the state of the scattering system unchanged[10]. Applying this to our problem, we see that Rayleigh scattering is completely coherent, whereas for the nuclear-resonance scattering the coherence condition is satisfied only if the projection of the spin of the ground state of the nucleus is conserved. This corresponds to the ampli-

tude $N(\mathbf{k}\mathbf{k}'pp'j_n)$. In addition, M_{coh} should be taken to mean the scattering amplitude averaged over the initial state of the scattering system[11]. We thus have in our problem

$$M_{ког}=\frac{\langle N(\mathbf{k}\mathbf{k}'pp'j)\rangle}{E-E_0+i\Gamma/2}+R(\mathbf{k}\mathbf{k}'pp'), \tag{5}$$

where the angle brackets denote averaging over the spin states of the scattering nucleus.

Starting from the general theory of radiation and scattering of a photon (see, for example,[10]), we can write

$$N(\mathbf{k}\mathbf{k}'pp'j)=(2L+1)$$

$$(2I+1)\varepsilon\frac{\Gamma_R}{4k}\sum\begin{pmatrix}J & L & I\\ j & m & -j-m\end{pmatrix}^2 D^{L*}_{pm}(\mathbf{k})D^{L}_{p'm}(\mathbf{k}'), \tag{6}$$

where Γ_R and I are the radiation width and the spin of the excited state of the nucleus, L is the multipolarity of the γ transition, $\epsilon = 1$ for a magnetic transition, and $\epsilon = pp'$ for an electric transition. The amplitude of the Rayleigh scattering is conveniently written in the form

$$R(\mathbf{k}\mathbf{k}'pp')=Zr_0pp'\sum_{\mu=0,\pm1}D^{1*}_{p\mu}(\mathbf{k})D^{1}_{p'\mu}(\mathbf{k}'), \tag{7}$$

where $r_0 = e^2/mc^2$ is the classical radius of the electron, and Z is the atomic form factor. In formula (7) the indices $\mu = 0$ and ± 1, and in formula (6) the values of the summation indices m depend on the concrete conditions of the scattering problem. Let us consider certain cases.

1. Case of Total Degeneracy of Nuclear Levels

In this case it can readily be shown, using for convenience the direction of the wave vector k as the quantization axis, that

$$\langle N(\mathbf{k}\mathbf{k}'pp'j)\rangle=\frac{1}{2J+1}\sum_j N(\mathbf{k}\mathbf{k}'pp'j)=\frac{2I+1}{2J+1}\frac{\Gamma_R}{4k}\varepsilon D^{L}_{p'p}(\mathbf{k}'). \tag{8}$$

and

$$R(\mathbf{k}\mathbf{k}'pp')=Zr_0pp'D^{1}_{p'p}(\mathbf{k}'). \tag{9}$$

The energy dependence of the differential cross section of the Bragg scattering is expressed in terms of the quantity $|M_{coh}|^2$. It is convenient to write it in the form

$$|M_{coh}|^2=\frac{A\Gamma^2/4+B\Gamma(E-E_0)}{(E-E_0)^2+\Gamma^2/4}+C, \tag{10}$$

where A and C characterize the intensities of the nuclear-resonance and Rayleigh scatterings, and B is the contribution of the interference between these channels. For the case of a polarized incident beam we obtain after simple manipulations

$$A=\left(\frac{2I+1}{2J+1}\frac{\lambda}{2\pi}\frac{\Gamma_R}{\Gamma}\right)^2\frac{1}{2}\sum_\nu(4\nu+1)\begin{pmatrix}L & L & 2\nu\\ 1 & -1 & 0\end{pmatrix}^2 P_{2\nu}(\cos 2\alpha), \tag{11}$$

$$C=\tfrac{1}{3}(Zr_0)^2(1+\cos^2 2\alpha), \tag{12}$$

$$B=\frac{2I+1}{2J+1}\frac{\lambda}{2\pi}Zr_0\frac{\Gamma_R}{\Gamma}\sum_\nu(2\nu+1)\begin{pmatrix}L & 1 & \nu\\ 1 & -1 & 0\end{pmatrix}P_\nu(\cos 2\alpha), \tag{13}$$

where λ is the photon wavelength and 2α is the scattering angle. In (13), the indices ν assume odd values for a magnetic transition and even ones for an electric one. In the experiments one usually has the dipole-magnetic transition M1. For this case, Eqs. (11)–(13) take the form

$$\sqrt{\frac{A}{C}}=\frac{1}{4\pi}\frac{2I+1}{2J+1}\frac{\Gamma_R}{\Gamma}\frac{\lambda}{Zr_0} \tag{14}$$

$$\frac{B}{C}=\sqrt{\frac{A}{C}}\frac{2\cos 2\alpha}{1+\cos^2 2\alpha} \tag{15}$$

2. Case of Zeeman Splitting of Nuclear Levels

We consider now the case of Zeeman splitting of nuclear levels in a scatterer. We shall assume that the energy spectrum of the incident beam is a single line, and the magnitude of the Zeeman splitting is sufficiently larger than the width of the excited state of the nucleus so that a "Doppler adjustment" of the frequency on the incident beam makes it possible to investigate the Bragg scattering by individual Zeeman components. Thus, we assume that we investigate in the experiment scattering by the Zeeman component corresponding to a γ transition from the sublevel of the ground state j to the sublevel of the excited state j + m. For the differential cross section of the Bragg scattering we also have formula (4) with

$$M_{coh}=\frac{1}{2J+1}\frac{N(\mathbf{k}\mathbf{k}'pp'jm)}{E-E_0+i\Gamma/2}+R(\mathbf{k}\mathbf{k}'pp'), \tag{16}$$

where E_0 now denotes the resonant energy of the given Zeeman component, and

$$N(\mathbf{k}\mathbf{k}'pp'jm)=(2L+1)(2I+1)\varepsilon\frac{\Gamma_R}{4k}$$

$$\begin{pmatrix}J & L & I\\ j & m & -j-m\end{pmatrix}^2 D^{L*}_{pm}(\mathbf{k})D^{L}_{p'm}(\mathbf{k}'). \tag{17}$$

The amplitude of the Rayleigh scattering is determined from the general formula (7). The quantity $|M_{coh}|^2$ can also be written conveniently in the form (10), where the coefficients A and B defined by formulas (10), (16), and (17) take the form

$$A=A_1+A_2, \tag{18}$$

$$A_1=\frac{1}{2}a^2\sum_{\nu\nu'}(4\nu+1)(4\nu'+1)\begin{pmatrix}L & L & 2\nu\\ 1 & -1 & 0\end{pmatrix}\begin{pmatrix}L & L & 2\nu'\\ 1 & -1 & 0\end{pmatrix}$$

$$\times\begin{pmatrix}L & L & 2\nu\\ m & -m & 0\end{pmatrix}\begin{pmatrix}L & L & 2\nu'\\ m & -m & 0\end{pmatrix}P_{2\nu}(\cos\theta)P_{2\nu'}(\cos\theta'), \tag{19}$$

$$A_2=i(G-G^*), \tag{20}$$

where

$$G=Zr_0a\sum_{\mu\nu\nu'}(-1)^{\mu+m}(2\nu+1)(2\nu'+1)\begin{pmatrix}L & 1 & \nu\\ 1 & -1 & 0\end{pmatrix}\begin{pmatrix}L & 1 & \nu'\\ 1 & -1 & 0\end{pmatrix}$$

$$\times\begin{pmatrix}L & 1 & \nu\\ m & -\mu & \mu-m\end{pmatrix}\begin{pmatrix}L & 1 & \nu'\\ -m & \mu & m-\mu\end{pmatrix}D^{\nu}_{0\,m-\mu}(\theta\varphi)D^{\nu'}_{0\,\mu-m}(\theta'\varphi'),$$

$$a=\frac{(2L+1)(2I+1)}{2J+1}\frac{\Gamma_R}{\Gamma}\frac{\lambda}{2\pi}\begin{pmatrix}J & L & I\\ j & m & -j-m\end{pmatrix}^2 \tag{21}$$

In (19) and (21), θ, φ and θ', φ' denote the spherical angles of the vectors $\mathbf{k}$ and $\mathbf{k}'$ in a coordinate system having a z axis parallel to the direction of the magnetic field acting on the nucleus. In (21), the indices ν and ν' are even for an electric transition and odd for a magnetic transition. The coefficient B is determined from the formula

$$B = {}^1/_2(G + G^*), \tag{22}$$

and the coefficient C from formula (12).

The use of formulas (18)–(20) in the case of an M1 transition leads to the rather simple results:

$$A = \frac{a^2}{8(1+|m|)^2}[1+(-1)^{m+1}\cos^2\theta][1+(-1)^{m+1}\cos^2\theta'] + \frac{a}{8}Zr_0\sin\theta\sin\theta'\sin m(\varphi'-\varphi), \tag{23}$$

$$B = \frac{a}{8(1+|m|)}Zr_0[\cos 2\alpha - (1-|m|)\cos\theta\cos\theta']. \tag{24}$$

3. Bragg Scattering of Resonant γ Rays by Antiferromagnetic Crystals

Belyakov and Aĭvazyan[5] were the first to call attention to the possibility of using Bragg scattering of resonant γ rays for the determination of the structure made up by the crystal magnetic fields at the lattice nuclei. Physically this is connected with the dependence of the amplitude of the Mossbauer scattering on the direction of the magnetic field acting on the resonant nucleus. If the periods of the magnetic and crystal structures are different, then additional, purely nuclear Bragg maxima should appear in the diffraction of resonant γ rays, just as in the case of neutron diffraction by magnetically-ordered crystals. Smirnov et al.[1] noted, and demonstrated experimentally with hematite as an example, that even when the magnetic and crystal structures coincide, new maxima can appear in directions corresponding to the extinction of the Rayleigh scattering. Under the special experimental conditions of[1], the nuclei of each antiferromagnetic sublattice scatter γ quanta of definite circular polarization, so that the waves scattered by the two sublattices do not interfere with each other and make independent contributions to the intensity of the new diffraction maximum. We note also that in both cases noted above there is no Rayleigh scattering at all in the new magnetic diffraction maxima. Such a singularity of magnetic diffraction produces favorable conditions for the investigation of certain effects connected with purely nuclear scattering by single crystals[1,6].

In this section of the paper we consider, for concreteness, Bragg scattering of resonant γ rays by antiferromagnetic crystals of the collinear type. We assume also that each antiferromagnetic cell contains two atoms, at which the magnetic fields have opposite directions. (The results can be generalized to the case of a complex magnetic cell by the standard method[11].) As stated above, it is necessary here to distinguish between two cases.

A. If the magnetic and crystal unit cells are identical, then purely nuclear maxima can appear along the directions corresponding to the extinction of the Rayleigh scattering. Since the structure factor of the Rayleigh scattering is equal to zero along these directions, we have in accordance with the general formula (3)

$$\left(\frac{d\sigma}{d\Omega}\right)_{\text{coh}} = \frac{1}{2}\frac{(2\pi)^3}{V_0}\frac{|N_\uparrow - N_\downarrow|^2}{(E-E_0)^2+\Gamma^2/4}\delta(\mathbf{k}-\mathbf{k}'-2\pi\tau_{\text{ext}}), \tag{25}$$

where τ_{ext} is the vector, in reciprocal-lattice space, corresponding to extinction of the Rayleigh scattering, and indices $\uparrow\downarrow$ are introduced to denote the amplitudes of resonant scattering by nuclei at which the magnetic fields have opposite directions.

B. If the magnetic cell is larger than the crystal cell, then additional maxima, connected with the increased period of the magnetic lattice, can appear on the diffraction pattern. In this case we can show that

$$\left(\frac{d\sigma}{d\Omega}\right)_{\text{coh}} = \frac{1}{2}\frac{(2\pi)^3}{V_{0(\text{mag})}}\frac{|N_\uparrow - N_\downarrow|^2}{(E-E_0)^2+\Gamma^2/4}\delta(\mathbf{k}-\mathbf{k}'-2\pi\tau_{\text{mag}}), \tag{26}$$

where $V_{0\text{mag}}$ is the volume of the magnetic unit cell and τ_{mag} are the reciprocal-lattice vectors of the magnetic structure, which do not coincide with the reciprocal-lattice vectors of the crystal structure.

Thus, in both cases the coefficients B and C in (10) are equal to zero, and the coefficient A is determined from the quantity $|N_\uparrow - N_\downarrow|^2$. We have:

$$|N_\uparrow - N_\downarrow|^2 = \left[\frac{(2L+1)(2I+1)}{2J+1}\frac{\Gamma_R}{2k}\begin{pmatrix} J & L & I \\ j & m & -j-m\end{pmatrix}^2\right]^2 \times \sum_{\nu\nu'}(4\nu+1)(4\nu'+1)\begin{pmatrix} L & L & 2\nu \\ m & -m & 0\end{pmatrix}\begin{pmatrix} L & L & 2\nu' \\ m & -m & 0\end{pmatrix} \times \left\{\begin{pmatrix} L & L & 2\nu \\ 1 & -1 & 0\end{pmatrix}\begin{pmatrix} L & L & 2\nu' \\ 1 & -1 & 0\end{pmatrix} P_{2\nu}(\cos\theta)P_{2\nu'}(\cos\theta) - \left[\frac{(2\nu-2)!(2\nu'-2)!}{(2\nu+2)!(2\nu'+2)!}\right]^{1/2}\begin{pmatrix} L & L & 2\nu \\ 1 & 1 & -2\end{pmatrix} \times \begin{pmatrix} L & L & 2\nu' \\ 1 & 1 & -2\end{pmatrix} P^2_{2\nu}(\cos\theta)P^2_{2\nu'}(\cos\theta')\right\}, \tag{27}$$

where $P^2_{2\nu}(\cos\theta)$ is the associated Legendre polynomial.

The use of formula (27) in the case of a M1 transition yields

$$N_\uparrow - N_\downarrow|^2 = \frac{9}{4}\left(\frac{\lambda}{2\pi}\frac{2I+1}{2J+1}\Gamma_R\right)^2 \begin{pmatrix} J & 1 & I \\ j & m & -j-m\end{pmatrix}^4 \frac{|m|}{8}(\cos^2\theta \div \cos^2\theta$$

4. Mossbauer Diffraction Spectrum. Allowance for Absorption in the Scatterer

Let us now calculate the Mossbauer diffraction spectrum. This question was already considered in[8], where, however, the absorption of the primary and secondary beams inside the scatterer, which is known to greatly influence the intensity and the form of the diffraction spectrum, were actually not taken into account.

It is convenient to represent the differential diffraction cross section[3,4,10] in the form

$$\left(\frac{d\sigma}{d\Omega}\right)_{\text{coh}} = \left(\frac{d\sigma}{d\Omega}\right)_\infty\left[\frac{A'+B'x}{1+x^2}+1\right], \tag{29}$$

where $x = 2(E - E_0)/\Gamma$, $A' = A/C$, $B' = B/C$, $(d\sigma/d\Omega)_\infty$ is the differential diffraction cross section far from resonance $E - E_0 \gg \Gamma/2$.

Let the incident beam of the γ quanta (emission line) have a natural width Γ with a center of gravity shifted from the resonant energy E_0 by an amount $\Delta E_0 + E_0 v/c$, where v is the Doppler velocity and ΔE_0 is a certain energy shift existing at zero Doppler velocity. Then, when account is taken of the absorption of the primary and secondary beams inside the scatterer, the diffraction intensity can be represented in the form

$$I(y) = \text{const} \cdot \int \left(\frac{A' + B'x}{1+x^2} + 1 \right) \left(\frac{1 - \exp\{-2\mu_x L \operatorname{cosec} \alpha\}}{2\mu_x \operatorname{costc} \alpha} \right) \frac{dx}{1+(x+y)^2}, \quad (30)$$

where $y = 2(\Delta E_0 + E_0 v/c)/\Gamma$, L is the scatterer thickness, μ_x is the absorption coefficient of the γ quanta:

$$\mu_x = \mu_e + \mu_n/(1+x^2), \quad (31)$$

μ_e is the coefficient of electron absorption, and μ_n is the coefficient of nuclear absorption at resonance.

It is impossible to obtain an exact expression for I(y) in the general case. However, by approximating the function

$$1 - \exp\left\{ - \frac{2\mu_n L \operatorname{cosec} \alpha}{1+x^2} \right\},$$

by the method proposed in[12,13] for the calculation of Mossbauer absorption spectra, we can readily obtain a fairly exact expression for I(y) in a wide range of variation of the effective scatterer thickness $L_{eff} = 2\mu_n L \operatorname{cosec} \alpha$. According to[12]

$$1 - \exp\left(- \frac{L_{eff}}{1+x^2} \right) \approx \frac{K(L_{eff})\varkappa(L_{eff})}{\varkappa^2(L_{eff}) + x^2}, \quad (32)$$

where

$$K(L_{eff}) = L_{eff} e^{-L_{eff}/2} [I_0(L_{eff}/2) + I_1(L_{eff}/2)], \quad (33)$$

$$\varkappa(L_{eff}) = \frac{K(L_{eff})}{1 - e^{-L_{eff}/2} I_0(L_{eff}/2)} - 1;$$

I_0 and I_1 are the zeroth- and first-order Bessel functions of an imaginary argument. The use of the approximation (32) and of the inequality $\mu_c \ll \mu_n$, which is usually satisfied in the experiments, yields

$$I(y) = \text{const} \cdot \int \frac{x^2 + B'x + A' + 1}{[1+(x+y)^2][\varkappa^2(L_{eff}) + x^2]} dx. \quad (34)$$

From the experimental point of view it is convenient to measure the quantity $W(y) = [I(y) - I(\infty)]/I(\infty)$, which takes the form

$$W(y) = \frac{\varkappa + 1}{\varkappa} \frac{A' + 1 - \varkappa^2 + B'y}{(\varkappa+1)^2 + y^2}, \quad (35)$$

where $\kappa = \kappa(L_{eff})$ determined from formula (33). Thus, the coefficients in the formula for the differential cross section can be determined directly from the Mossbauer diffraction spectrum.

We note that W(0), calculated from formula (35), is exact, independently of the employed approximation (32)

$$W(0) = (A' + 1 - \varkappa^2)/\varkappa(\varkappa+1). \quad (36)$$

Consequently, the quantity $A' = A/C$ can be determined accurately from the intensity of the diffraction at resonance. It is also interesting to note that at a certain critical scatterer thickness, when

$$\varkappa^2(L_{eff}) = A' + 1, \quad (37)$$

the Mossbauer spectrum represents the dispersion curve

$$W(y) = B'y / [(\varkappa+1)^2 + y^2]. \quad (38)$$

This critical scatterer thickness does not depend on the contribution of the interference scattering (B'). According to (33) and (37), the larger the ratio of the intensity of the nuclear-resonance and Rayleigh scatterings (A'), the larger this thickness. This interesting fact was observed in some experiments (see, for example,[1,14]), which serve by the same token as a convincing confirmation of the existence of interference between the Rayleigh and nuclear-resonance scatterings.

We note in conclusion that when account is taken of electronic absorption, formulas (35), (37), and (38) do not change qualitatively, and in place of $\kappa(L_{eff})$ it is necessary to introduce some other quantity that depends on the electronic-absorption coefficient[12]. In addition, the use of the approximation (32) makes it possible to generalize the calculation to include the case when the emission and absorption lines have widths larger than the natural width Γ.

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Translated by J. G. Adashko
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