## Asymmetry of photon emission by <sup>57</sup>Fe nuclei in a Mössbauer transition

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The asymmetry of  $\gamma$ -photon emission from polarized <sup>57</sup>Fe nuclei in a 14.4-keV.Mössbauer transition was measured. The experiment was performed at source temperatures 300 and 500 K with immobile source and detector. The radiation of nuclei having a definite polarization at the instant of the transition was separated by using a resonant (Mössbauer) scintillation detector whose absorption line coincided with one of the split lines of the Mössbauer emission spectrum of an iron isomer located in a ferromagnetic nickel matrix. The quantity 2R, which is a measure of the violation of spatial parity in <sup>57</sup>Fe Mössbauer transitions and is equal to double the ratio of the reduced matrix element of the impurity E1 and regular M1 transitions, is found to be  $2R = (5.8 \pm 1.2) \cdot 10^{-4}$ .

Weak nucleon–nucleon interaction leads in nuclear processes to the appearance of effects connected with spatial parity violation.<sup>1-4</sup> In particular, mixing of multipoles of opposite parity in a gamma transition of an excited nucleus to the ground state can cause circular polarization of the radiation from an unpolarized source, or  $\gamma$ -photon emission that is asymmetric relative to the nuclear-spin direction. In the latter case the source must contain polarized excited nuclei. The customary quantitative mesure of spatial-parity violation in these phenomena is the quantity 2*R*, double the ratio of the reduced matrix elements of the impurity and regular multipoles.

We report here an investigation of the asymmetry of photon emission in a Mössbauer (14.4. keV) transition of  $^{57}$ Fe, an asymmetry due to mixing of a regular M1 multipole and an impurity P-parity-violating E1 multipole. To analyze in the experiment the radiation by nuclei in a definite polarized state, we used the Mössbauer-transition level splitting of the nucleus in a magnetic field, when each transition through the Zeeman sublevels corresponds to a definite energy of the emitted photons and definite components of the spins of the excited and ground states of the nucleus along the field direction.<sup>5,6</sup>

By energy-selective recording of the photons of one of these transitions, using a tuned detector having high selective sensitivity to Mössbauer photons, it is possible to separate the radiation by that part of the nuclear ensemble which has definite spin components along the quantization axis at the instant of transition. In other words, it is possible to use radiation from "polarized" nuclei without polarizing the entire ensemble.

It follows from Fig. 1 that the <sup>57</sup>Fe gamma transition of interest to us corresponds to six transitions between the Zeeman sublevels of the excited and ground states. Information on the "*P*-odd" effect, however, can be provided only by transitions whose spin component along the quantization axis change by an amount  $\Delta M = m_e - m_g = \pm 1$  ( $m_e$  and  $m_g$  are the nuclear spin components in the excited and ground state, respectively).

In fact, radiation with M 1 or E 1 multipolarity in these transitions is elliptically polarized if the photon emission angle is not perpendicular to the magnetic field. The electricfield vectors of these multipoles can be resolved into two orthogonal components whose oscillations differ in phase by  $\kappa/2$ . The phases of the oscillations of the corresponding components of the magnetic and electric multipoles differ in turn likewise by  $\kappa/2$ . The relative orientation of these components is shown in Fig. 2a. The expression for the intensity I of the resultant radiation contain squares of the sums of the orthogonal components of the two intermixing multipoles. The expression for I contains thus terms linear in the amplitude of the electric multipole, meaning also linear in R (except when the angle between the photon momentum and the magnetic-field direction is equal to  $\kappa/2$ ), where R = Q(E1)/Q(M1) is the ratio of the reduced matrix elements of intermixing multipoles.

The radiation corresponding to a transition with  $\Delta M = 0$  is always linearly polarized. In this case field vectors of the impurity and regular multipoles oscillate in mutually perpendicular planes (see Fig. 2b), the parity-violating increment to the emission intensity depends on Ronly quadratically. Note that for this transition the parity-violating effect linear in R is the circular polarization of the radiation.

Spatial parity violation in a nuclear radiative transition can be investigated in experiment by measuring the change produced in the intensity of the radiation-spectrum Zeeman component when the direction of the applied magnetic field is reversed. In the general case, as shown in Ref. 5, when interference takes place between the impurity E 1 and ground M 1 multipoles, such a radiation asymmetry  $\Delta I$  is



FIG. 1. Zeeman splitting of  $^{57}$ Fe Mössbauer transition. The thick line indicates the transition investigated.



FIG. 2. Relative arrangement of the electric-field vector components for radiation of E 1 and M 1 multipolarity:  $\mathbf{a} - \Delta M = \pm 1$ ;  $\mathbf{b} - \Delta M = 0$ .

described in first-order approximation by the equation  $\Delta I / I = -2R\xi_c$ , where *I* is the radiation intensity of the investigated Zeeman component,  $\xi_c (\Delta M = \pm 1) = 2\Delta M \cos \theta / (1 + \cos^2 \theta) = \Delta M \xi$  is the degree of the circular polarization of radiation of transitions with  $\Delta M = \pm 1$ , and  $\theta$  is the angle between the photon momentum and the quantization axis, and is set by the magnetic-field vector.

*Experiment*. To implement the quasipolarization method<sup>5</sup> we used the effective magnetic field of the hyperfine interaction between the radiating <sup>57</sup>Fe nucleus and the ferromagnetic electronic surrounding of the matrix material. This has made it possible to reverse the quantization-axis direction by remagnetizing the matrix with an electromagnet.

In precision measurements it is desirable to leave the experimental geometry unchanged and not to use the mechanical scanning, traditional in Mössbauer spectroscopy, of the gamma-radiation energy via the Doppler effect. We created therefore for this experiment a resonant pair consisting of "a source in a ferromagnetic matrix" and "a resonant scintillation detector." In this pair the Mössbauer resonance was effected at zero relative velocity.

The ferromagnetic matrix used for the source was a nickel foil into which parent radioactive <sup>57</sup>Co nuclei were introduced. The Mössbauer spectrum of this source was a well-resolved sextet of lines. The natural width of the internal lines, measured with a calibrated absorber of diferrocyanide of potassium and magnesium, was 0.14 mm/s. The effective magnetic field at the iron nuclei at room temperature was  $266 \pm 1.5$  kOe.

The radiation detector was a plastic scintillator containing absorbing matter based on iron phosphate. Resonant (Mössbauer) absorption of a gamma photon was revealed by production of conversion electrons. Such a detector has low sensitivity to non-Mössbauer gamma radiation, since the cross section for photoabsorption is much smaller than that for resonant absorption.

Figure 3a shows the Mössbauer spectra of the <sup>57</sup>Co (Ni) at temperatures 300 and 450 K, measured with a working resonant detector. The dashed line denotes the position of zero source velocity relative to the detector. Figure 3b shows in enlarged scale the  $-1/2 \rightarrow +1/2$  transition line of interest to us. It constitutes a weakly resolved doublet determined by the detector absorption line shape. The weak splitting of the detector line was due to hyperfine interaction of the iron nucleus with the electric field gradient of the electronic surrounding in the employed chemical compound.

It can be seen from the figures that the resonance of the source radiation line and of the detector absorption line (at



FIG. 3. Mössbauer radiation spectra of  $^{57}$ Fe in Ni matrix at temperatures 300 (1) and 450 K (2). Figure 3b shows the  $-1/2 \rightarrow 1/2$  transition line in enlarged scale.

V=0) is partial at room temperature and practically complete when the source is heated to 450 K. The reason for this is that the gamma-photon energy decreases and the emission line shifts to the right by 0.18 mm/s, both on account of the temperature red shift ( $\approx 60\%$  of the total shift) and as a result of the decrease of the effective magnetic field of the hyperfine interaction on heating.

The experimental setup is shown schematically in Fig. 4. The gamma-photon source 2 (the  ${}^{57}Co(Ni)$  sample) measured  $12 \times 8 \times 0.03$  mm and was glued to aluminum vessel 2 which equalized the source temperature. A heating element 4 was placed inside the vessel. On the outside the element was covered by asbestos thermal insuation, and on the inside by a reflecting thermal screen of metallized lavsan (mylar). The source temperature could be varied in the range 300– 470 K and was monitored by thermocouple 5.

The source unit was a light-weight assembly mounted during the preliminary investigations on the vibrator of the Mössbauer spectrometer. In the main experiment this unit was rigidly secured in the gap of electromagnet 6. The source surface and the field in the gap of the electromagnet were oriented 45° (or 135°) relative to the average direction to the detector. The electromagnet-winding current strength was stabilized and reversed every four seconds. This reversed periodically the field at the radiating iron nuclei, and the viola-



FIG. 4. Setup for the measurement of the radiation asymmetry:  $1-{}^{57}$ Co (Ni) sources; 2—aluminum vessel, 3—Be filter, 4—heater, 5—thermocouple, 6—electromagnet, 7—resonant scintillator, 8—hollow light pipe, 9—photomultiplier.

tion of the spatial parity caused a change in the intensity of the recorded line of the source Mössbauer spectrum.

The gamma radiation was recorded by resonant Mössbauer scintillator 7 mounted on hollow light pipe 8 of metallized lavsan. A solid light pipe of transparent material could not be used in view of the need to reduce to a minimum the background signal due to the Compton scattering and parasitic scintillations generated in the interior of the light pipe by the accompanying cobalt-source radiation of energy 122 and 136 keV. The intensity of this radiation exceeds by more than an order of magnitude the Mössbauer-transition radiation intensity. The photomultiplier was provided with a triple magnetic screen to protect it against the stray fields of the electromagnet.

We used in the experiment an integral (current) method of recording the  $\gamma$  photons, since no limit is imposed in it on the radiation intensity. We measured the photomultiplier anode-current dc and ac components *i* and  $\Delta i$  converted into voltages; the ac component contained the information on the sought *P*-odd effect. The employed recording procedure makes provision for separating the useful signal from the aditive noise due to the fluctuations of the total radiation intenstiy. This noise is a random function of the time and has a normal distribution.

The signal can be effectively separated from such a noise if the signal is periodic. In our experiment the useful periodic signal was the result of moduating the intensity signal by reversing the source magnetization every four seconds. This signal was amplified in the recording channel and stored after lock-in detection. The lock-in detector and the electromagnet current inverter were controlled by the same oscillator.

Operating in parallel was a control memory channel, indentical with the main one and governed by a reference voltage shifted  $\kappa/2$  in phase. The output voltage of this channel was determined only by the noise component of the detector signal.

The ac component  $\Delta i$  was calibrated by applying to the input of the recording channel a four-second step signal  $V_s$ . A voltage  $U_s$  was then accumulated in the main-channel integrator after a time  $T_s$ . The experimentally measured quantity  $2\Delta i/i$  is equal to the ratio of the ac and dc ( $U_{\perp}$ ) components of the electrometric-amplifier output signal. It is connected with the quantities known from the channel calibration, vis,  $V_s$ ,  $U_s$ , and  $T_s$ , the effect-accumulation time T, the value of  $U_{\perp}$ , and the integrator output voltage U by the simple relation

$$2\Delta i/i = V_{\rm c}T_{\rm c}U/U_{\rm c}TU_{\rm c},$$

and it was this which enabled us to determine the sought quantity.

Measurement of the  $\gamma$ -photon emission asymmetry. It was established in the course of the experiment that the sought P-odd signal was accompanied by, and in synchronism with, a signal from the  $\beta$  particles emitted by the radioactive impurities of the source and recorded by a  $\beta$ -particle scintillator. Naturally, the intensity of the electron flux incident on the detector depended on the direction of the applied magnetic field and was synchronized with its reversal. Furthermore, the  $\beta$ -electron ionization-loss energy in the scintillator was three-four times larger than the energy of the conversion electrons produced in the acts of resonant absorption of the Mössbauer photons. The synchronous noise from the  $\beta$  electrons was decreased by placing a beryllium plate 3 mm thick between the source and the electron; the plate absorbed a noticeable fraction of the electrons but attenuated the resonant Mössbauer gamma radiation insignificantly.

To separate the true *P*-odd  $\gamma$ -ray asymmetry from the background of the residual synchronous loss, we used the temperature dependence of the energy of the investigated line of the Mössbauer-spectrum source. The measurements were performed at 300 and 450 K under conditions of partial and exact resonance of the source and detector lines (see Fig. 3). Both the ac component  $\Delta i$  (containing the *P*-odd signal) and the dc component *i* of the detector signal were measured.

Obviously, a change in sample temperature affects only the efficiency with which the detector records the chosen Mössbauer  $\gamma$ -photon line. Therefore the expressions for the indicated quantities per unit flux intensity of 14.4-keV photons take the form

$$i(T) = w_{nr} + I_r f(T) w_r(T) + I_0,$$
  

$$\Delta i(T) = I_r f(T) w_r(T) \delta + \delta_0,$$

where  $W_{nr}$  is the probability of nonresonant recording, via the photoeffect, of the 14.4-keV  $\gamma$  photons by the detector;  $W_r(T)$  is the probability of resonant recording of the Mössbauer photons of the investigated Zeeman component of the spectrum, determined by the degree of overlap of the source and absorber lines;  $I_r$  is the fraction of this component in the total Mössbauer radiation; f(T) is the Debye-Waller factor of the radiation source;  $I_0$  is the signal from the accompanying radiation;  $\delta = 2\Delta I/I = 2 \cdot 2R |\bar{\xi}|$  is the sought *P*-odd effect produced when the source is remagnetized and equal to double the radiation asymmetry of the polarized nuclei;  $\bar{\xi}$  is the quantity  $\xi = 2 \cos \theta / (1 + \cos^2 \theta)$ , averaged over the radiation-beam aperture; and  $\delta_0$  is the synchronous noise. The difference between the ac and dc components of the detector signal at 300 and 450 K are given by  $i(450) - i(300) = I_r[f(450) w_r(450) - f(300) w_r(300)],$  $\Delta i(450) - \Delta i(300) = I_r[f(450) w_r(450) - f(300) w_r(300)]\delta.$ 

The ratio of these differences determines explicitly the sought-for value of  $\delta$ .

Measurements results. The observed change of the ac component of the detector signal following heating of the source to 450 K amounted to about 10%, i.e., the synchronous noise due to the  $\beta$  electrons was quite high. In addition, the contribution of the Mössbauer photons to the detector signal was  $\approx 1\%$  at 300 and  $\approx 2\%$  at 450 K. This is due not only to the presence of a high-power 122-keV accompanying radiation, but also to the fact that only one of the six Mössbauer spectrum lines was recorded,  $\approx 5-10\%$  of the resonant photons.

The measurements were performed at alternating temperatures. This decreased the influence of the decay of the source of  $\beta$ -active impurities and of the possible instabilities of the apparatus. A total of 58 measurement runs were made alternately at both temperatures. The durations of the runs were gradually increased to compensate for the source decay and to equalize the statistical weights of the results of all the measurement runs.

The data were reduced by the standard procedure, assuming a Gaussian distribution. A zero mean value of the *P*odd signal was obtained for the control channel, in which the lock-in detector operated with a  $\kappa/2$  phase shift. The value of the average statistical deviation from the mean turned out to be the same for both channels. The geometric factor  $\xi$  of the radiation beam was determined by numerical integration and found to equal  $0.78 \pm 0.05$ .

The resultant degree of spatial-parity violation in a Mössbauer transition of the <sup>57</sup>Fe nucleus was  $2R = (5.8 \frac{6}{1.2}) \cdot 10^{-4}$ .

The appreciably larger effect for <sup>57</sup>Fe and for the <sup>119</sup>Sn previously investigated by us,<sup>5</sup> compared with the earlier values observed for  $\gamma$  transitions of undeformed <sup>19</sup>F and <sup>41</sup>K nuclei, can be attributed to an enhancement mechanism connected with the nuclear structure. Indeed, the investigated M 1 transitions of <sup>57</sup>Fe and <sup>119</sup>Sn are strongly suppressed compared with the single-particle estimates of the probabilities of radiative transitions (by ~40 and ~80 times, respectively. Note that theoretical calculations performed for this transition by Platonov<sup>7</sup> with allowance for the contributions of the neutral currents yield a value  $2R \approx 3.8 \times 10^{-4}$ .

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