Mössbauer Effect of the Second Excited State of Fe^{57}†

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The Mössbauer effect of the 136.4-keV decay of the second excited state of Fe^{57} has been observed using a new scattering technique. The decay of the second excited state following resonant absorption to the first excited state was detected. A 6-mg/cm^2 Fe^{57} scatterer was used. The Mössbauer effect was unambiguously observed using 15 mCi of Co^{57} diffused into an iron host lattice as a source. With both source and scatterer at liquid-helium temperature, the recoilless fraction for an iron host lattice was estimated to be approximately 2×10^{-3}. Partially resolved magnetic hyperfine structure was observed using a 15-mCi source of Co^{57} diffused into palladium. Since the structure was only partially resolved, the sign of the magnetic moment was not determined with certainty. However, the assumption that the magnetic moment is negative led to agreement with the magnitude of the magnetic moment previously reported by Körner. The gyromagnetic ratio that was determined was $g=-0.39\pm0.04$.

INTRODUCTION

PREVIOUS Mössbauer-effect experiments have usually been performed by observing either the resonant absorption or the resonant scattering of recoilless gamma rays. An alternative technique has been to observe the decay of a level, following resonant absorption, by processes other than gamma emission to the ground state. This technique has an advantage over conventional absorption experiments in that the signal-to-noise ratio can be greater than the recoilless fraction. Both the electrons resulting from internal conversion,[1] and the x rays following internal conversion,[2,3] have been used in this way. The Mössbauer effect of the second excited state of Dy^{161} [4] has been observed by requiring coincidence between the two gamma rays which result from the decay of this level by way of the first excited state. In this experiment, the Mössbauer effect in the second excited state of Fe^{57} has been detected by observing the internal conversion of the first excited state following the decay of the second excited state.

Although the properties of Fe^{57} have been studied in great detail since the discovery of the Mössbauer effect in the 14.4-keV transition from the first excited state, the observation of recoilless transitions involving the second excited state at 136.4 keV (see Fig. 1) has not been reported previously. The observation of the Mössbauer effect of the second excited state, using the conventional techniques of scattering and absorption, is more difficult than observing the Mössbauer effect of the first excited state. This is because both the recoilless fraction and the resonant-absorption cross section of the former are very much smaller. Furthermore, the effective resonant-absorption cross section is comparable to the Compton and Rayleigh scattering cross sections.

The method used to perform this experiment is somewhat similar to the technique used to observe the Mössbauer effect in the second excited state of Dy^{161}. Following resonant absorption of a 136-keV gamma ray, the decay of the second excited state by way of the first excited state was detected. This was advantageous, because the second excited state decays in this way about 90% of the time.[5] However, coincidence between 14.4- and 122-keV gamma rays was not practical, because of the high count rate of 122-keV gamma rays produced in the Co^{57} source. Furthermore, very little of the 14.4-keV radiation produced in the scatterer would have been detected, because of the high resonant-absorption cross section for this radiation in the scatterer. Since the 14.4-keV level decays by internal conversion about 90% of the time,[5] and since the iron scatterer is thick to electrons, it was decided to detect the 6.4-keV K x rays following internal conversion. The 6.4-keV x rays produced in the Co^{57} source were filtered out by a nonresonant energy-dependent filter which reduced their intensity substantially below the level of K x rays produced in the scatterer. Using this technique, it was possible to observe the Mössbauer effect of the 136-keV transition and to observe partially resolved hyperfine structure. From this structure the magnetic moment was estimated.

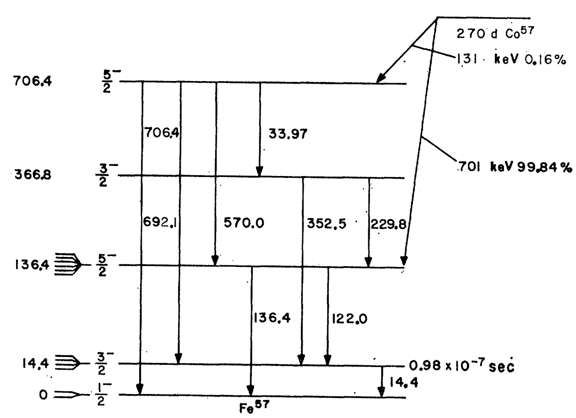

FIG. 1. Decay scheme of Co^{57}.

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[2] H. Frauenfelder, D. R. F. Cochran, D. E. Nagle, and R. D. Taylor, Nuovo Cimento **19**, 183 (1961).
[3] N. Hershkowitz, J. S. Eck, and J. C. Walker, Bull. Am. Phys. Soc. **10**, 577 (1965).
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EXPERIMENTAL CONSIDERATIONS

The details of the scattering geometry are given in Fig. 2. Both source and scatterer were cooled to liquid-helium temperature. This was necessary in order to increase the recoilless fraction to its maximum value of approximately 10^{-3}. A 6-mg/cm^2 Fe^{57} scatterer was clamped to the $\frac{1}{4}$-in. aluminum bottom plate of the liquid-helium container. The aluminum bottom plate served as the nonresonant energy-dependent filter which reduced the intensity of the incident 6.4-keV x rays and the 14.4-keV gamma rays to well below that produced in the scatterer. The 136-keV gamma rays were only attenuated to about 80% of their initial intensity in passing through the filter. This filter was necessary in order to prevent resonant scattering from taking place in the first excited state. Therefore, the 14.4-keV level of a nucleus in the Fe^{57} scatterer could only be excited as a result of the resonant absorption of a 136-keV gamma ray.

Mössbauer spectra were obtained using the constant acceleration method.[6] A loudspeaker-based velocity drive[7] was coupled to the cooled source through a double bellows[8] arrangement. Data were accumulated in a TMC Model 401 multichannel analyzer operating as a multiscaler. Background was subtracted from the data to remove the double parabola caused by moving the source relative to the detector, and to eliminate possible systematic effects in the data acquisition. Velocity calibrations were obtained by making measurements on the first excited state of Fe^{57} and comparing these measurements to the values of the hyperfine splitting of this level reported by Hanna *et al.*[9]

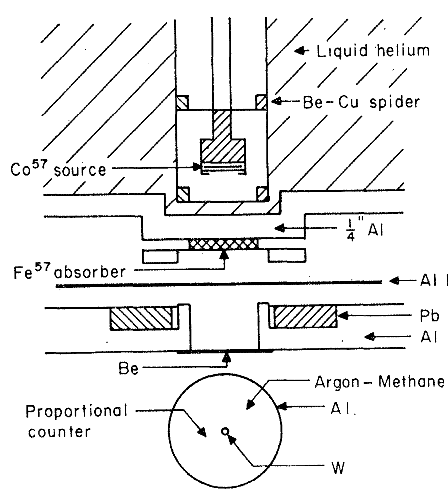

FIG. 2. Schematic drawing of the scattering geometry showing the Co^{57} source, helium container, bottom plate, Fe^{57} scatterer, and proportional counter.

Because of the difficulties inherent in this experiment, the size of the observed Mössbauer effect was not appreciably greater than the statistical uncertainty in the data. For this reason, several experiments were performed. First, iron metal was chosen as the host lattice for both source and scatterer nuclei. In such an environment, the second excited state is split by the magnetic hyperfine interaction into six sublevels. The Mössbauer spectra resulting from the use of both a split source and a split absorber was a broad peak with unresolved structure centered about zero veloctity. Secondly, in order to observe hyperfine structure, a metallic iron scatterer (split) was used in conjunction with a Co^{57} source diffused into a palladium host lattice.[10] These spectra were obtained at several maximum velocities, in order to verify that the apparent Mössbauer effects were real and that the peaks occurred at the same velocities in each case.

GENERAL DISCUSSION

The effective scatterer thickness for the ith absorption line of a scatterer having a thickness t and a recoilless fraction f is given by

$$T_i = W_i f n \sigma_0 (\Gamma_\gamma/\Gamma) t, \tag{1}$$

where n is the atomic density of Fe^{57}, σ_0 is the cross section, and W_i is the relative transition probability. The ratio[5] $\Gamma_\gamma/\Gamma = \frac{1}{8}$ is the fraction of decays from the second excited state which take place by gamma emission to to ground state.

The maximum intensity $I_{6.4\,\mathrm{res}}(i,j)$ of 6.4-keV K x rays resulting from resonant absorption of 136-keV gamma rays from the jth sublevel of the source with intensity I_{136} by the ith absorption line can be estimated from[3]

$$I_{6.4\mathrm{res}}(i,j) \approx \tfrac{1}{2} I_{136} f_s \omega_k \frac{T_i W_j \rho}{\mu_{6.4} t} (1 - e^{-\mu_{6.4} t}) E, \tag{2}$$

where f_s is the recoilless fraction of the source, ω_k is the K fluorescent yield ≈ 0.3 for iron, E takes into account losses related to attenuation of windows, geometric losses, and detector efficiency, W_j is the relative intensity of the jth emission line, ρ is the density of the scatterer, and $\mu_{6.4}$ is the atomic-mass absorption coefficient of the scatterer for 6.4-keV x rays. Equation (2) assumes that both source and absorber are thin to 136-keV gamma rays, and that the absorption lines are well separated. The factor of $\frac{1}{2}$ results from the assumption of equal source and absorber natural linewidths.

In this experiment, it was necessary to detect 6.4-keV x rays in the presence of a background of 122-keV gamma rays which had an intensity of about 10^5 relative

[6] G. K. Wertheim, *Mössbauer Effect: Principles and Applications* (Academic Press Inc., New York, 1964).
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[10] R. M. Bozorth, P. A. Wolff, D. D. Davis, V. B. Compton, and J. H. Wernick, Phys. Rev. **122**, 1157 (1961).

to that of the K x rays. Although lithium-drifted detectors have recently been designed to have resolutions the order of 1 keV at 6.4 keV, the noise produced by the high background obscures the signal. Scintillation counters having resolutions and relative insensitivity to 122-keV gamma rays comparable to proportional counters are not available. Therefore, a proportional counter was chosen as a detector.

The best signal-to-noise ratio was obtained using a 90% argon-10% methane gas mixture in a cylindrical flow counter having a 1-in. diameter. This detector had an efficiency of about 60% for 6.4-keV x rays. The most prominent features of the pulse-height spectrum observed with this counter were peaks at 6.4 and 3.2 keV and a broad background of noise. The peak at 6.4 keV was largely due to the characteristic K x rays produced in the scattering foil following photoelectric absorption of 122-keV gamma rays. The remainder of the noise was produced within the proportional counter. The peak at 3.2 keV corresponded to the characteristic K x rays of argon which were produced following photoelectric absorption of 122-keV gamma rays. The broad background of noise was produced by electrons resulting from the Compton scattering of 122-keV gamma rays. The count rate observed within the window set on the peak at 6.4 keV was approximately 10^3 counts/sec.

RESULTS

The Mössbauer spectra that were obtained, using an iron host lattice for both source and scatterer, are shown in Fig. 3. A rough estimate of the recoilless fraction made from the observed signal-to-noise ratio of the peak at zero velocity is 2×10^{-3}.

TABLE I. Relative intensities and energy separations of the transitions from the sublevels of the 136-keV level to the sublevels of the ground state.

Negative magnetic moment Relative intensity		Positive magnetic moment Relative intensity	
5	$5\Delta E_2+\Delta E_0$	5	$5\Delta E_2-\Delta E_0$
4	$3\Delta E_2+\Delta E_0$	1	$3\Delta E_2+\Delta E_0$
1	$3\Delta E_2-\Delta E_0$	4	$3\Delta E_2-\Delta E_0$
3	$\Delta E_2+\Delta E_0$	2	$\Delta E_2+\Delta E_0$
2	$\Delta E_2-\Delta E_0$	3	$\Delta E_2-\Delta E_0$

The magnetic hyperfine structure was seen by observing the Mössbauer spectra of an unsplit source (palladium host lattice) with a split scatterer (Fe^{57} foil). It was assumed that only magnetic hyperfine structure was present, as the quadrupole structure was expected to be negligibly small from results of first-excited-state measurements in Fe^{57}. The five pairs of lines of the magnetic hyperfine spectrum have relative intensities 5:4:1:3:2, as indicated in Table I. The observed spectra were not well resolved, and showed velocity broadening due to the geometry employed. Because of this and the small size of the observed effects, only the six lines with the highest relative intensities were fitted to the data. The energy spacing between the two outermost lines is equal in magnitude to $5\Delta E_2\pm\Delta E_0$, depending upon the size of the magnetic moment of the second ex-

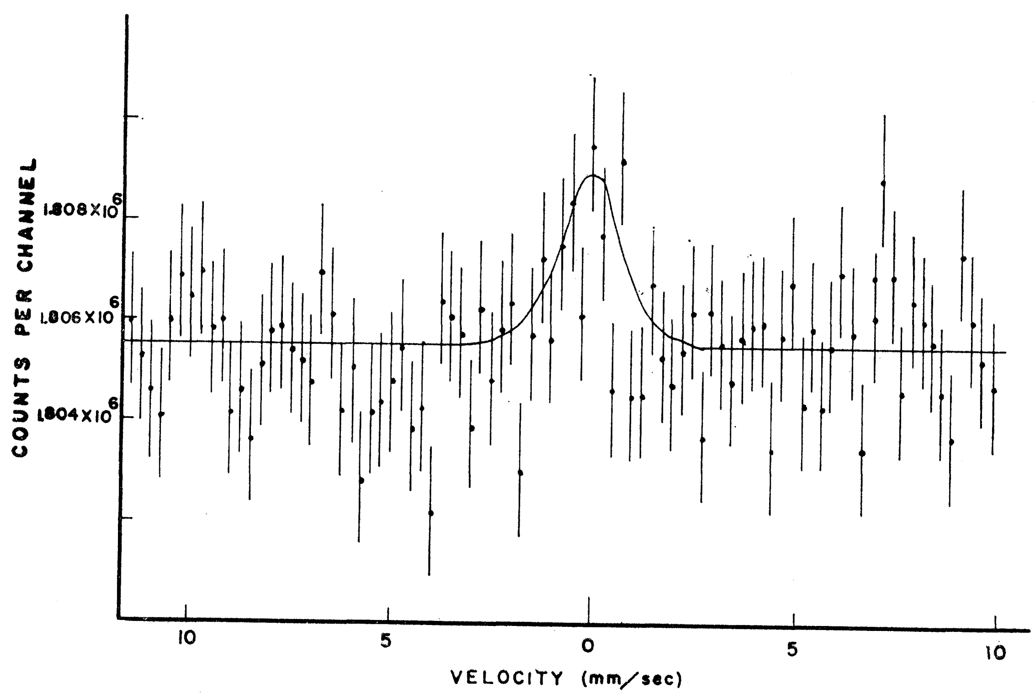

FIG. 3. Results of the split-split experiment showing peak at zero relative velocity. The source was 15 mCi of Co^{57} in an iron host lattice. The scatterer was Fe^{57}.

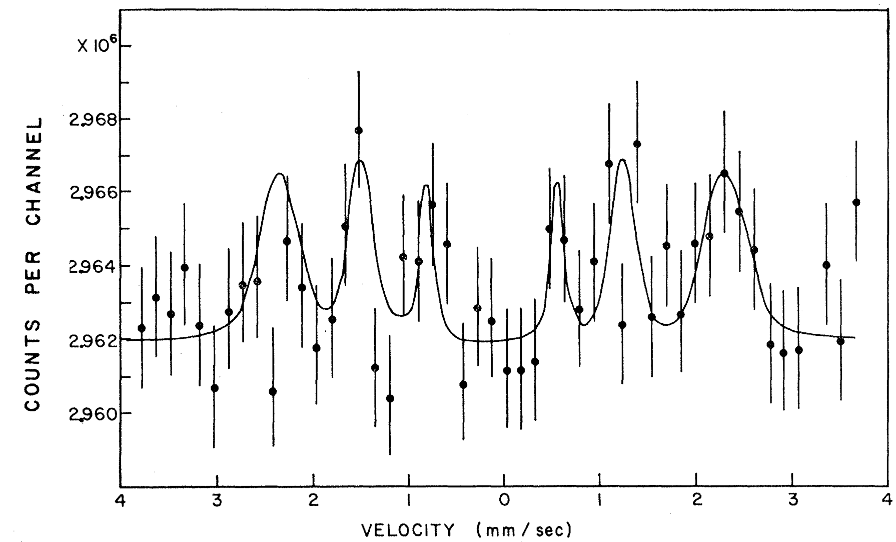

FIG. 4. Results of the first unsplit-split experiment. The source was 15 mCi of Co^{57} diffused into palladium. The scatterer was a 6 mg/cm² Fe^{57} foil.

cited state. The quantities ΔE_2 and ΔE_0 are the energy separations of the sublevels of the second excited state and the ground state, respectively. Here, $\Delta E_2 = g\mu_n H$, where g is the gyromagnetic ratio, μ_n the nuclear magneton, and H the effective magnetic field. The data show that ΔE_2 is greater than ΔE_0. The choice of the sign of g for the 136-keV state determines the order of the transitions in terms of decreasing energy. Table I indicates the energy separations of the pairs of lines for each choice of the sign of g for the 136-keV state.

The spectra shown in Figs. 4 and 5 were taken at different maximum velocities to check the consistency of the peak positions. The curves drawn through these data consisted of Gaussians whose heights and widths were determined to be consistent with the velocity broadening[11] and shifts that were present because of the geometry used in this experiment. The curves were specified by five parameters. They were ΔE_2, the height and width of one of the peaks (which took into account the velocity broadening), the zero-velocity point, and the number of counts corresponding to velocities far from resonance. The values of χ^2 determined for these fits are summarized in Table II. A negative magnetic moment for the 136-keV state gave a somewhat better fit than a positive value, and the curves in Figs. 4 and 5 are drawn for that choice of sign.

Because the statistical uncertainty in the data is comparable to the size of the Mössbauer effect, the χ^2 of the curves that have been fit to the data have been compared to the χ^2 of the mean. Only data points corre-

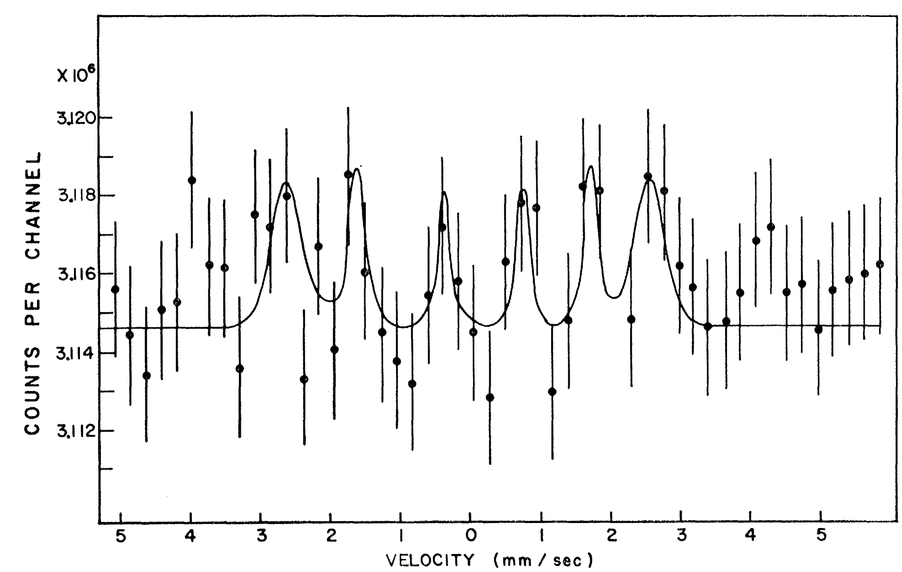

FIG. 5. Results of the second unsplit-split experiment. The maximum velocity was chosen to be greater than that which was used to obtain the results shown in Fig. 4.

[11] N. Hershkowitz and J. C. Walker (to be published).

TABLE II. χ^2 of the mean and χ^2 of the curves fit to the data for unsplit-split experiments. P is the probability of observing a greater value of χ^2.

	χ^2 of mean	Degrees of freedom	P	χ^2 of theoretical fit	Degrees of freedom	P
Figure 4	60	49	0.14	45	45	0.50
Figure 5	40	29	0.08	38	45	0.76

sponding to the range in velocities in which structure was observed have been included in the calculations of the χ^2 of the mean. The inclusion of the data points for which no structure was expected and for which the mean is a good fit would have reduced the significance of χ^2. The probability P of obtaining a χ^2 that was greater than or equal to the value obtained for each of these two sets of data was 0.50 and 0.76 for the curves fit to the data, compared to 0.14 and 0.08 for the mean as an assumed fit; therefore, the mean is not a good fit.

The results for a negative moment are summarized in Table III. Since ΔE_0 is known, the energy corresponding to the hyperfine splitting of the second excited state can be determined. Uncertainties in the positions of peaks have been combined with those of the velocity calibration to give the estimated uncertainties in ΔE_2. An average of these results was taken after multiplying each value by the reciprocal of the square of its uncertainty, and this resulted in $\Delta E_2 = 0.91 \pm 0.09$ mm/sec, which corresponds to $g_2 = -0.39 \pm 0.04$ for an assumed value of H of 333 kG. Using a similar analysis to the above, the choice of a positive moment results in a value of $g = +0.51 \pm 0.05$.

TABLE III. Results from curve fitted to data assuming a negative magnetic moment.

Theoretical peak separation	Figure 4 ΔE_2(mm/sec)	Figure 5 ΔE_2(mm/sec)
$5\Delta E_2 + \Delta E_0$	0.9 ± 0.1	1.0 ± 0.1
$3\Delta E_2 + \Delta E_0$	0.8 ± 0.1	1.1 ± 0.2
$\Delta E_2 + \Delta E_0$	1.0 ± 0.6	0.7 ± 0.7

The above results can be compared to those previously presented by Körner *et al.*[12] The preliminary value of the gyromagnetic ratio that was quoted for that experiment was $+0.366 \pm 0.018$. The magnitude of that result agrees with the magnitude of the gyromagnetic ratio determined in this experiment for a negative moment. Körner *et al.* used a differential perturbed angular-correlation technique. The results of that experiment for the magnitude of g are unambiguous. From the data presented, one can only speculate as to the correctness of the sign. We do not understand the discrepancy in sign between the two experiments.

CONCLUSION

The Mössbauer effect has been observed for the first time in the 136-keV transition from the second excited state of Fe^{57}. Estimates of the magnetic moment have been obtained.

As far as the technique is concerned, this experiment has extended the use of the Mössbauer effect to a nuclear level with both small resonant cross section and small recoilless fraction. For this experiment, $\sigma f \approx 10^{-22}$ cm^2. Because of the difficulties involved, it is unlikely that the Mössbauer effect will prove useful in measuring the properties of levels which have σf much smaller than this value.

[12] H. J. Körner, J. Braunsfurth, H. F. Nesemann, S. J. Skorka, and B. Zeitnitz, in *Comptes Rendus du Proceedings of the Congrès International de Physique Nucléaire, II*, edited by P. Gungenberger (Centre National de Recherche Scientifique, Paris, 1964).