

Emission of gamma quanta from a crystal in the case of hyperfine splitting

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The γ radiation produced upon decay of an excited nucleus of the Mössbauer type located in a crystal containing the same nuclei, but in the ground state, is considered. The influence of the hyperfine splitting on the radiation intensity, both in the emitting nucleus and in the crystal nuclei, is taken into account. The problem is solved by means of the reciprocity theorem, which permits the use of results previously obtained for an external γ quantum source. The influence of the type of nuclear transitions and of the nature of the hyperfine splitting on the intensity is considered. Special attention is paid to the suppression of inelastic channels of the nuclear reaction in this case. By way of example, the case of a ferromagnet is considered in detail.

1. INTRODUCTION

In an earlier study^[1] we considered the emission of γ quanta produced in the decay of an excited nucleus of the Mössbauer type in an ideal crystal consisting of the same atoms, but in the ground state. We investigated in that study the influence of the effect of suppression of the inelastic channels of the nuclear reaction (SE) on the angular and frequency distributions of the γ quanta emitted from the crystal. It turned out that the emission from a thick crystal takes place in the presence of the SE only along surfaces of cones with axes along the reciprocal-lattice vector and with aperture angle $90^\circ - \theta_B$ (θ_B is the Bragg angle). The character of the angular distribution of the intensity turns out to differ greatly, depending on whether the radiating nucleus is in a site or in interstice, and it is this which makes it possible to analyze the position of the nucleus in the unit cell.

The problem was solved by using the solution for an external source of photons incident in the form of a plane wave on an ideal crystal (see the paper of Kagan and Afanas'ev^[2]), and also by establishment of a corresponding reciprocity theorem that makes it possible to connect both solutions. It should be noted that the emission of resonant photons from a crystal was investigated also in an earlier paper by the author from the point of view of the time dependence of the intensity of the radiation emitted from the crystal^[3].

In the foregoing papers it was assumed for simplicity in the analysis of the resonant interaction of the protons with the nuclei that there is no hyperfine splitting. On the other hand, the hyperfine splitting has a strong influence on the SE. The appropriate analysis for the scattering of γ radiation by a crystal was recently carried out by Afanas'ev and Kagan^[4] (see also^[5]). They developed a general dynamic theory of the diffraction of γ quanta that interact resonantly with crystal nuclei under hyperfine splitting conditions, and analyzed the condition for complete realization of the SE.

The present paper is devoted to the emission of Mössbauer γ quanta from a crystal with decaying nuclei that undergo hyperfine interaction. To this end, we first generalize the reciprocity theorem to include the case of hyperfine splitting. Then, using the results of the earlier studies^[4,5], we obtain a general relation for the intensity of the radiation from the crystal and consider the influence of the SE on the intensity of the emitted radiation.

2. RECIPROCITY THEOREM

Assume that radiation sources are located at some point r_ξ in space ($\xi = 1, 2$); then the field E_ξ produced by the action of one of the sources is determined with the aid of Maxwell's equation

$$(k^2 - \omega^2/c^2)E_\xi^i(k, \omega) + k^i k E_\xi^i(k, \omega) = \frac{4\pi i \omega}{c^2} \left(\sum_{k'} \sigma_{\omega}^{ii}(k, k') E_\xi^i(k', \omega) + J_\xi^i(k, \omega) \right). \quad (2.1)$$

Here J_ξ is the current corresponding to the source at the point r_ξ , and the quantity $\sigma_{\omega}^{i'l}$ describes the scattering of the γ quanta. We assign the equation with $\xi = 1$ to the source (excited nucleus) located inside the crystal, and $\xi = 2$ to the source far enough from the crystal.

Since the current J_1 in (2.1) is determined by the current operator averaged over the quantum mechanical state and the statistical distribution, we can write

$$J_1(k, \omega) = J_{01}(k, \omega) \exp[-ikr_1 - Z(k)/2]. \quad (2.2)$$

In this expression, $\exp[-Z(k)/2]$ characterizes the amplitude of the Mössbauer-effect probability, i.e., the emission or absorption of the γ quantum without excitation of the phonons, while $J_{01}(k, \omega)$ is the current of the rigidly secured nucleus.

The first term in the right-hand side of (2.1) describes the scattering of the γ quanta, and $\sigma_{\omega}^{i'l}$ can be represented in the form

$$\sigma_{\omega}^{ii}(k, k') = \sum_m \sigma_{\omega m}^{ii}(k, k') \exp[-i(k' - k) \cdot r_m]. \quad (2.3)$$

The summation is carried out here over all the atoms in the crystal. $\sigma_{\omega m}^{i'l}$ is a complex quantity proportional to the amplitude for scattering by an individual atom, and in the general case it has an arbitrary ratio of the imaginary and real parts, in other words, of the absorption and the scattering. $\sigma_{\omega m}^{i'l}$ depends principally on the hyperfine structure of the ground and excited states of the nucleus, a structure due mainly to the magnetic dipole and electric quadrupole interactions. Henceforth, to simplify the notation, we shall omit the subscript ω of $\sigma_{\omega}^{i'l}$.

With an aim towards using a quantum mechanical relation for the direct and inverse scattering amplitude, we make the substitution $k \rightarrow -k$ in Eq. (2.1) with $\xi = 2$ (source outside the crystal), and also reverse the signs of the projections of the angular momenta of the ground and excited states of the nucleus on the direc-

tion of the magnetic field (M_0 and M , respectively) and of the effective magnetic field H that produces the hyperfine splitting, i.e.,

$$M \rightarrow -M, \quad M_0 \rightarrow -M_0, \quad H \rightarrow -H. \quad (2.4)$$

The quantities contained in those Maxwell's equations in which this substitution is made will be designated by a minus sign, while the quantities in the initial equations will be marked by a plus sign. We multiply Eq. (2.1) with $\xi = 1$, without the indicated substitution, by $E_{2-}^i(-\mathbf{k}, \omega)$, and the equation with $\xi = 2$ with the substitution by $E_{1+}^i(\mathbf{k}, \omega)$. Then, summing over \mathbf{k} and subtracting one equation from the other, we obtain

$$\sum_{\mathbf{k}, \mathbf{k}'} [E_{2-}^i(-\mathbf{k}, \omega) \sigma_{+}^{il}(\mathbf{k}, \mathbf{k}') E_{1+}^i(\mathbf{k}', \omega) - E_{1+}^i(\mathbf{k}, \omega) \sigma_{-}^{il}(-\mathbf{k}, \mathbf{k}') E_{2-}^i(\mathbf{k}', \omega)] = - \sum_{\mathbf{k}} [J_{1+}(\mathbf{k}, \omega) E_{2-}(-\mathbf{k}, \omega) - J_{2-}(-\mathbf{k}, \omega) E_{1+}(\mathbf{k}, \omega)]. \quad (2.5)$$

In the second term on the left, we make the substitution $\mathbf{k}' \rightarrow -\mathbf{k}'$ followed by $\mathbf{k} \rightleftharpoons \mathbf{k}'$ and $i \rightleftharpoons l$; we then obtain

$$- \sum_{\mathbf{k}, \mathbf{k}'} E_{2-}^i(-\mathbf{k}, \omega) \sigma_{-}^{il}(-\mathbf{k}', -\mathbf{k}) E_{1+}^i(\mathbf{k}', \omega).$$

We now use the well known relation for the scattering amplitudes (see, e.g., [6]).

$$\sigma_{m+}^{il}(\mathbf{k}, \mathbf{k}') = \sigma_{m-}^{il}(-\mathbf{k}', -\mathbf{k}). \quad (2.6)$$

Since σ_{m}^{il} is proportional to the elastic-scattering amplitude, the initial and final states of the nucleus coincide as a result of the interaction with the γ quantum; thus, we can dispense with the replacement of the final state of the nucleus by the initial state, a replacement required by the quantum-mechanical reciprocity theorem. As a result, the left-hand side of (2.4) vanishes. We then obtain

$$\sum_{\mathbf{k}} J_{1+}(\mathbf{k}, \omega) E_{2-}(-\mathbf{k}, \omega) = \sum_{\mathbf{k}} J_{2-}(-\mathbf{k}, \omega) E_{1+}(\mathbf{k}, \omega). \quad (2.7)$$

This is the final expression, and now we must change from the Fourier components of the field to the coordinate representation. We are interested principally in the decay of the nucleus in the direction corresponding to the Bragg condition. In the problem for the external source, the incidence of γ quanta with wave vector $\kappa_0 = -\mathbf{k}_0$ inside the crystal will correspond to a superposition of two waves with wave vectors κ_0 and $\kappa_1 = \kappa_0 + \mathbf{K}$, where \mathbf{K} is the reciprocal-lattice vector. In this case, the transition to the coordinate representation can be carried out in exactly the same manner as in [1]. When considering an external source with definite polarization s , we must use the solution for E_{2-} corresponding to this polarization s . Omitting the subscript s from now on, we obtain directly

$$I(\mathbf{k}_0, \omega) = \xi \sum_{\alpha, \beta} \exp\left(\frac{-Z(\kappa_\alpha) - Z(\kappa_\beta)}{2}\right) \mathbf{E}_{\alpha-}(\mathbf{r}_1) \mathbf{E}_{\beta+}(\mathbf{r}_1) I_{\alpha\beta}^{il}, \quad (2.8)$$

$$I_{\alpha\beta}^{il} = \langle (J_{01}^i(\kappa_\alpha, \omega))_{M_0 M_0} (J_{01}^{i*}(\kappa_\beta, \omega))_{M_0 M_0} \rangle.$$

The symbol $\langle \dots \rangle$ denotes here the sum over the spin state of the ground level and averaging over the spin state of the excited level. The indices $\alpha, \beta = 0, 1$ (denote the numbers of the wave vectors κ_0 and κ_1 , while ξ is a constant independent of ω).

It is easy to separate in the current contained in (2.8) the resonant factor

$$J_{01}^i(\kappa, \omega) = \frac{j^i(\kappa) \Gamma/2}{\omega - \omega'(M, M_0) - i\Gamma/2}. \quad (2.9)$$

Here $\omega'(M, M_0)$ is a well-known quantity that deter-

mines the position of the written level as a function of the character of the hyperfine splitting. The matrix elements of the transition currents for the multipolarities E1, M1 and E2 are respectively [5]

$$(\hat{j}_1(\mathbf{k}))_{M_0 M} = \left[\frac{3}{4} (2I+1) \frac{c^2 \Gamma_1}{\omega} \right]^{1/2} \sum_{q=0, \pm 1} (-1)^q \begin{pmatrix} I_0 & 1 & I \\ -M_0 & q & M \end{pmatrix} \mathbf{n}_{-q}, \quad (2.10)$$

$$(\hat{j}_1(\mathbf{k}))_{M_0 M} = \left[\frac{3}{4} (2I+1) \frac{c^2 \Gamma_1}{\omega^3} \right]^{1/2} \sum_{q=0, \pm 1} (-1)^q \begin{pmatrix} I_0 & 1 & I \\ -M_0 & q & M \end{pmatrix} [\mathbf{k} \times \mathbf{n}_{-q}], \quad (2.11)$$

$$(\hat{j}_1(\mathbf{k}))_{M_0 M} = \left[\frac{5}{2} (2I+1) \frac{c^2 \Gamma_1}{\omega^3} \right]^{1/2} \sum_{m=-2}^2 (-1)^m \begin{pmatrix} I_0 & 2 & I \\ -M_0 & m & M \end{pmatrix} N_{-m}^{(2)}. \quad (2.12)$$

Here

$$N_{-m}^{(2)} = (-1)^m \sqrt{5} \sum_{q, q'=0, \pm 1} \begin{pmatrix} 1 & 1 & 2 \\ q & q' & m \end{pmatrix} (\mathbf{k} \mathbf{n}_{q'}) \mathbf{n}_q,$$

$\mathbf{q} = M_0 - M$, \mathbf{n}_0 is the used vector along the magnetic field, $\mathbf{n}_{\pm 1} = \mp(\mathbf{n}_x \pm i\mathbf{n}_y)$, and $\mathbf{n}_{x,y}$ are two mutually perpendicular vectors, both perpendicular to \mathbf{n}_0 ; the remaining symbols are standard. In quadrupole splitting, \mathbf{n}_0 is directed along the axial-symmetry axis of the electric-field gradient tensor.

3. EMISSION FROM CRYSTAL

The motion of the γ quanta in the crystal (in other words, the radiation field) is determined by the well known system of equations (see [5]) containing the dynamic coefficients $g_{\alpha\beta}^{il}$:

$$g_{\alpha\beta}^{il} = -g_0 \sum_p A_{\alpha}^i(p) A_{\beta}^{i*}(p) \frac{\Gamma/2}{\omega - \omega(M, M_0) + i\Gamma/2}. \quad (3.1)$$

In this expression, p is the aggregate of indices M, M_0 and J , determining the position of the nucleus in the unit cell;

$$g_0 = \frac{4\pi\eta}{\kappa^3 V_0} \frac{2I+1}{2(2I_0+1)} \frac{\Gamma_1}{\Gamma}, \quad (3.2)$$

$$A_{\alpha}(p) = \frac{2 \exp[Z_j(\kappa_{\alpha})/2]}{c[\Gamma_1(2I+1)]^{1/2}} (\hat{j}_1(\kappa_{\alpha}))_{M_0 M} \exp[i\kappa_{\alpha} \mathbf{R}_j]. \quad (3.3)$$

Here \mathbf{R}_j is a vector describing the position of the nucleus in the unit cell. Thus, the coefficients $g_{\alpha\beta}^{il}$ contain the same product of transition currents as $\frac{il}{\alpha\beta} \mathbf{k}$.

To determine the fields that enter in the reciprocity theorem, we must ascertain the effect of the substitution (2.4) on the SE. Let us see first what changes occur in the coefficients $g_{\alpha\beta}^{il}$. We note first that the same analysis applies to the transitions E1 and M1; if we make the substitution $\mathbf{E} \rightarrow \mathbf{H}$, then we can easily show that the coefficients $g_{\alpha\beta}^{il}$ in the equations for the electric and magnetic fields respectively, coincide for both transitions. Using (2.9), we can easily find in the case when $\exp(i\mathbf{K} \cdot \mathbf{R}_j) = 1$ (see [5])

$$g_{\alpha\beta}^{il} = g_0 \sum_j \sum_{M_0^j M_0^j} \begin{pmatrix} I & 1 & I_0 \\ -M^j & q & M_0^j \end{pmatrix} \frac{T_j^{il}(M^j, M_0^j) \Gamma/2}{\omega - \omega(M^j, M_0^j) + i\Gamma/2}, \quad (3.4)$$

$$T_j^{il} = \delta_{\alpha} \delta_{\beta} n_{0j}^i n_{0j}^{j+1/2} \delta_{\alpha, -1} (\delta^{il} - n_{0j}^i n_{0j}^j - i\epsilon^{ilk} n_{0j}^k) + 1/2 \delta_{\alpha, -1} (\delta^{il} - n_{0j}^i n_{0j}^j + i\epsilon^{ilk} n_{0j}^k). \quad (3.5)$$

The subscript j takes into account the fact that in the general case the hyperfine-splitting field can be different for different nuclei in the unit cell.

It is seen immediately from (3.4) and (3.5) that for transitions with $q = 0$, the value of $g_{\alpha\beta}^{il}$ remains unchanged when the signs of \mathbf{n}_0 and of the momentum projections are reversed. On the other hand, in transitions with $q = \pm 1$ (if there is no corresponding degen-

eracy of the levels with respect to the signs of the projections of the angular momenta), the direction of the elliptic polarization is reversed. In the case of degeneracy with respect to the times of the angular momenta (quadrupole splitting) there are no terms linear in n_0 at all, and $g_{\alpha\beta}^{il}$ retains the same form.

In the case of the E2 transition, no terms linear in n_0 appear, and the $g_{\alpha\beta}^{il}$ are invariant to the transformation (2.4). Violation of the invariance is possible in the case of interference between the transitions M1 and E2, for in this case there appear terms with odd powers of n_0 . Usually, however, the admixture of the other transition is small, and we shall not consider this effect here.

So far we have considered only nuclear resonant scattering. If elastic scattering by the electrons is present simultaneously, then $g_{\alpha\beta}^{il}$ acquires an additive term (see^[5]) connected only with scattering by electrons. Obviously, in this case, under the transformation (2.4), the resultant quantity will not change, since the scattering by the electrons does not depend on H , M_0 , and M .

We note that the interference of two inelastic processes, the photoeffect and conversion^[7] for transitions of the E1 type, which usually appears in the total scattering cross section, is reflected also in the amplitude of the elastic scattering of the γ quanta, i.e., in the coefficient $g_{\alpha\beta}^{il}$ (for details see^[3]). However, a direct analysis shows that even this part of the scattering amplitude is invariant to the transformation (2.4).

Thus, the coefficients $g_{\alpha\beta}^{il}$, which determine the field inside the crystal, are not altered by the transformations $H \rightarrow -H$, $M \rightarrow -M$, and $M_0 \rightarrow -M_0$, except that the sign of the elliptic polarization is reversed in the cases of E1 and M1.

We can now examine the intensity of the radiation from the crystal. Let at first the Bragg conditions not be satisfied. In this case the radiation field produced by external incidence of γ quanta attenuates exponentially with increasing depth. For the radiation from the crystal, this corresponds, as seen from the reciprocity theorem, to an intensity proportional to $e^{-\mu l}$, where $\mu = \kappa_0 \text{Im} \frac{1}{g_{00}^{ii}}$ is the usual absorption coefficient and l is the effective depth of the radiating nucleus. In the general case, however, a situation is also possible in which γ quanta of definite polarization do not interact at all with the nuclei. This occurs if the vector $A_\alpha(p)$ (see (3.3)) is perpendicular to the corresponding polarization vector, as a result of which the nuclei in the crystal are not excited by radiation with this polarization. This causes the radiation with this polarization not to appear when the nuclei decay. Indeed, this follows directly from the fact that $I_{\alpha\beta}^{lk}$ in (2.8) is determined by the same product of currents as $g_{\alpha\beta}^{il}$. We note that in the case of decay in an arbitrary direction, far from the Bragg condition, the position of the radiating nucleus in the unit cell does not influence the intensity at all.

We now consider decay in a direction k for which the Bragg condition is exactly satisfied; assume that complete SE is realized for external incidence of γ quanta in the direction $-k$. As is well known, the SE is characterized by the fact that there is produced in the crystal a coherent wave superposition for which the amplitude for the formation of an excited nucleus vanishes at the lattice site, whereas in the interstice this amplitude is different from zero. Using the re-

ciprocity theorem, we can conclude that no non-absorbing superposition of the waves will be produced as a result of the decay of the nucleus located at the lattice site, and the radiation will be exponentially small. At a small deviation from the exact Bragg condition, a weakly-absorbing superposition of waves begins to be generated already with finite intensity, but the absorption is still small and only the indicated wave superposition will emerge from a sufficiently large depth of the crystal (for details see^[1]).

We note that if the detector is tuned to one allowed hyperfine-structure line, then we have for the intensity of the radiation from the crystal the same result, in the sense of the angular and frequency distributions, as for the unsplit line^[1].

On the other hand, if the radiating nucleus is located at an interstice, then the result is radically altered. Since in this case the amplitude for the production of the excited nucleus is large precisely for the nonabsorbed superposition of the wave, the intensity of the radiation following decay of interstitial nuclei will be maximal when the Bragg condition is exactly satisfied. This result is a direct manifestation of the fact that the decay of the nucleus is accompanied by generation of a superposition of waves for which the SE is effective and which is thus weakly absorbed. In the case of deviation from the exact Bragg conditions, the intensity decreases with increasing angle to the value characteristic of the absence of the SE.

In the case when the Bragg condition is satisfied, a situation is also possible in which there is no interaction at all between nuclei and γ quanta of definite polarization. As a result, radiation with the corresponding polarization will not be emitted. It is clear that this statement remains valid regardless of the position of the nucleus in the unit cell.

So far we have not spelled out concretely the values of the hyperfine splitting in the radiating nucleus and in the nuclei of the crystal from which the scattering takes place. For a radiating nucleus in the regular position, the splitting, naturally, coincides with the splitting in the crystal, but for interstitial nuclei, in the general case, the hyperfine splitting may turn out to be different. Assume that the Bragg condition is satisfied and the splitting in the crystal is large. If none of the lines of the crystal coincides with the lines in the source, then we have the interaction of the quanta only with the electrons of the atomic shell (the Cossel effect). The case when the emission line of the decaying nuclei is not split and coincides with one line or with a group of two or three lines in the crystal, is the most favorable for the observation of the radiation from the crystal, for in this case the SE always exists (see^[4,5] for details). If, to the contrary, the hyperfine splitting in the radiating nucleus is large, and in the crystal the line is unsplit and coincides with one of the lines of the source, then it can be easily seen that this case reduces to that described earlier^[1].

Let us dwell in greater detail on a ferromagnet with one resonant nucleus per unit cell. For simplicity we assume that the hyperfine splitting does not depend on the position of the nucleus in the unit cell, and choose a reflection with $\exp(i\mathbf{K} \cdot \mathbf{R}) = 1$. We consider a nuclear transition of the E1 type (it was shown earlier that the analysis is similar for M1 transitions). Let at first the detector be tuned to a line with $M - M_0 = 0$. This

transition corresponds to scattering without a change of the polarization of the quantum, and the tensor is given by $g^{il} \sim n_0^i n_0^l$. This corresponds to a situation in which waves with polarization vector $\mathbf{e}_\alpha \perp \mathbf{n}_0$ do not interact at all with the crystal, and complete SE is realized for waves of the other polarization. In the case of radiation from the crystal, the tensor $g_{\alpha\beta}^{il}$ is not altered by the transformation (2.4), and no radiation will be observed at all in a narrow interval near the Bragg angle for a decaying nucleus at a site. For an interstitial nucleus, the γ quanta with non-interacting polarization will likewise not be emitted, and the γ quanta of the other polarization will be weakly absorbed near the Bragg angle.

We now consider an E1 transition with $M - M_0 = \pm 1$. We choose a reflection such that the effective field at the nucleus is directed perpendicular to the scattering plane—along the z axis. We then have $\mathbf{A}_\alpha \sim n_x \pm iny$ (see (3.3)). Waves with polarization \mathbf{e}^σ (\mathbf{e}^σ is perpendicular to the scattering plane) do not interact at all with the nuclei of the crystal, meaning that radiation of this polarization is not emitted from the crystal. The radiation of the other polarization, $\mathbf{e}_\alpha^\pi = \mathbf{e}^\sigma \times \kappa_0 / \kappa_0$, interact resonantly with the nuclei, but, as shown by Kagan and Afanas'ev^[5], SE takes place for this polarization. Thus, when the γ quanta are incident at the Bragg angle, the radiation passes through the crystal without absorption. Inasmuch as in this case the scattering proceeds with a change of the γ -quantum polarization, a superposition of the direct and scattered waves produces at the lattice site a field with circular polarization, and at other points of the unit cell the field polarization will in general be elliptic and will vary from point to point (for more details see^[5]), the direction of location of the polarization vector being dependent on the sign of $M - M_0$. If we make the substitution (2.12), then the direction of the circular polarization of the field at the lattice site is reversed, and it can be shown that $\mathbf{E}_2 \sim n_x \mp iny$. Let us determine the current corresponding to the decay of a radiating nucleus. The current of interest to us, obviously, is the complex conjugate of the current contained in the definition of \mathbf{A}_α (see (3.3)) and corresponding to excitation of the nucleus and $\mathbf{J}_1 \sim n_x \mp iny$.

We note that it is precisely in transitions with $\Delta M = \pm 1$ that the specific features of the influence of SE on

the character of the radiation from the crystal become most clearly manifest. The amplitude of the radiation field of the nucleus (a quantity proportional to \mathbf{J}_1) does not vanish, since the field from the external source of the nucleus is \mathbf{E}_2 , but the product $\mathbf{J}_1 \cdot \mathbf{E}_2$ is the amplitude of the production of a non-absorbed superposition of the waves during the decay of the nucleus, and is strictly equal to zero for a radiating nucleus at a lattice site. Thus, radiation with polarization \mathbf{e}^π (see (2.7)) is not emitted from the crystal. On the other hand, if the radiating nucleus is located at an interstice, then \mathbf{J}_1 , naturally, retains its form, and \mathbf{E}_2 varies with the position of the nucleus in the unit cell, so that the product $\mathbf{J}_1 \cdot \mathbf{E}_2$ does not vanish, i.e., the non-absorbed superposition will be radiated from the crystal with finite intensity.

Thus, in spite of the more complicated situation when account is taken of the hyperfine splitting, it can be concluded that if the SE is produced when the photons are incident from the outside, then it is realized also in the case of radiation from the crystal.

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