Interference of hyperfine components in the emission spectrum of Mössbauer nuclei

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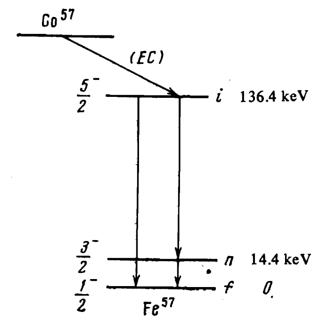
The emission spectrum of resonance γ quanta in a magnetic field is considered. It is shown that the hyperfine components in the emission spectrum interfere, and the interference can be observed by the $\gamma\gamma$-coincidence method. The possibility of isolating experimentally the interference contribution and separating the splitting caused by magnetic dipole and electric quadrupole interactions when they are present simultaneously is discussed. The necessity of taking this effect into consideration in Mössbauer experiments on the nonconservation of the T invariance of electromagnetic interactions is indicated.

Earlier[1] we considered the interference of hyperfine components during the resonance scattering of γ quanta by nuclei. It was shown that the interference terms in the scattering cross section have an appreciable magnitude and lead to an asymmetrical angular distribution. The interference of hyperfine components in the resonance emission spectrum is considered below.

Long-life isotopes, such as Co^{57}, Mn^{57}, Sn^{119}, etc., serve as sources of resonance γ quanta. The decay scheme of Co^{57} (see[2]) is shown in the figure. This decay is a cascade process. As a result of K capture, represented by the oblique arrow, the nucleus is found in the level i. Then a primary γ quantum $\mathbf{k}$ is emitted, the nucleus enters the first excited level n, and only then is a γ-quantum $\mathbf{k}'$ emitted, for which the resonant-emission probability is high because of its comparatively low energy.



Decay scheme of the Co^{57} isotope.

Let us assume that the nucleus is in a crystal and interacts with a magnetic field $\mathbf{H}$. This interaction leads to Zeeman splitting of the nuclear levels with a total angular momentum Y into $2Y + 1$ components characterized by a total angular-momentum projection M. We will not be interested in K capture and assume that the initial state of the system is $|Y_iM_i\rangle$ and has a width $2\Gamma_i$. We will regard the state $|Y_nM_n\rangle$ with width 2Γ as the intermediate state and $|Y_fM_f\rangle$ as the final state. The transitions $|Y_nM_n\rangle \to |Y_fM_f\rangle$ form the hyperfine structure of the Mössbauer line. De-excitation of the $|Y_iM_i\rangle$ level can proceed via the different sublevels $|Y_nM_n\rangle$ of the intermediate state, while the final states of the nucleus and of the γ-quanta give no information on the course the process. This indicates the possibility of interference of the hyperfine components in the emission spectrum. It is clear from the same considerations that the hyperfine components do not interfere if the momenta of the primary or secondary γ quanta are parallel to the magnetic field. The amplitude of a cascade transition with emission of two γ quanta is given by the formula[3]

$$A_{fi}=\sum_{M_n}\frac{V_{M_fM_n}V_{M_nM_i}}{(\omega'-\omega_{M_nM_f}+i\Gamma)(\omega+\omega'-\omega_{M_iM_f}+i\Gamma_i)}, \tag{1}$$

where $\omega_{M_nM_f}$ is the frequency of the $|Y_nM_n\rangle \to |Y_fM_f\rangle$ transition, $V_{M_fM_n}$ and $V_{M_nM_i}$ are the matrix elements of the emission operators of γ quanta with frequencies ω' and ω.

Let us point out that since the width Γ_i is much less than the frequency ω_i of the transition between the initial and intermediate states, we can set $\omega = \omega_i$ in the matrix element $V_{M_nM_i}$. To obtain the probability of emission of two photons we square the modulus of the transition amplitude. Assume that the energy of the primary γ quantum is not registered. Integrating the square of the modulus of the amplitude with respect to the frequency of the primary photon, we have

$$|A_{fi}|^2=\frac{\pi}{\Gamma_i}\sum_{M_nM_m}\lambda^{M_f}_{M_mM_n}V'_{M_fM_n}V_{M_nM_i}V'^{*}_{M_fM_m}V^{*}_{M_mM_i},$$
$$\lambda^{M_f}_{M_mM_n}\equiv[(\omega'-\omega_{M_nM_f}+i\Gamma)(\omega'-\omega_{M_mM_f}-i\Gamma)]^{-1}. \tag{2}$$

Since the wavelengths of the primary and secondary emissions are much larger than the dimensions of the nucleus, we assume in the calculation only magnetic dipole transitions between levels with different total angular momenta. In addition, we assume that the initial and final states of the nucleus are not recorded, and the γ detectors are not sensitive to polarizations. Under these assumptions the general formulas are all still quite cumbersome; therefore let us examine in detail only the special case of the decay of the widely used Mössbauer isotope Co^{57}, for which $Y_i = 5/2$, $Y_n = 3/2$, and $Y_f = 1/2$.

The averaging Eq. (2) over the initial states and summation over the final states of the nucleus, as well as the summation over the polarizations of the primary and secondary photons, are analogous to those used earlier[1]. Upon completion of these operations we find that the probability of detecting a primary photon in the direction $n \equiv \mathbf{k}/\omega$ and of a secondary photon in the direction $\mathbf{n}' \equiv \mathbf{k}'/\omega'$ with a frequency in the interval from ω' to $\omega' + d\omega'$ contains an interference term equal to

$$\mathrm{Int}\overline{|A|^2}=\alpha\,\mathrm{Re}[(n_1n_{-1}')^2(\lambda^{1/2}_{3/2-1/2}+\lambda^{-1/2}_{1/2-3/2})-2n_1n_0n_{-1}'n_0'(\lambda^{1/2}_{3/2\,1/2}+\lambda^{-1/2}_{-1/2-3/2})],$$
$$\alpha\equiv\frac{\pi^3\omega'\omega_i}{180\Gamma_i}\left(\frac{5}{2}\,\|\mu\|\,\frac{3}{2}\right)^2\left(\frac{1}{2}\,\|\mu\|\,\frac{3}{2}\right)^2, \tag{3}$$

where $n_{\pm1}$, n_0; $n'_{\pm1}$, n'_0 are the spherical components of the vectors **n** and **n**′ in a coordinate system with the z-axis directed along the external field **H**.

In Mössbauer emission spectrum studies, as a rule, one counter is used and there is no interest in the propagation direction of the primary photons of the cascade. This situation corresponds to an averaging of Eq. (3) over the emission directions of the primary photons. It is easy to see that such an averaging causes the interference term to vanish, and the resonance emission spectrum is the usual Zeeman sextuplet. Not all of the resonance quanta **k**′ have to be recorded to observe the interference of the hyperfine components in the emission spectrum, but only those which correspond to certain directions **k** of the primary photons, i.e., the $\gamma\gamma$-coincidence method must be used.

Equation (3) is simplified considerably if we set $\theta = \theta' = \pi/2$. This means that the photons whose wave vectors **k** and **k**′ lie in a plane perpendicular to the quantization axis are recorded. Transforming Eq. (3), we have

$$\mathrm{Int}\overline{|A|^2} = \frac{\alpha}{4}\left\{\frac{[(\omega'-\omega_1)(\omega'-\omega_2)+\Gamma^2]\cos 2(\varphi-\varphi')-\Gamma(\omega_2-\omega_1)\sin 2(\varphi-\varphi')}{[(\omega'-\omega_1)^2+\Gamma^2][(\omega'-\omega_2)^2+\Gamma^2]} + \frac{[(\omega'-\omega_3)(\omega'-\omega_4)+\Gamma^2]\cos 2(\varphi-\varphi')-\Gamma(\omega_4-\omega_3)\sin 2(\varphi-\varphi')}{[(\omega'-\omega_3)^2+\Gamma^2][(\omega'-\omega_4)^2+\Gamma^2]}\right\}. \quad (4)$$

The frequency ω_1 corresponds to the $|\,{}^3\!/_2\,-{}^1\!/_2\rangle \to |\,{}^1\!/_2\,{}^1\!/_2\rangle$ transition, ω_2 to the $|\,{}^3\!/_2\,{}^3\!/_2\rangle \to |\,{}^1\!/_2\,{}^1\!/_2\rangle$ transition, ω_3 to the $|\,{}^3\!/_2\,-{}^3\!/_2\rangle \to |\,{}^1\!/_2\,-{}^1\!/_2\rangle$ transition, and ω_4 to the $|\,{}^3\!/_2\,{}^1\!/_2\rangle \to |\,{}^1\!/_2\,-{}^1\!/_2\rangle$ transition. Equation (4) is analogous in its structure to that obtained in[1] for interference during scattering in a plane orthogonal to the external field. In studying the scattered radiation spectrum we must take into consideration the interference of the nuclear resonance channel and the Rayleigh channel. This effect is absent in our case being considered, so that the interference spectrum is somewhat simpler. In addition, a study of interference in the emission spectrum may be simpler for nuclei having a large conversion coefficient of the first excited state.

The interference term (4) changes sign when $\varphi' - \varphi$ is replaced by $\varphi - \varphi'$, if $\varphi' - \varphi = {}^1\!/_4\pi(1 + 2p)$ (p is an integer). This can be utilized to isolate the interference term in an experiment. Let $N(\varphi - \varphi')$ be the number of coincidences for counters oriented at angles of φ and φ'. Recognizing that the principal terms in the probability of the cascade transition (2) are independent of the azimuthal angles φ and φ', it is not hard to determine the relative magnitude of the interference contribution

$$\beta = \frac{N(\pi/4)-N(-\pi/4)}{N(\pi/4)+N(-\pi/4)}. \quad (5)$$

The effect considered above should appear not only in the presence of magnetic splitting but also in the presence of electric quadrupole splitting of the nuclear levels. In the case of the quadrupole Stark effect, however, degeneracy with respect to the sign of the projection of the angular momentum remains. As a result the interference term will be invariant to the transformation $\varphi' - \varphi \to \varphi - \varphi'$. This fact, on the one hand, prevents the use of (5) to determine the relative magnitude of the interference contribution and, on the other hand, makes it possible to separate from the experimental data the splittings associated with electric quadrupole and magnetic dipole interactions if they are present simultaneously.

In conclusion let us discuss another interesting aspect associated with the possibility of verifying the time parity of electromagnetic interactions in Mössbauer experiments[4-6]. In a discussion of these experiments[5] it was suggested that the asymmetry of the T-odd angular correlation be used to verify T-invariance and that the maximum of the right-left asymmetry occurs at the angles 45 and 135° in the plane perpendicular to the polarization direction of the nuclei. It should be pointed out that the maximum of the asymmetry introduced by interference of the hyperfine components occurs at these same angles. The asymmetry caused by interference is much larger than the asymmetry due to the assumed absence of T-invariance both for small and for large splittings of the resonance line within a fairly wide interval of splittings. This indicates that the interference of the hyperfine components must be taken into consideration in the appropriate experiments.

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