Nuclear-Orientation and Angular-Correlation Studies of ^{57}Fe and ^{58}Fe

R. A. Fox, W. D. Hamilton, and M. J. Holmes
School of Mathematical and Physical Sciences, University of Sussex, Brighton, England
(Received 7 October 1971)

The $E2:M1$ multipole mixing ratio of the $\frac{5}{2}^- \rightarrow \frac{3}{2}^-$ and $2^+ \rightarrow 2^+$ transitions between the second and first excited states in ^{57}Fe and ^{58}Fe, respectively, have been measured. The nuclear-orientation sources were prepared by diffusing cobalt activity into iron. Nuclear polarization was obtained by cooling the sample to 25 mK using a ^{3}He-^{4}He dilution refrigerator and applying sufficient external magnetic field to saturate the iron. The value $\delta = 0.120 \pm 0.002$ was obtained for the 122-keV transition in ^{57}Fe by a nuclear-orientation measurement. The ^{58}Fe $\gamma\gamma$ angular-correlation results used (a) NaI(Tl)-NaI(Tl) and (b) Ge(Li)-NaI(Tl) detector combinations and gave consistent results: (a) $A_{22} = 0.460 \pm 0.013$, $A_{44} = 0.150 \pm 0.021$; (b) $A_{22} = 0.45 \pm 0.04$, $A_{44} = 0.08 \pm 0.09$. The orientation experiment yielded $A_2 = 0.33 \pm 0.03$. These values give $\delta = 0.69 \pm 0.05$ for the 865-keV transition. From a knowledge of the temperature of the orientated nuclei the Fermi to Gamow-Teller matrix element ratio of the two 2^+-2^+ electron-capture and β^+ transition from ^{58}Co were obtained. For the transition to the 1675-keV level $\lambda = 0.07 \pm 0.06$ and to the 810-keV level $\lambda = 0.04 \pm 0.06$. The mixing ratio measurements remove existing possible ambiguities and illustrate the very useful advantages of combining nuclear-orientation and angular-correlation methods.

INTRODUCTION

There have been many investigations of the electron capture and positron decay of the two isotopes ^{57}Co and ^{58}Co so that the decay scheme and nuclear parameters are known.[1] However, in both cases the decay is predominantly to the first excited states in iron nuclei, which makes measurement of the mixing ratio of the γ rays between the second and first excited states difficult. In view of the considerable disagreement between the published results for the mixing ratio of the $2^+ \rightarrow 2^+$ transition in ^{58}Fe,[1] the experiment was repeated making use of both the methods of $\gamma\gamma$ angular correlation and γ-ray emission following equilibrium nuclear orientation. Likewise, the precise angular-correlation results on ^{57}Fe have an uncertainty due to the long half-life ($t_{1/2} = 9.8 \times 10^{-8}$ sec) of the 14.4-keV intermediate level, and therefore it was felt necessary to perform a nuclear-orientation experiment using a high-resolution germanium detector.

The angular distribution of radiation from a polarized sample is given by[2]

$$W(\theta) = \sum_{k \text{ even}} B_k U_k A_k Q_k P_k(\cos\theta). \tag{1}$$

The quantities B_k contain the dependence on the equilibrium nuclear-level populations and are therefore a function of temperature. For example,

$$B_2 = 3\left\{\sum_m [m^2 - \tfrac{1}{3}I(I+1)]p_m\right\} \times [\tfrac{1}{5}I(I+1)(2I-1)(2I+3)]^{-1/2}, \tag{2}$$

where p_m is the population of the state m. In the case of a nucleus of spin I and magnetic moment $\vec{\mu}$ experiencing a pure magnetic interaction in a magnetic field $\vec{H}$, then

$$p_m = e^{-m\vec{\mu}\cdot\vec{H}/IkT}.$$

The coefficients U_k give the degree of depolarization caused by an unobserved intermediate transition, and A_k are the angular-distribution coefficients. The Q_k factors correct for the finite solid angle of the detector and in many circumstances these are close to unity. From these definitions it can be seen that for two γ rays from the same nuclear level the values of $B_k U_k$ will be the same. This will be true even if some depolarization occurs in the intermediate state before emission of the γ rays provided that cylindrical symmetry is maintained. It is also true if not all nuclei experience the same hyperfine field because of poor solubility in the host. It is simple to show that for two γ rays, represented by primed and unprimed coefficients, emitted from the same state,

$$\frac{A_2}{A_2'} = \frac{\frac{3}{8}[1 - W(0)] + [W(\pi/2) - 1]}{\frac{3}{8}[1 - W'(0)] + [W'(\pi/2) - 1]} \tag{3}$$

and

$$\frac{A_4}{A_4'} = \frac{\frac{1}{2}[1 - W(0)] - [W(\pi/2) - 1]}{\frac{1}{2}[1 - W'(0)] - [W'(\pi/2) - 1]}, \tag{4}$$

provided $W(0)$ and $W(\pi/2)$ are measured at the same temperature. It has been assumed that the Q_k factors are identical for the two transitions.

The unperturbed angular-correlation function

for two successive γ rays, 1 and 2, is given by[3]

$$W(\theta)=\sum_{k\ \text{even}} A_k(1)Q_k(1)A_k(2)Q_k(2)P_k(\cos\theta). \tag{5}$$

For the case in which radiation 1 is a mixed transition containing multipoles L and L' and with a mixing ratio δ, defined by

$$\delta=\frac{\langle I_i \| L' \| I\rangle}{\langle I_i \| L \| I\rangle},$$

then the $A_k(1)$ coefficients have the form

$$A_2=\frac{1}{1+\delta^2}[F_2(LLI_iI)-2\delta F_2(LL'I_iI)+\delta^2 F_2(L'L'I_iI)] \tag{6}$$

and

$$A_4=\frac{1}{1+\delta^2}[F_4(LLI_iI)+\delta^2 F_4(L'L'I_iI)], \tag{7}$$

where I_i is the spin of the intermediate level and $L'=L+1$. However, for the nuclear-orientation experiment, $\gamma(1)$ is in fact the second transition following the decay of the nucleus, so that now

$$A_2=\frac{1}{1+\delta^2}[F_2(LLI_iI)+2\delta F_2(LL'I_iI)+\delta^2 F_2(L'L'I_iI)]. \tag{8}$$

The A_4 coefficient remains unchanged. (Note that we are using the notation of Krane and Steffen,[4] so that the sign of δ which indicates the relative phase of the multipoles in the mixed transition remains unchanged.) These two possibilities for A_2 are well illustrated in a subsequent figure (Fig. 5) which is a reproduction of the Arns-Wiedenbeck[5]

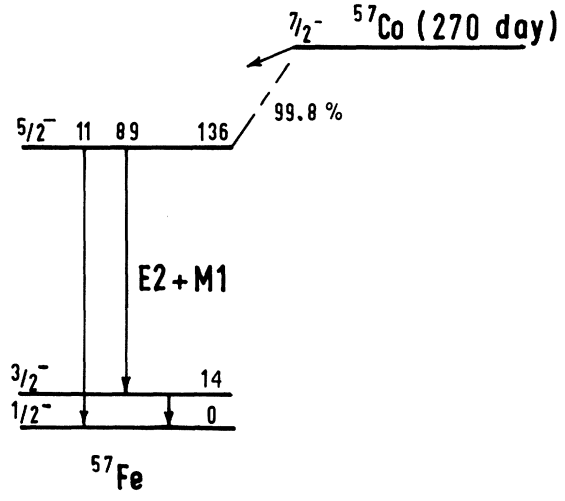

FIG. 1. The decay scheme of ^{57}Co taken from Ref. 1. Level energies are given in keV and the percentage intensity of the branching transitions is indicated.

plot for the transition $2(M1,E2)2$. It can be seen from this diagram that the angular-correlation and nuclear-orientation measurements complement one another so that a measurement of A_4 is not necessary. Furthermore, should one of these measurements of A_2 not be a sensitive function of δ, the other measurement will be sensitive. This situation is a general feature of all spin sequences, and the nuclear-orientation method obtains particular importance in mixing-ratio measurements for those cases in which the angular-correlation A_2 coefficient is insensitive and the corresponding A_4 coefficient does not have sufficient accuracy or precision.

EXPERIMENTAL PROCEDURE

A. Nuclear-Orientation Measurements

Samples were prepared by diffusion of the ^{57}Co and ^{58}Co activity, in the form of carrier-free cobaltous chloride obtained from the Radiochemical Center, Amersham, into small pieces of 99.999% pure iron at 850°C for 24 h in an atmosphere of hydrogen. Since both iron isotopes decay predominantly by electron capture, radioactive heating is not a problem, so that source strengths of about 10 μCi were used. The cobalt of the source constituted an impurity of about 10^{-6} in the iron.

The samples were cooled to approximately 30 mK by attaching them to the mixing chamber of an Oxford Instruments Mark IV ^{3}He-^{4}He dilution refrigerator. Thermal contact was obtained by soldering the samples with Cd-Bi eutectic solder to a copper holder. This was screwed, with a coating of Apiezon N grease, to the base of a sintered copper plug which formed part of the mixing chamber.

The iron samples were polarized with a small superconducting magnet. A field of 1.5 kOe was found to be sufficient to saturate the iron; however, 2 kOe was used in all experiments. The field at the cobalt nuclei was then[6] 288 kOe aligned in the direction of the external field so that $W(0)$ and $W(\pi/2)$ counts were taken relative to this direction.

The detector used was a 30-cm^3 Ge(Li) detector with a resolution of 2.1 keV at 1.33 MeV. Since this was placed at 14 cm from the sample, the values of Q_k were extremely close to unity and effectively independent of γ-ray energy.

The ^{57}Co measurements were performed to investigate the $E2:M1$ mixing ratio of the 122-keV $\frac{5}{2}^- \rightarrow \frac{3}{2}^-$ transition in ^{57}Fe (see Fig. 1.) Since the 136-keV $\frac{5}{2}^- \rightarrow \frac{1}{2}^-$ transition is pure $E2$ and arises from the same level in iron, Eqs. (3) and (4) may be applied so that a knowledge of the sample temperature is not required. Counts were taken when the

source was cold, with the Ge(Li) detector at successive $W(0)$ and $W(\pi/2)$ positions by rotating an arm on which the detector was mounted. These were then normalized with an equivalent set of warm (1-K) counts. The γ-ray spectra were fed via an OTREC 716A amplifier into 200 channels of an Intertechnique multichannel analyzer which was backbiased to increase the separation between 122- and 136-keV peaks. Figure 2 shows a typical warm spectrum with such an arrangement. Shifts of the spectra were never more than 5% of the peak width (full width at half maximum) and did not introduce significant errors. The background beneath the peaks was extremely small and was obtained by observing the counts a few keV on the high-energy side of the peak, and subtracting these from the counts under the peak.

The nuclear-orientation experiment using ^{58}Co, in addition to yielding the $E2:M1$ mixing ratio of the 865-keV $2^+ \rightarrow 2^+$ transition, also provides a value for the Fermi to Gamow-Teller matrix element ratio of the $2^+ \rightarrow 2^+$ electron-capture transition to the 1.675-MeV level in ^{58}Fe (see Fig. 3). To measure the latter it is necessary to know the sample temperature and, since a small quantity of ^{60}Co was present as an impurity in the source, observation of the 1.117- and 1.332-MeV transitions in ^{60}Ni provided a convenient method.

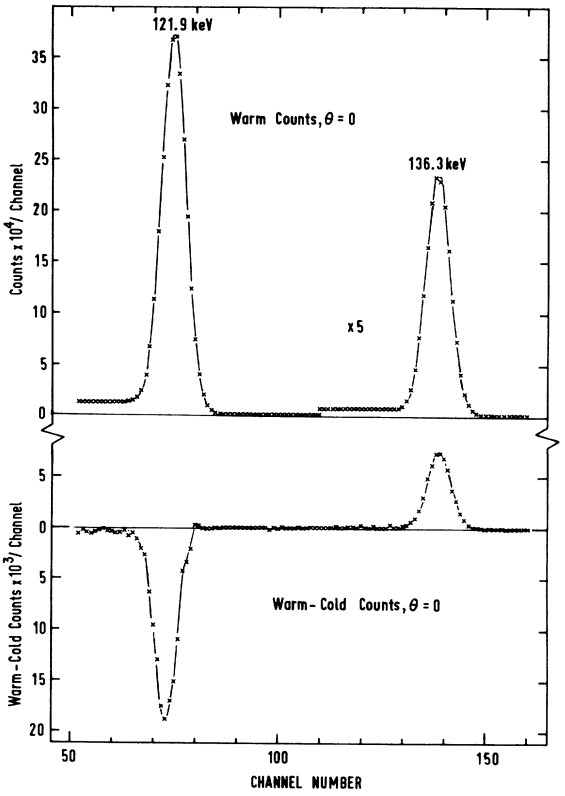

FIG. 2. The upper part of the figure represents the spectrum recorded at 0° with respect to the axis of polarization when the source was warm (1 K). The lower part is the difference between the warm and cold (~30-mK) spectra, and indicates clearly the difference in the behavior of the two transitions.

At the temperature of the experiment (23 mK) the A_4 coefficient is small, and therefore difficult to measure sufficiently accurately to distinguish between the two possible values of δ allowed by the measured A_2 value. However, it is possible to combine the results of an angular-correlation measurement with an A_2 coefficient obtained from an orientation experiment to give a unique solution for δ. In these circumstances it is necessary to record counts only along the field axis. An estimate of the magnitude of the fourth-rank statistical tensor $B_4 U_4 A_4 Q_4$ may be obtained from a knowledge of the sample temperature, the tabulated U_4 and Q_4 coefficients, and the value of A_4 derived from the measured A_2 coefficient.

The output from the germanium detector was fed into two multichannel analyzers to observe the 800- to 900-keV and 1100- to 1700-keV regions of the spectrum, respectively. Sets of cold (23-mK) and warm (1-K) counts were taken for 60-min periods to obtain the anisotropies of each peak; the background was obtained in the same way as for the ^{57}Co measurements.

B. Angular-Correlation Experiments

Liquid sources were used consisting of cobaltous chloride in 0.1N HCl contained in a nylon cylinder

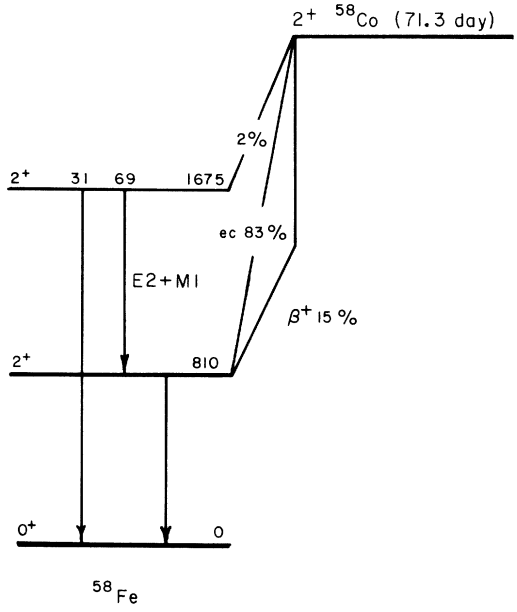

FIG. 3. The decay scheme of ^{58}Co taken from Ref. 1.

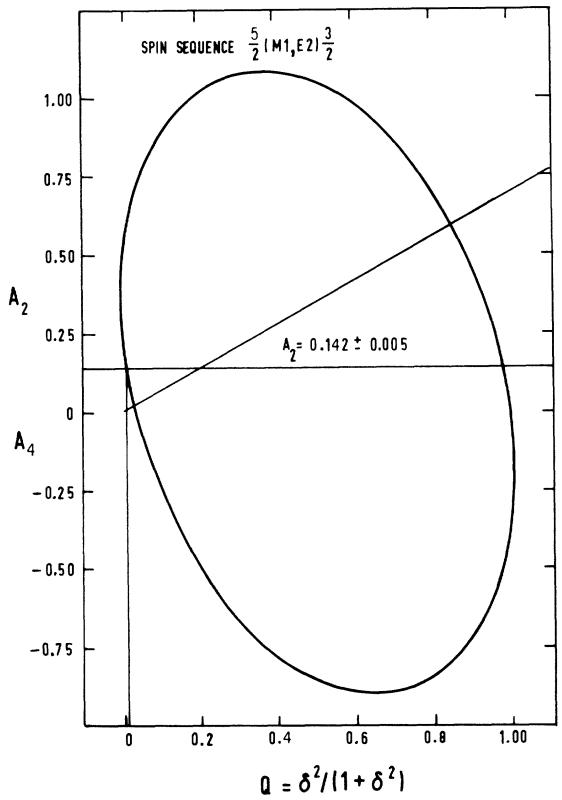

FIG. 4. The Arns-Wiedenbeck plot for a $\frac{5}{2}(E2, M1)\frac{3}{2}$ transition and the present result.

with internal dimensions 0.15 cm diameter by 0.5 cm long. The axis of the source was perpendicular to the plane of the detectors.

Two separate $\gamma\gamma$ angular-correlation measurements were performed. The first used two NaI(Tl) detectors, 7.6 cm by 7.6 cm and 3.8 cm by 5.1 cm, with a conventional fast-slow coincidence system and leading-edge timing. The energy resolution of NaI(Tl) does not allow the 810- and 865-keV photopeaks to be resolved and the composite correlation was measured. Because of the intense feeding to the 810-keV level of ^{58}Fe the majority of 810-keV radiations have no associated 865-keV event, so that the accidental contribution to the coincidence rate may be high. In the experiment, in which the resolving time was $2\tau = 30$ nsec, it was of the order of 40% of the true rate, since a source strength of ~50 μCi was used. Under these circumstances the correlation coefficients are very sensitive to the value of the accidental-to-true ratio, and while this may be accurately determined, the application of large correction factors in the evaluation of the A_{kk} angular-correlation coefficients is not very satisfactory.

Therefore, it was decided to use a high-resolution detector to separate the two lines and consequently very significantly reduce the accidental-coincidence contribution associated with the 865-keV line. A 30-cm^3 coaxial Ge(Li) detector was used together with a 7.6-cm by 7.6-cm NaI(Tl) crystal. The coincidence condition gated a multichannel analyzer in which the 810- and 865-keV full-energy lines recorded by the Ge(Li) were stored. This coincidence spectrum on the analyzer showed that the 810-keV line contained about one hundred times as many accidental coincidences as the 865-keV line. Upon subtraction of the accidental spectrum, obtained by inserting a time delay, the two peaks were reduced to equal size. The 865-keV peak had less than a 1% contribution from accidentals. The coincidence spectra were taken at four angles; angles in the vicinity of 180° being omitted to remove the possibility of 511-keV coincidence-summing effects. The data were accumulated in a group of independent measurements which were analyzed separately using a standard fitting procedure to extract the normalized coefficients of the second- and fourth-order Legendre polynomials. The program corrected for the small source miscentering and for the extension of the source. Weighted averages of the coefficients were obtained from the group of data.

EXPERIMENTAL RESULTS

A. 122-keV Transition in ^{57}Co

Several experiments were performed at temperatures of approximately 30 mK. Use of Eqs. (3) and (4) produced a consistent set of results, the weighted mean of which was

$$\frac{A_2}{A_2'} = -0.265 \pm 0.008\,, \quad \frac{A_4}{A_4'} = -0.2 \pm 0.2\,.$$

Since the 136-keV transition is pure $E2$, then $A_2' = -0.5345$ and $A_4' = -0.6172$, so that for the 122-keV transition

$$A_2 = 0.142 \pm 0.005\,,$$

$$A_4 = 0.12 \pm 0.12\,.$$

TABLE I. Measured anisotropies for the γ rays observed with the ^{58}Co source which contained a small ^{60}Co admixture.

Transition energy (keV)	810	865	1173	1332	1675
$[1 - W(0)]$ (%)	12.5 ± 0.1	−8.04 ± 0.06	17.1 ± 0.6	16.3 ± 0.6	13.15 ± 0.1

TABLE II. The angular-correlation and nuclear-orientation results. Also included in the table are the A_{kk} coefficients derived using the nuclear-orientation result for δ.

Measuring method	A_{22}	A_{44}	A_2	δ
NaI(Tl)-NaI(Tl)	0.460 ± 0.013	0.150 ± 0.021		$-0.94^{+0.09}_{-0.08}$
Ge(Li)-NaI(Tl)	0.45 ± 0.04	0.08 ± 0.09		$-1.00^{+0.60}_{-0.20}$
Nuclear orientation	0.487 ± 0.001	0.105 ± 0.010	0.33 ± 0.03	−0.69 ± 0.05

The value of A_4 suggests strongly that the 122-keV transition is mainly $M1$; the other solution would require an A_4 of approximately 0.7 (cf. Fig. 4). Assuming this, the value of A_2 gives $\delta = 0.120 \pm 0.002$. This result is in good agreement with the angular-correlation results of $\delta = 0.121 \pm 0.003$[7] and $\delta = 0.131 \pm 0.006$.[8] Previous nuclear-orientation results were considerably less accurate because of the poor resolution of the detectors which were used. However, our attention has recently been drawn to a similar nuclear-orientation result[9] on ^{57}Co of $\delta = 0.123 \pm 0.001$.

B. 865-keV Transition in ^{58}Co

The average of several experiments at approximately the same temperature produced anisotropies, for each of the γ-ray energies as shown in Table I. The anisotropy obtained for ^{60}Co indicates that the sample temperature was 23.0 ± 1.0 mK. Therefore, using the known values of the magnetic moment μ and the magnetic field H for cobalt in iron,[6] the values of B_2 and B_4 are given by

$$B_2 = 0.456 \pm 0.025,$$

$$B_4 = 0.021 \pm 0.0025.$$

Equation (3) can now be applied to the 865- and 1675-keV γ rays, using values of $W(\pi/2)$ estimated from this value of B_4. The error introduced in this way is less than the statistical error, and one obtains for the 865-keV transition $A_2 = 0.33 \pm 0.03$.

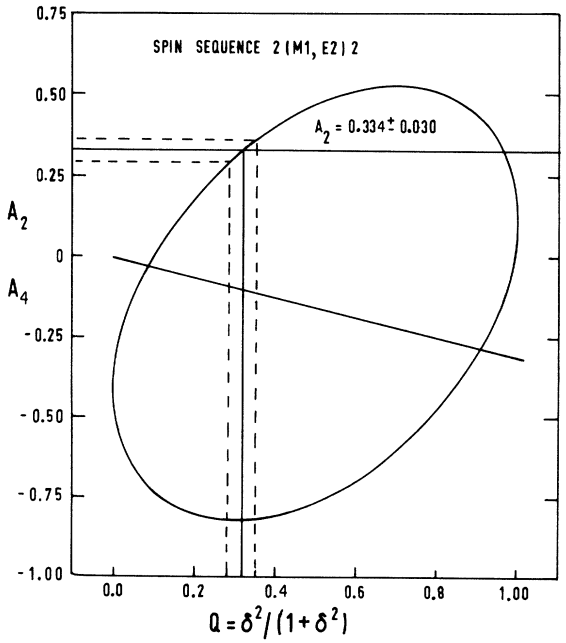

FIG. 5. The Arns-Wiedenbeck plot for a $2(E2, M1)2$ transition on which the A_2 coefficient obtained from the nuclear-orientation measurement of the 865-keV transition in ^{58}Fe is shown.

The angular-correlation results are presented in Table II. The result obtained using the pair of NaI(Tl) detectors is particularly sensitive to the accidental contribution and great care was taken to assess it correctly.[10] The present values for the A_{kk} coefficients are consistent for both detector combinations, but they are significantly different from previous angular-correlation results obtained using NaI(Tl) detectors.[1] The most recent measurements[11,12] also use a Ge(Li)-NaI(Tl) detector combination and obtain results in agreement with the present work when data analysis was confined to events in coincidence with the 865-keV transition. However, if the angle-dependent coincident intensities of the 810-keV line were used, significant discrepancies are reported.[12] We chose to analyze only the 865-keV peak in the coincidence spectrum, since the very large accidental contribution in the 810-keV peak reduced the statistical significance of this data to a useless level.

The A_2 coefficient from the nuclear-orientation measurement yields for the $E2:M1$ mixing ratio of the 865-keV transition the value

$$\delta = -0.69 \pm 0.05.$$

The other possible solution is completely inconsistent with the A_{22} angular-correlation coefficients as may be seen in Fig. 5. It is now possible, using Eqs. (6) and (7) and the above value of δ, to calculate the corresponding A_{kk} coefficients appropriate to the angular-correlation measurements. These values are included in Table II and they are consistent with the angular-correlation results; however, because the A_{22} coefficient lies on a turn-

ing point of Eq. (6), i.e., an insensitive region of the Arns-Wiedenbeck plot, the δ values derived from the angular-correlation measurements have poor precision.

The value of δ is in good agreement with the result $\delta = -0.57 \pm 0.06$ obtained by measuring γ-ray correlations following n capture by Fe.[13] This experiment was free from large accidental contributions to the measured cascades, since the n-capture reaction strongly fed the relevant excited states.

C. $2^+ \rightarrow 2^+$ Electron-Capture Transition from ^{58}Co

The U_k coefficients in Eq. (1) depend on the nature of the electron-capture and β^+ transitions which feed the two 2^+ states whose γ decay is observed. These transitions in each case are $2^+ \rightarrow 2^+$, and thus may be a mixture of Fermi and Gamow-Teller components whose matrix element ratio is denoted by λ.

For the 1675-keV transition we measured the value $U_2 = 0.53 \pm 0.03$, and by an iterative process obtain $U_4 = -0.56 \pm 0.09$. These values are consistent with the result

$$\lambda = 0.07 \pm 0.06$$

for the transition to the upper 2^+ level in ^{58}Fe. Similarly, the analysis of the 810-keV transition, after allowing for the 1.2% feeding via the 865-keV γ ray, gives the values $U_2 = 0.52 \pm 0.03$ and $U_4 = -0.60 \pm 0.9$; and thus for the decay to the 810-keV level we have

$$\lambda = 0.04 \pm 0.06 .$$

This result is in agreement with that of Chapman *et al.*[14]:

$$\lambda = 0.01 \pm 0.01 .$$

CONCLUSIONS

The combination of angular-correlation and nuclear-orientation techniques is a very sensitive way to determine a multipole mixing ratio. The techniques complement each other, and this is well exemplified by the 865-keV transition in ^{58}Fe for which the angular-correlation measurements do not provide results that are very sensitive to δ.

The nuclear-orientation measurements on transitions in both ^{57}Fe and ^{58}Fe also illustrate the strength of the technique of using a pure transition from the same level that feeds the transition of interest to monitor the degree of orientation. This effectively removes many problems associated with accurate hyperfine-field and temperature measurements and with knowledge of the nuclear environment and possible depolarization effects.

It is also interesting to note the comparatively large $M1$ admixture in $2^+ \rightarrow 2^+$ transition in ^{58}Fe, which is normally thought to be of a vibrational character and might thus be expected to be predominantly $E2$. A study of similar transitions in this mass region is being made.

This work was supported by the Science Research Council, and M. J. H. acknowledges receipt of a Science Research Council studentship.

[1] J. Rapaport, Nucl. Data B3 (Nos. 3,4), 103 (1970); S. Raman, *ibid.* B3 (Nos. 3,4), 145 (1970).
[2] M. A. Grace and R. J. Blin-Stoyle, *Handbook of Physics* (Academic, New York, 1957), Vol. 42, p. 555.
[3] H. Frauenfelder and R. M. Steffen, in *Alpha-, Beta-, and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North-Holland, Amsterdam, 1966).
[4] K. S. Krane and R. M. Steffen, Phys. Rev. C 3, 240 (1971).
[5] R. G. Arns and M. L. Wiedenbeck, Phys. Rev. 111, 1631 (1958).
[6] J. E. Templeton and D. A. Shirley, Phys. Rev. Letters 18, 240 (1967).
[7] C. Hohenemser, R. Reno, H. C. Benski, and J. Lehr, Phys. Rev. 184, 298 (1969).
[8] T. Lindquist and E. Heer, Nucl. Phys. 2, 680 (1956).
[9] R. D. Taylor, Los Alamos Laboratory Report No. LA-DC-12276, 1971 (unpublished).
[10] M. J. Holmes, M.S. dissertation, University of Sussex, 1969 (unpublished).
[11] U. Schotzig, H. Schrader, R. Stippler, and F. Munnich, Z. Physik 222, 479 (1969).
[12] N. C. Singhal, A. V. Ramayya, and J. H. Hamilton, Z. Physik 245, 50 (1969).
[13] U. Finger, M. Michaelis, H. Schmidt, and H. Ottmar, Nucl. Phys. A128, 641 (1969).
[14] C. J. S. Chapman, J. M. Gregory, R. W. Hill, and M. W. Johns, Proc. Roy. Soc. (London) A262, 541 (1961).