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# *THE INVESTIGATION OF NUCLEAR QUADRUPOLE INTERACTION IN EXPERIMENTS ON THE MÖSSBAUER EFFECT*

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Certain kinematic effects in resonance absorption and scattering are considered which occur when there is nuclear quadrupole interaction. Methods are proposed for determining all the characteristic parameters of quadrupole interaction from Mössbauer spectra of resonance absorption and scattering in single and polycrystals. These methods are convenient to apply even when the quadrupole interaction is small and only causes a broadening of the Mössbauer line.

## INTRODUCTION

THE study of nuclear quadrupole interaction is an interesting branch of investigations using the Mössbauer effect. Unfortunately, as yet we have not succeeded in using the experimental data for a complete determination of all the characteristic parameters of the quadrupole interaction (such as the quantity $eQ\varphi_{zz}$, the asymmetry parameter for the electric field $\eta = (\varphi_{xx} - \varphi_{yy})/\varphi_{zz}$ and the orientation of the principal axes of the electric field gradient tensor (e.f.g)). This situation is connected with the fact that in the majority of experiments, which are usually on the principal Mössbauer isotopes ($Fe^{57}$, $Sn^{119}$, $Te^{125}$, etc) that have $\gamma$ transitions with $I_b = 3/2 \rightarrow I_a = 1/2$, the only source of information about the quadrupole interaction is the separation between components of the Mössbauer spectrum. This defect of the experiments done so far has obviously seriously limited possible application of the Mössbauer effect to the study of atomic and crystal structure.

In the present paper we note certain kinematical effects in the resonance absorption and scattering of $\gamma$ quanta and suggest a method for determining the characteristic quantities for this interaction.

## DEVIATION OF GENERAL FORMULAS

The Hamiltonian describing the nuclear quadrupole interaction can be written in the form

$$\hat{H} = \frac{eQ\varphi_{zz}}{4I(2I-1)}[3I_z^2 - I(I+1) + \eta(I_+^2 + I_-^2)], \quad (1)$$

where I and Q are the spin and quadrupole moment of the nucleus, and $I_\pm = I_x \pm iI_y$. In the case of an axially symmetric field ($\eta = 0$) the quadrupole interaction gives rise to a splitting of the nuclear level into sublevels with definite values of $|I_z|$. The sublevel states are doubly degenerate.

In the general case $0 < \eta < 1$, it follows from (1) that the split sublevels do not have definite values of $I_z$. Each degenerate state of a sublevel is a superposition of states with definite values of the spin projection:

$$\psi_j{}^i = \sum_{m_j} A_{m_j}{}^i \psi_{m_j}, \quad (2)$$

where j labels the degenerate state, i labels the sublevel. In the case of half-integral spin, j takes on two values and the subscripts $m_j$ differ from one another by two units.

Let us assume for simplicity that there is no quadrupole interaction in the ground state. Consider a $\gamma$ transition $I_b \rightarrow I_a$. If for the excited level $eQ\varphi_{zz} > \Gamma$, the resonance absorption (emission) line is a multiplet, for which the intensities of the individual components depend on the orientation of the wave vector **k** of the photon in the system of the principal axes of the e.f.g. tensor. It is not difficult to see that the cross section for resonance absorption of the i'th component of the multiplet has the form

$$f^i(\mathbf{k}) = \mathrm{const} \sum_{\substack{p=\pm 1 \\ m_{aj}}} \Big[ \sum_{m_{bj}} A^i_{m_{bj}} C(I_a L I_b; m_a, m_{bj} - m_a) D^L_{m_{bj} m_a p}(\mathbf{k}) \Big]^2, \quad (3)$$

where C is a Clebsch-Gordan coefficient and D is a generalized spherical function. For specific calculations formula (3) is conveniently written in the form

$$f^i(\mathbf{k}) = \text{const} \sum_{m_{aj}} \sum_{m_{bj} m_{bj}'} (-1)^{m_{bj}-I_b} A^i_{m_{bj}} A^i_{m_{bj}'}$$
$$\times \sum_{\nu \text{ even},} C(LL\nu; 1, -1)\, C(I_b I_b \nu; -m_{bj}, m_{bj}')$$
$$\times W(I_b I_a \nu L; L I_b) D^{\nu}_{m_{bj}'-m_{bj}, 0}(\mathbf{k}),$$

where the W's are Racah coefficients, $0 \leq \nu \leq \min(2I_b, 2L)$. Let us consider resonance scattering. Using the usual methods of the theory of $\gamma - \gamma$ correlations, one can show that the angular dependence of the cross section for resonance scattering of the i-th component of the multiplet has the form

$$F^i(\mathbf{k}, \mathbf{k}') = \text{const} \sum_{jj'} \sum_{m_{bj} m_{bj}'} \sum_{m_{bj'} m'_{bj'}} (-1)^{m_{bj}+m_{bj}'-2I_b}$$
$$\times A^i_{m_{bj}} A^i_{m_{bj}'} A^i_{m_{bj'}} A^i_{m'_{bj'}} \sum_{\nu \text{ even}} C(LL\nu; 1, -1)$$
$$\times C(I_b I_b \nu; -m_{bj} m_{bj'})\, W(I_b I_a \nu L; L I_b) D^{\nu}_{m_{bj'}-m_{bj}, 0}(\mathbf{k})$$
$$\times \sum_{\nu' \text{ even}} C(LL\nu'; 1, -1)\, C(I_b I_b \nu'; -m'_{bj'} m'_{bj})$$
$$\times W(I_b I_a \nu' L; L I_t) D^{\nu'}_{m'_{bj} - m'_{bj'}, 0}(\mathbf{k}'),$$
$$0 \leqslant \nu, \nu' \leqslant \min(2I_b, 2L), \tag{4}$$

where the subscripts $m_{bj'}$ run through the same values as $m_{bj}$ in (1) and (2).

If the direction of the $\gamma$ quantum coincides with the z axis, $D^{\nu}_{m_{bj'}-m_{bj}, 0} = \delta_{m_{bj'} m_{bj}}$, and formula (4) gives a clear picture of the angular distribution of scattered $\gamma$ quanta $F^i(k_i)$ in the system of the principal axes of the e.f.g. tensor.

The formulas given are valid only for a single crystal. For a polycrystal the resonance absorption and scattering cross sections for the quadrupole components are obtained by averaging (3) and (4) over the orientation of the system of principal axes of the tensor.

## THE CASE OF $\gamma$ TRANSITIONS $^3\!/_2 \rightarrow {}^1\!/_2$

Let us consider the most common case in experiments on the Mössbauer effect, corresponding to a $^3\!/_2 \rightarrow {}^1\!/_2$ transition. Quadrupole interaction is absent in the ground state. The excited state is split into two sublevels with energies $\pm(eQ\varphi_{zz}/4)\sqrt{1 + \eta^2/3}$, and these sublevels are doubly degenerate. Their wave functions are easily found from perturbation theory:

$$E^1 = \frac{eQ\varphi_{zz}}{4}\sqrt{1 + \eta^2/3} \qquad \begin{cases} \psi_1{}^1 = \lambda(\psi_{3/2} + A\psi_{-1/2}) \\ \psi_2{}^1 = \lambda(\psi_{-3/2} + A\psi_{1/2}) \end{cases},$$
$$E^2 = -\frac{eQ\varphi_{zz}}{4}\sqrt{1 + \eta^2/3} \qquad \begin{cases} \psi_1{}^2 = \lambda(\psi_{1/2} - A\psi_{-3/2}) \\ \psi_2{}^2 = \lambda(\psi_{-1/2} - A\psi_{3/2}) \end{cases}, \tag{5}$$

where $A = (\eta/\sqrt{3})(1 + \sqrt{1 + \eta^2/3}$, while $\lambda$ is a normalization factor. Thus in the general case ($\eta \neq 0$) the $\gamma$ line is a doublet with a separation of the components equal to $\frac{1}{2} eQ\varphi_{zz}\sqrt{1 + \eta^2/3}$. It is obvious that a measurement by the Mössbauer effect of just one such separation cannot determine all the parameters and the orientation of the principal axes of the e.f.g. tensor.

Let us consider the angular dependence of the resonance absorption cross section for the two components of the doublet. In our case it follows from (3) that

$$f^{(1)}(\theta\varphi) \sim \left(1 + \frac{A^2}{3}\right)(1 + \cos^2\theta) + \frac{4}{3}A^2 \sin^2\theta$$
$$+ \frac{2A}{\sqrt{3}} \sin^2\theta \cos 2\varphi,$$
$$f^{(2)}(\theta\varphi) \sim \left(\frac{1}{3} + A^2\right)(1 + \cos^2\theta) + \frac{4}{3}\sin^2\theta$$
$$- \frac{2A}{\sqrt{3}} \sin^2\theta \cos 2\varphi, \tag{6}$$

where $\theta$ and $\varphi$ are the spherical angles of the vector k in the system of the principal axes of the e.f.g. tensor. For $\eta = 0$ ($A = 0$) we have the simple formulas for an axially symmetric field:

$$f^{(1)}(\theta) \sim 1 + \cos^2\theta, \quad f^{(2)}(\theta) \sim {}^5\!/_3 - \cos^2\theta. \tag{7}$$

Comparison of (6) and (7) shows that any asymmetry of the electric field sensitively influences the angular dependence of the absorption cross sections of the doublet components. Of particular interest is the azimuthal dependence of the cross sections for resonance absorption of the two components of the doublet, where the effect is proportional to $\eta$, i.e., becomes important even for small $\eta$.

From the experimental point of view it is more convenient to investigate the relative absorption of the two components, i.e., $f^{(1)}/f^{(2)} = a$. As an example, we take $\eta = 0.5$, $\theta = \pi/2$; then a varies from $a = 0.46$ when $\varphi = \pi/2$ to $a = 0.8$ when $\varphi = 0$. Thus, by studying the azimuthal dependence of the relative intensity of the components of the doublet in the Mössbauer absorption spectrum in a single crystal (the $\gamma$ emission line is single), we can determine the asymmetry parameter $\eta$ and the directions of the principal axes of the e.f.g. tensor. Then the quantity $eQ\varphi_{zz}$ can be determined from the spacing (5) of the components. The sign of $eQ\varphi_{zz}$ is easily established from the relative position of the two components of the doublet in the Mössbauer spectrum.

The resonance absorption spectrum in a polycrystal does not give sufficient information for determining all the characteristic parameters of the quadrupole interaction.

We consider resonance scattering. Formula (4) now becomes

$$F^{1,2}(\mathbf{kk}') = \text{const}\left[1 + A^2 \pm \frac{1}{2}(1 - A^2)D_{00}{}^2(\mathbf{k}) \pm \frac{A}{\sqrt{2}}(D^2_{-20}(\mathbf{k}) + D_{20}{}^2(\mathbf{k}))\right]\left[1 + A^2 \pm (1 - A^2)D_{00}{}^2(\mathbf{k}') \pm \frac{A}{\sqrt{2}}(D^2_{-20}(\mathbf{k}') + D_{20}{}^2(\mathbf{k}'))\right]. \tag{8}$$

If the direction of the incident $\gamma$ quantum is along the z axis, (8) will have the form

$$F^1(\theta'\varphi') = \text{const}\,(3 + A^2)[3 + 5A^2 + 3(1 - A^2)\cos^2\theta' + 2\sqrt{3}A\sin^2\theta'\cos 2\varphi'],$$

$$F^2(\theta'\varphi') = \text{const}(1 + 3A^2)[5 + 3A^2 - 3(1 - A^2)\cos^2\theta' - 2\sqrt{3}A\sin^2\theta'\cos 2\varphi']. \tag{9}$$

Here the effect of the azimuthal dependence is even more clearly apparent. Thus by rotating a single crystal absorber sample around the direction of the $\gamma$ quantum (the z axis) we can, from the dependence of the relative intensities of the Mössbauer doublet components, determine A (or $\eta$). The resonance scattering method has other advantages:

1) If the characteristic quantity for the quadrupole interaction, $eQ\varphi_{zz}$, is small, but $eQ\varphi_{zz} > \Gamma$, so that the Mössbauer line is not split, but only noticeably broadened, we can determine $\eta$ from the azimuthal dependence of the total cross section for resonance scattering:

$$F(\theta'\varphi') = F^{(1)}(\theta'\varphi') + F^{(2)}(\theta'\varphi'); \tag{10}$$

$F^{(1)}(\theta'\varphi')$ and $F^{(2)}(\theta'\varphi')$ are defined in (9). For a thin scatterer $F(\theta'\varphi')$ may be assumed to be proportional to the area under the curve in the Mössbauer spectrum. When the inequality $eQ\varphi_{zz} > \Gamma$ is not satisfied, the total resonance scattering cross section still depends on the ratio of $eQ\varphi_{zz}$ and $\Gamma$, and can be calculated in the same way as in the theory of $\gamma - \gamma$ correlation. But in this case the azimuthal dependence is less sensitive, and therefore is not of interest.

2) The spectrum for resonance scattering in a single crystal is a markedly asymmetric doublet. Therefore, when $eQ\varphi_{zz}$ is small and the Mössbauer spectrum becomes a single broadened line, this quantity can be determined precisely from the shift of the center of gravity of the spectrum (if the parameter $\eta$ is already known). Thus, even when the quadrupole interaction is small, all its characteristics can be determined completely.

3) The cross section for resonance scattering in a polycrystal depends on the angle between the directions of the incident and scattered $\gamma$ quanta. If we assume that the probabilities for the Mössbauer effect in the single crystal, f, f′ are independent of the direction of propagation of the $\gamma$ quantum, the cross section for resonance scattering of the doublet components in the polycrystal is obtained by simply averaging (8) over the orientations of the system of principal axes of the tensor:

$$F^{1,2}(\vartheta) = \text{const}\,[1 + 2A^2 + {}^1/_{20}P_2(\cos\vartheta) + {}^3/_{20}A^2P_2(\cos\vartheta)], \tag{11}$$

where $\vartheta$ is the angle between **k** and **k′**. On this assumption the two components of the doublet from resonance scattering have the same intensity. Expression (11) enables one to determine A (or $\eta$) by measurements on a polycrystal.

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[1] T. R. Das and E. L. Hahn, Nuclear Quadrupole Resonance Spectroscopy, Solid State Physics, Suppl. I, 1958.

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