PHYSICAL REVIEW VOLUME 129, NUMBER 3 1 FEBRUARY 1963

$E2/M1$ Mixing Ratio of the 123-keV Transition in Fe^{57} Determined from a Mössbauer Coincidence Experiment

H. DE WAARD*
Laboratory of Physics, University of Groningen, Groningen, Netherlands

AND

F. VAN DER WOUDE
Institute for Crystal Physics, University of Groningen, Groningen, Netherlands

(Received 12 September 1962)

For gamma cascades $(2) \xrightarrow{\gamma_2} (1) \xrightarrow{\gamma_1} (0)$ in nuclei where the separate magnetic components of the ground-state transition $(1) \to (0)$ can be observed by the Mössbauer effect because there is a strong internal magnetic field, information about the multipole character of γ_2 and γ_1 can be obtained by measuring the relative intensities of the magnetic components of γ_1 in coincidence with γ_2 for specified emission angles of γ_1 and γ_2 with respect to an external polarizing field. In particular, the mixing ratio of γ_2 can be determined if the spins of the levels and the multiple character of γ_1 are known. One advantage of this method over normal γ-γ directional correlation experiments is that the populations of the sublevels of state (1) are independent of the precession of the nuclear spin axis around the field direction. This makes the method especially suitable for cases where the lifetime of state (1) is so long that the directional correlation is appreciably affected by the precession. Another advantage of the method is that there is no averaging effect of the summation over the sublevels. The method has been used to determine the $E2/M1$ mixing ratio of the 123-keV transition in the $(\frac{5}{2}-) \xrightarrow{123\text{ keV}} (\frac{3}{2}-) \xrightarrow{14.4\text{ keV}} (\frac{1}{2}-)$ cascade in Fe^{57}. Both gamma rays were observed under angles of 90° with respect to the field magnetizing the Armco foil in which the Co^{57} activity was diffused. Designating the intensities of the components of γ_1 observed in coincidence with γ_2 by $I_1^c \cdots I_6^c$ (from low to high energy) the following ratios were measured: $I_1^c/I_2^c = 0.91 \pm 0.035$ and $I_6^c/I_5^c = 0.89 \pm 0.035$. Taking into account incomplete magnetization of the source and finite solid angles of the counters, an $E2/M1$ mixing ratio $\delta = -0.15 \pm 0.035$ is derived from these ratios.

RECOILLESS resonance absorption permits the measurement of the relative intensities of the separate magnetic components of gamma transitions (γ_1) from excited states $J_1\pi_1$ to ground states $J_0\pi_0$ (J = nuclear spin, π = parity) in cases where the magnetic field H at the gamma emitting nuclei is so strong that the Zeeman splitting of the levels exceeds the natural linewidth. These intensities are determined by (1) the population of the magnetic sublevels of state $J_1\pi_1$, (2) the multipole character of the transition, and (3) the angle θ_1 between the direction of the magnetic field and that of the gamma rays.

In normal Mössbauer experiments the populations of the sublevels of state $J_1\pi_1$ are equal. Let us suppose now that there is a second excited state $J_2\pi_2$ which decays by γ emission (γ_2) to state $J_1\pi_1$. If we observe this state only if formed after γ_2 emission in a certain direction (say, under an angle θ_2 with the direction of H), the apparent populations of the magnetic sublevels of state $J_1\pi_1$ are, in general, no longer equal. These populations depend on J_2, J_1, the multipole character of γ_2, and on θ_2. Therefore, the relative intensities of the components of γ_1 are changed if they are observed in coincidence with γ_2 and from this change the multipole character (in particular, the mixing ratio) of γ_2 can be derived. The calculations involved here are, of course, identical to those pertaining to γ-γ angular correlations, with the difference that there is no summing over the sublevels of the intermediate state. Since the populations of the sublevels are independent of the precession of the nuclear spin axis around the field direction, the intensities of the components of γ_1 are not affected by this precession, which in normal γ-γ angular correlation measurements may complicate the interpretation of the measurement. Another advantage of the method is that there is no averaging effect of the summation over the sublevels, which may make the angular correlation less sensitive to the mixing ratio of γ_2.

The method has been used to determine the $E2/M1$ mixing ratio of the 123-keV transition in Fe^{57}.

Most of the electron capture decay of Co^{57} goes through the

$$\frac{5}{2}- \xrightarrow{123\text{ keV}(\gamma_2)} \frac{3}{2}- \xrightarrow{14.4\text{ keV}(\gamma_1)} \frac{1}{2}-$$

cascade to the ground state of Fe^{57} [see Fig. 2(a)]. Assuming the 14.4-keV transition to be pure $M1$ (in accordance with the $L_{\text{I}}/(L_{\text{II}}+L_{\text{III}})$ conversion ratio measured by Ewan, Graham, and Geiger[1] the relative intensities $I_1 \cdots I_6$ of the six components in a single count spectrum should be 3:4:1:1:4:3 if the source is completely magnetized perpendicular to the direction of observation of $\gamma_1(\theta_1 = 90°)$. The populations of the sublevels of the 3/2− state in the coincidence experiment were calculated as a function of the angle θ_2 between γ_2 and the field direction from the formula given by Ling and

* Present address: Department of Physics, University of Illinois, Urbana, Illinois.

[1] G. T. Ewan, R. L. Graham, and J. S. Geiger, Nucl. Phys. 19, 221 (1960).

Falkoff[2] for the intensities of mixed $(L, L-1)$ transitions between magnetic substates. Equal populations for the sublevels of the $5/2-$ state were assumed. From this calculation the intensity ratios I_1^c/I_2^c and I_6^c/I_5^c in the coincidence experiment were obtained as a function of the ratio δ of the $E2$ and $M1$ matrix elements of the $5/2- \rightarrow 3/2-$ transition. In Fig. 1 the intensity ratio of the coincident lines divided by that of the single count lines is plotted as a function of δ for angles $\theta_2=\theta_1=90°$ (broken line).

In the experiment [see Fig. 2(b)] a magnetized Co^{57} source of about 20 μC was used, obtained by electroplating Co^{57} on an Armco foil and diffusing it into the foil at 900°C. In order to obtain a good coincidence counting efficiency, an absorber with a high ratio of resonant to nonresonant absorption was needed. Also, for ease of interpretation of the results the absorber had to be "unsplit." For these reasons the absorber was made of $Na_4Fe(CN)_6$ enriched to 67% in Fe^{57}.

The absorber was given a sinusoidal motion by coupling it through an aluminum driving rod to the moving coil of a loudspeaker. A second moving coil, also coupled to this rod, served as a velocity pickup.

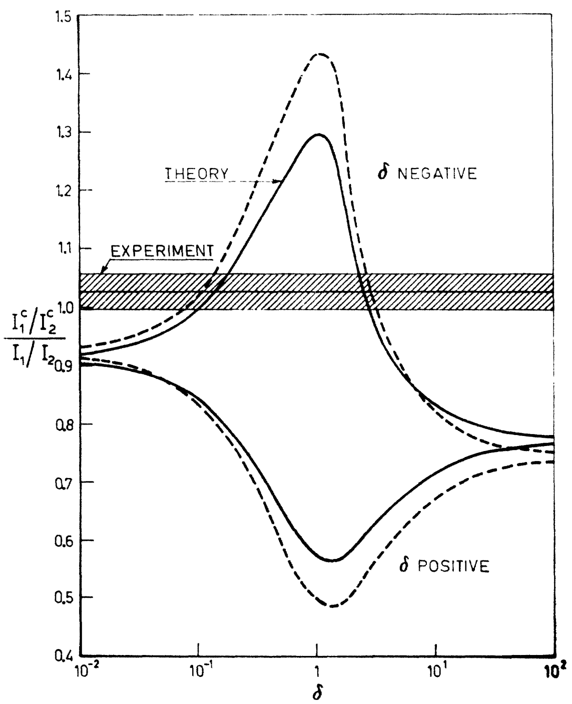

FIG. 1. Intensity ratio of components 1 and 2 of 14.4-keV line of Fe^{57} observed in coincidence with 123-keV line divided by intensity ratio of single count lines as a function of the $E2/M1$ mixing ratio δ. Dashed line: theoretical curve for complete magnetization of source perpendicular to direction of emission of both γ rays and for infinitely small solid angles. Solid line: theoretical curve for incomplete magnetization and finite solid angles encountered in the experiment. Shaded area: experimental value of intensity ratio with statistical limits of error.

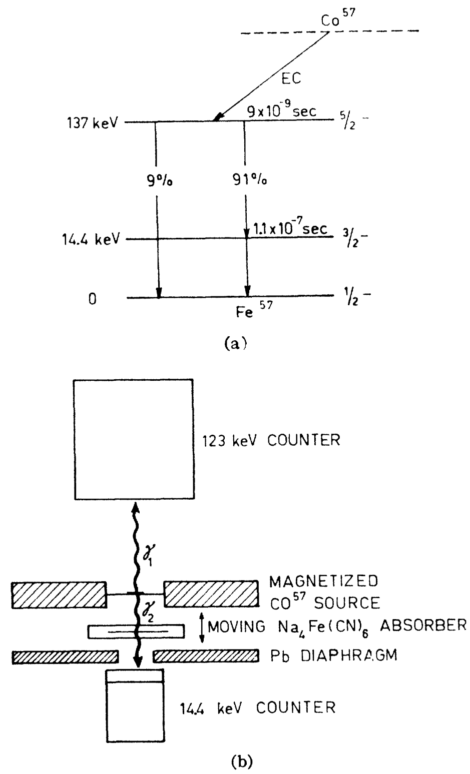

FIG. 2. (a) Decay of Co^{57}; (b) experimental arrangement.

The amplified signal from the latter coil was used to modulate the amplitude of the pulses from the 14.4-keV channel. The amplitude-modulated pulses were applied to a 256-channel analyzer, gated by coincidences between the 123- and 14.4-keV channels.[3] The coincidence resolving time of about 0.2 μsec yielded a real to chance coincidence ratio of about 8. Coincidence runs were alternated with runs in which the single-count hyperfine spectrum of the 14.4-keV line was recorded.

The shape of the background spectrum was also repeatedly recorded by replacing the Co^{57} source by a Co^{60} source.

From the single-count spectrum, intensity ratios $I_1/I_2=0.89\pm0.02$ and $I_6/I_5=0.87\pm0.02$ were determined by least-squares fits of the measured points (all runs added together) to Lorentzian distributions. Taking into account the finite solid angle of the 14.4-keV detector but not the finite size of the source, a ratio of 0.79 is expected for the intensities of these lines if the source is completely magnetized. Due to the somewhat irregular distribution of the Co^{57} activity, the effect of the finite size of the source could not be calculated very accurately. This effect might increase the ratio just quoted by at most 2%. Comparing this ratio

[2] D. S. Ling and D. L. Falkoff, Phys. Rev. **76**, 1639 (1949).

[3] Actually, for this type of experiment, a constant velocity arrangement would have been preferable, since much time was wasted in counting events that were of no interest. However, such an arrangement was not available at the time the experiment was carried out.

with the experimental value, it is found that $(6.3\pm1.4)\%$ of the magnetization is effectively perpendicular to the direction of the external field. Including this effect as well as the finite solid angles of both counters in the calculation of the intensity ratio of the coincidence lines as a function of δ, the solid line in Fig. 1 is obtained. The experimental value of this ratio was determined from a least-squares fit of the sum of all coincidence runs to Lorentzian distributions. The results are $I_1^c/I_2^c=0.91\pm0.035$ and $I_6^c/I_5^c=0.89\pm0.035$. The average of these values, divided by the average intensity ratio of the single count lines and corrected for chance coincidences, is also indicated in Fig. 1, together with its statistical limits of error. From the intersections with the theoretical curve (solid line), values of $\delta=-0.15\pm0.035$ or $\delta=-2.58\pm0.024$ are found. The first value yields an enhancement factor of about 4 for the $E2$ part of the 123-keV transition; the second value may be discarded because it would give a much too large $E2$ transition probability.

The value of δ found from the present experiment is in good accordance with a value previously determined by Bishop *et al.*[4] from a study of the directional distribution and the polarization of 123-keV γ rays emitted by oriented Co^{57} nuclei.

[4] G. R. Bishop, M. A. Grace, C. E. Johnson, A. C. Knipper, H. R. Lemmer, J. Perez y Jorba, and R. G. Surlock, Phil. Mag. **46**, 951 (1955).

PHYSICAL REVIEW VOLUME 129, NUMBER 3 1 FEBRUARY 1963

Nuclear Moments and Hyperfine Structure of 13-Year Eu^{152}*

SEYMOUR S. ALPERT†
Lawrence Radiation Laboratory, University of California, Berkeley, California
(Received 30 August 1962)

The magnetic dipole interaction constant a and the electric quadrupole interaction constant b for Eu^{152} (13 yr) were measured by the method of atomic beams. These values are $a=\pm9.345\pm0.006$ Mc/sec and $b=\mp1.930\pm0.165$ Mc/sec. By comparison with the known moment of Eu^{151}, the nuclear dipole moment of Eu^{152} was found to be $\mu=\pm1.912\pm0.004$ nm. The sign of this moment cannot be inferred from the experimental results. The zero-field hyperfine separations between levels of different total angular momentum were directly measured.

INTRODUCTION

In recent years much work has been done on the isotopes of europium $(4f^75s^25p^66s^2,{}^8S_{7/2})$. Pichanick *et al.* directly determined the magnetic dipole moment of stable Eu^{151} in an atomic beam experiment using three rf loops.[1] Sandars and Woodgate, also using the atomic beam method and mass-spectrographic detection, determined the interaction constants for the stable europium isotopes.[2] By use of the results of these experiments, it is possible by means of comparison to determine the nuclear magnetic dipole moment for all the other europium isotopes for which the interaction constants can be measured in the free atom.

Since there are seventeen isotopes of europium with atomic weights in the range 144 to 159, it would seem that the validity of the collective model which is generally taken to hold in the region $150<A<190$ could be checked or modified with knowledge of the nuclear moments of many of the isotopes of europium.

Abraham *et al.*, working with divalent europium ions bound in crystalline KCl, have performed electron paramagnetic resonance experiments on Eu^{151}, Eu^{152}, Eu^{153}, and Eu^{154} and measured the hyperfine interaction constants of these species in ionic form.[3] The spin of Eu^{152} was found to be $3\hbar$. Similarly, Baker and Williams measured the hyperfine interaction in ionic Eu^{151} and Eu^{153} bound in crystalline CaF_2 by means of the electron nuclear double resonance (ENDOR) technique.[4] When the results relating to the crystalline ionized Eu isotopes are compared with similar results derived for the atomic state by means of the atomic beam method, significant differences are seen in the magnetic dipole interaction constants. This, when subjected to the theoretical analysis, may furnish useful information about the electronic wave function of atomic and doubly ionized europium.

THEORY

In the free atom there generally exists an angle-dependent interaction between the nucleus and the surrounding electrons. This interaction can be represented in the nuclear Hamiltonian by a series of terms of which only the first two are ordinarily significant.

* Work supported by the U. S. Atomic Energy Commission.
† Present address: Bell Telephone Laboratories, Murray Hill, New Jersey.

[1] F. M. Pichanick, P. G. H. Sandars, and G. K. Woodgate, Proc. Roy. Soc. (London) **A257**, 277 (1960).

[2] P. G. H. Sandars and G. K. Woodgate, Proc. Roy. Soc. (London) **A257**, 269 (1960).

[3] M. Abraham, R. Kedzie, and C. D. Jeffries, Phys. Rev. **108**, 58, (1957).

[4] J. M. Baker and F. I. B. Williams, Proc. Roy. Soc. (London) **A267**, 283 (1962).