

## Beta radiations from $^{59}\text{Fe}$ and $^{60}\text{Co}$ nuclei polarized in iron foils cooled by dilution refrigeration\*

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(Received 2 August 1976)

Apparatus for observing the  $\beta$  emissions from radioactive nuclides diffused into iron foils cooled to 25 mK by a simple dilution refrigerator is briefly described. Tests using  $^{60}\text{Co}$  gave a  $\beta$  asymmetry coefficient  $A = -0.80 \pm 0.04$ , compared with the theoretical value  $-1$ , indicating the effect of scattering. For the  $^{59}\text{Fe}$  467-keV  $\beta$  transition the asymmetry was very small,  $(0.8 \pm 0.5)\%$  at 30 mK; no convincing asymmetry could be observed for the 273-keV  $\beta$  rays. This is ascribed to the recently determined small value of the magnetic moment of  $^{59}\text{Fe}$ ; the results are consistent with previous measurements of the  $\beta$ - $\gamma$  circular polarization correlations. An earlier experiment with  $^{59}\text{Fe}$  in rare earth double nitrate crystals is reevaluated. The magnitudes of the  $\beta$  matrix elements are briefly compared with single-particle shell model estimates including configuration mixing. Various model estimates of the magnetic moment are also presented.

[RADIOACTIVITY  $^{59}\text{Fe}$ ,  $^{60}\text{Co}$ ; measured  $I_\beta(\theta)$ , deduced  $\mu A(\gamma)$ ,  $\mu < 0$ . Calculated experimental and single particle  $|M_{GT}|^2$ , also model estimates of  $\mu$ .]

Earlier research<sup>1</sup> on the angular distributions of 1099- and 1292-keV  $\gamma$  radiations from  $^{59}\text{Fe}$  impurities oriented in rare earth double nitrate crystals seemed to suggest the possibility of anomalously large Fermi contributions to the 467- and 273-keV  $\beta$  transitions, but the interpretation was complicated by uncertainties concerning possible small  $M3$   $\gamma$ -ray admixtures and large intermediate state reorientation preceding the  $\gamma$  emissions, as well as by lack of understanding of the impurity sites. An apparently reliable, though somewhat imprecise, estimate of the magnetic dipole moment of  $^{59}\text{Fe}$  was extracted from the temperature dependence of the unexpectedly large  $\gamma$  anisotropies.

In order to disentangle the various factors involved, it was proposed<sup>1</sup> to polarize  $^{59}\text{Fe}$  in an iron foil cooled by  $^3\text{He}$ - $^4\text{He}$  dilution refrigeration to a relatively stable temperature where good statistics could be obtained on the asymmetry of the  $\beta$  spectra directly. This would in principle provide a more sensitive and direct measure of the Fermi and Gamow-Teller mixing as well as a more refined value of the nuclear moment, if sufficiently high degrees of orientation could be produced, without the complications of  $\gamma$  mixing and intermediate state reorientation. These latter factors can also distort  $\beta$ - $\gamma$  circular polarization correlations, and may possibly have contributed to the wide scatter of results by different investigators.<sup>2</sup> This is not to say that there cannot be difficulties with  $\beta$  angular distribution experiments, for example due to source metallurgy problems, to inaccurate estimate of the degree of nuclear polarization, and to

internal scattering in the necessarily rather "thick" sources.

A simple dilution refrigerator cryostat was designed and constructed, and the proposed experiment was carried out<sup>3</sup> after repeating the famous parity experiment<sup>4</sup> on  $^{60}\text{Co}$ , this time in an iron foil, to demonstrate that the apparatus was working properly. In addition, since the  $\beta$  and  $\gamma$  spectra of  $^{60}\text{Co}$  are very similar to  $^{59}\text{Fe}$ , the anticipated effect of source scattering on the  $\beta$ -radiation spectrum and angular distribution could be gauged rather accurately. The  $^3\text{He}$ - $^4\text{He}$  dilution refrigerator is a small-scale conventional<sup>5</sup> single-stage design employing a continuous heat exchanger of the concentric tube, coaxial counterflow type. Full details in Ref. 3 may be of interest to persons contemplating experiments involving oriented nuclei in which heat extraction rates and low temperature requirements are modest. This experiment remains the only observation reported to date of the  $\beta$  rays from a low temperature  $^{59}\text{Fe}$  source.

The several samples<sup>6</sup> consisted of a few tens of microcuries of  $^{59}\text{Fe}$  and/or  $^{60}\text{Co}$  electrodeposited on a 5-mm-diam spot in the center of a 1-cm  $\times$  1-cm  $\times$  0.0025-cm, 99.99% pure iron foil and then diffused at 900 or 1200°C for 2–3 h in a hydrogen atmosphere. These foils were indium soldered to a vertical face of a fin projecting down from the oxygen free high conductivity copper bottom of the mixing chamber. A small superconducting split solenoid, located in the 4.2-K liquid  $^4\text{He}$  bath, provided fields up to approximately 0.15 T for magnetizing the iron foils to saturation. The  $^{60}\text{Co}$  was used to provide both a well understood  $\beta$  spectrum for testing and

calibration purposes and well understood  $\gamma$  rays for thermometry<sup>7</sup>; when serving only as a thermometer the  $^{60}\text{Co}$  was in a separate iron foil attached to the back of the copper fin, so that its  $\beta$  rays were blocked from reaching the  $\beta$  detector directly.

The  $\gamma$  rays were counted with a 24-cm<sup>3</sup> Ge(Li) detector located outside and directly below the cryostat along the axis of orientation at a distance of 25 cm. The  $\beta$  rays, which must be detected inside the cryostat, were counted with a commercial<sup>8</sup> "partially depleted" silicon surface barrier detector of active area 25 mm<sup>2</sup> and 1-mm Si thickness. The detector was mounted below and to one side of the source on the 1-K heat shield at a distance of 3.4 cm from the center of the source; the angle between the line joining the centers of source and detector and the magnetic field direction in the plane of the iron foil was 34°. This arrangement has the advantage that very low energy electrons spiral along the magnetic field and do not enter the  $\beta$  detector, reducing the dead-time correction but on the other hand leading to distortion of the  $\beta$  spectrum above 200 keV if the applied field exceeds 1200 Oe. The Si detector was connected through a coaxial cable, consisting of a 0.97-cm-diam thin wall stainless steel tube with a manganin center wire, to conventional room temperature solid state electronics; bias supply, charge-sensitive preamp, linear amplifier with baseline restorer, and 400-channel analyzer.

Though recurrent problems were encountered<sup>9</sup> with cracking of the gold and aluminum film contacts in the region where the silicon was epoxied to its ceramic ring mount, a fairly simple spring-loaded mounting was finally devised to make firm electrical contact directly to the edges of the gold front window of the silicon wafer itself and to an indium pad pressed against its aluminum back. With one detector especially this arrangement proved to be sufficiently reliable and when tested with  $^{133}\text{Ba}$  and  $^{113}\text{Sn}$  conversion electron sources gave a resolution less than 10 keV in the 300–400-keV range. A reverse bias of only 40 V was needed to deplete this detector at 1 K. Although the resolution and linearity were quite satisfactory, small gain shifts complicated analysis of the  $\beta$  spectra and, if more severe than two channels, caused us to reject some of the noticeably distorted spectra.

The  $^{60}\text{Co}$  and  $^{59}\text{Fe}$   $\beta$  spectra emitted from these sources and measured by these detectors were least squares fitted quite accurately by the empirical form  $N^{1/2} = an + b$  in the upper part of the spectrum, where  $N$  is the number of true counts due to that  $\beta$  component accumulated in channel  $n$  during 10 min of live time and  $a, b$  are fitted numerical constants. This form also accurately described

the upper portion of a penetrating, field independent background extending up to 1.3 MeV which was attributed primarily to Compton scattering of the 1.1–1.3-MeV  $\gamma$  rays in the 1-mm-thick Si wafer. The  $\beta$ -spectrum endpoints provided an energy calibration of each spectrum, from which total numbers of counts  $\sum N$  in a specified slice of a spectrum component could be calculated and correlated with an average  $\beta$ -particle speed  $v$  and with the temperature  $T$  at which the spectrum was accumulated.

These quantities could be used to determine experimentally the various factors in the expression<sup>10</sup> for the directional distribution of  $\beta$  particles in an allowed transition  $J(0, 1)J'$ :

$$W(\theta) = \frac{\sum N_{\text{cold}}}{\sum N_{\text{warm}}} = 1 + A_1 P_1(\theta) \\ = 1 + A(v/c) \langle J_z/J \rangle \cos \theta, \quad (1)$$

where  $\langle J_z/J \rangle = f_1(\mu B/Jk_B T) = \sum m N_m / J \sum N_m$  is the thermal average degree of nuclear polarization, i.e., the fractional magnetization of the nuclear spins of magnetic moment  $\mu$  and spin  $J$  at temperature  $T$  in the local magnetic field at the nucleus  $B$ ;  $\theta$  is the angle relative to the direction of nuclear spin  $J$  at which a  $\beta$ -particle of velocity  $\vec{v}$  is calculated to be emitted so as to reach the detector, in general taking into consideration the effect of the applied magnetic field on the trajectory (small in this case). The asymmetry coefficient  $A$  depends on the angular momenta of the initial and final nuclear states and, for mixed transitions, on the parameter  $y = C_V M_F / C_A M_{GT}$ , where  $M_F, M_{GT}$  are the Fermi and Gamow-Teller reduced matrix elements, respectively, and  $C_V, C_A$  are the vector and axial-vector coupling constants. Correction terms due to recoil effects<sup>11</sup> are negligible at the large mass numbers and low maximum electron energies involved here. Magnitudes of second order matrix elements have not been estimated but are assumed to have negligible effects, although the  $\log ft$  values for the  $\beta$  transitions studied (7.5 for  $^{60}\text{Co}$  and 6.7 for  $^{59}\text{Fe}$ ) are relatively large and indicate that the Gamow-Teller matrix elements involved are rather retarded.

Our experimental system and method of data reduction were tested by measuring the asymmetry coefficient of the 5(1)4, 314-keV allowed Gamow-Teller  $\beta$  transition of  $^{60}\text{Co}$ . With  $\mu = +3.754\mu_N$ <sup>12</sup> and  $B = -29.0$  T,<sup>13</sup> at temperatures between 25–30 mK  $\langle J_z/J \rangle = 0.47$ – $0.53$ . For  $\beta$  particles near the endpoint of the spectrum  $v/c = 0.78$ . Experimentally, we found  $A = -0.80 \pm 0.04$  at 475 Oe and  $-0.72 \pm 0.05$  at 950 Oe, compared to the theoretically expected value  $-1$ . The difference arises pre-

sumably because of electron scattering in the source, off the surroundings, and back out of the detector face, thereby providing an estimate of the corrections for these effects.<sup>14</sup> The attenuation due to detector solid angle and source extension, as well as to uncertainties in the proper value of  $v/c$  and possible incomplete parallelism of the iron hyperfine fields,<sup>15</sup> are also folded into the empirical correction factor this experiment provides. However, neither the  $\beta$  asymmetry nor the  $\gamma$  anisotropy showed any significant dependence on applied fields between 475 and 1140 Oe, indicating that even the lower fields were sufficient to line up the hyperfine fields at  $^{60}\text{Co}$  nuclei in domains of our *thin* (25.4  $\mu\text{m}$ ) foils. Purely magnetostatic considerations<sup>3</sup> indicate the importance of using foil thicknesses not exceeding 25.4  $\mu\text{m}$ , in order to minimize demagnetizing field effects.

When we substituted various iron foils containing  $^{59}\text{Fe}$  for those containing  $^{60}\text{Co}$  in our apparatus, we observed no 1099- or 1292-keV  $\gamma$ -ray anisotropy greater than 0.75%, in contrast to Ref. 1 but in agreement with Daly *et al.*<sup>16</sup> and Becker *et al.*<sup>17</sup> The  $\frac{3}{2}(0, 1)_{\frac{3}{2}}^{\pm}$ , 467-keV  $\beta$ -ray asymmetry [ $W(34^{\circ}) - 1$ ] was only  $(-0.8 \pm 0.5)\%$ <sup>18</sup> at  $T = 25\text{--}30$  mK. Using  $v/c = 0.85$  and the empirical scattering correction derived above yields

$$f_1 A(y) = f_1(-0.400 - 1.550y)/(1 + y^2) = -0.014 \pm 0.009. \quad (2)$$

For  $B = -33.9$  T,<sup>19</sup> pairs of values  $(\mu, y)$  consistent with Eq. (2) can be derived, using the leading term of the high-temperature expansion for  $f_1$ , from  $A\mu/\mu_N = +0.065 \pm 0.041$ . If  $\mu < 0$ , this relationship is consistent with a recent determination by Krane, Rosenblum, and Steyert<sup>20</sup> of  $|\mu(^{59}\text{Fe})| = 0.29 \pm 0.03$  from the anisotropy of the 192-keV  $\gamma$  ray and with previous  $\beta$ - $\gamma$  circular polarization correlation measurements by Mann *et al.*<sup>21</sup> which yielded  $y = -0.15 \pm 0.04$ . [If  $\mu$  were  $-1.1\mu_N$  (Ref. 1), Eq. (2) would imply  $y \approx -0.25$ .] Apparently the observation of the  $\beta$  rays from an iron source cooled by an adiabatic demagnetization cryostat to a few mK would be needed to make a significantly improved independent determination of  $y$  and  $\mu$ . The small value of  $\mu$  makes understandable the null results of Refs. 16 and 17, as well as of early attempts to observe  $\gamma$  anisotropies from oriented  $^{59}\text{Fe}$  in iron alum,<sup>22</sup> Armco iron,<sup>23</sup> and cerium magnesium nitrate.<sup>24</sup>

No convincing asymmetry could be determined for the 273-keV  $\beta$  transition, since this would require accurate extrapolation to low energies not only of the background but also of the observed 467-keV  $\beta$  spectrum. The difficulty of this spectrum stripping is illustrated by the results from

a mixed  $^{59}\text{Fe}$ - $^{60}\text{Co}$  sample which gave for the 314-keV  $^{60}\text{Co}$   $\beta$  rays  $A = -1.0 \pm 0.2$ . In any case we could find no evidence for a positive or negative asymmetry greater than a few percent, which is not in disagreement with the small moment of  $^{59}\text{Fe}$  and with the  $\beta$ - $\gamma$  circular polarization correlation results of Tirsell and Mann,<sup>25</sup>  $y = -0.04 \pm 0.05$ .

The results of Ref. 1 become even harder to understand now. For example, we have considered the possibility that axial electric quadrupole hyperfine coupling might provide most of the splitting in rare earth double nitrate crystals; however, the required splitting is at least a factor of 10 larger than what could reasonably be expected on the basis of estimates of the electric quadrupole moment of  $^{59}\text{Fe}$  ( $+0.15$  b<sup>26</sup>) and of the electric field gradient at the nucleus of a  $\text{Fe}^{3+}$  ion at any conceivable defect site in the structure.<sup>27</sup> Since the experiments of Ref. 1 were carried out with NaI(Tl) detectors and largely vacuum tube electronics, it must be considered possible that the 4–10%  $\gamma$  anisotropies reported earlier resulted from a spurious systematic effect<sup>17</sup> which escaped detection. In view of the low counting rates encountered with  $^{59}\text{Fe}$  as a trivalent impurity in double nitrate crystals, it does not seem desirable to attempt to repeat the experiments of Ref. 1 in order to check on these possible sources of error.

In conclusion, contrary to Ref. 1 there is now no evidence for large Fermi contributions to the  $^{59}\text{Fe}$   $\beta$  decay. Small  $|y|$  values  $< 0.2$  such as found by Mann and co-workers<sup>21,25</sup> are in agreement with the conventional picture that  $M_F$  arises from small isospin admixtures with  $T = \frac{7}{2}$  into the low-lying  $T = \frac{5}{2}$  states of  $^{59}\text{Co}$  due to a charge-dependent interaction.<sup>28</sup> The squares of the Gamow-Teller matrix elements for the various  $^{59}\text{Fe}$  allowed  $\beta$  transitions<sup>29</sup> can be derived from the relation<sup>30</sup>

$$|M_{\text{GT}}|_{\text{exp}}^2 = (4510 \text{ sec})/ft(1 + y^2). \quad (3)$$

The results are summarized in Table I. Rough estimates of the single-particle Gamow-Teller matrix elements<sup>31</sup>  $|M_{\text{GT}}|_{\text{sp}}^2$  were made on the basis of the only wave functions available to us: for the  $^{59}\text{Fe}$  ground state some intermediate coupling computations by Larner<sup>32</sup> and for the low-lying excited states of  $^{59}\text{Co}$  a unified anharmonic vibrational model calculation by Stewart, Castel, and Singh.<sup>33</sup> These values are listed in the last column of Table I. In the notation  $|NR, lj\rangle$  for the core phonon excitation number  $N$  and angular momentum  $R$  coupled to the odd particle with orbital quantum number  $l$  and total angular momentum  $j$  to give  $J^\pi$ ,  $|M_{\text{GT}}|^2 = |\langle f | \sum \vec{\sigma}_i \tau^+ | i \rangle|^2$  comes mainly from the  $|00, p_{3/2}\rangle$  and  $|12, p_{3/2}\rangle$  components of initial and final states; the  $|12, p_{3/2}\rangle - |12, p_{1/2}\rangle$  and  $|12, f_{5/2}\rangle$

TABLE I. Squared Gamow-Teller matrix elements  $|M_{\text{GT}}|^2$  for the allowed  $\beta$  transitions in the decay of  $^{59}\text{Fe}$  to final states in  $^{59}\text{Co}$ , compared to extreme single-particle model estimates including configuration mixing.

$E_{\beta}^{\text{max}}$ (keV)	$E_f$ (keV)	$J_f^{\pi}$	Branching (%)	$\log ft$	$ M_{\text{GT}} _{\text{exp}}^2$	$ M_{\text{GT}} _{\text{sp}}^2$
467	1099	$\frac{3}{2}^-$	53	6.7	$8.8 \times 10^{-4}$	0.69
273	1292	$\frac{3}{2}^-$	45	6.0	$4.5 \times 10^{-3}$	0.010
132	1434	$(\frac{1}{2})^-$	1.3	6.5	$1.4 \times 10^{-3}$	$2.0 \times 10^{-3}$
84	1482	$\frac{5}{2}^{-(+)}$	0.08	7.1	$3.8 \times 10^{-3}$	0.15

$-|12, f_{7/2}\rangle$  contributions are also important, with a great deal of destructive interference occurring especially in the 273- and 132-keV cases. Evidently other factors such as limited basis of  $|i\rangle$ , hindrance due to quasiparticle effects,<sup>34</sup> and possibly incompatibility of the core and particle-core interaction descriptions<sup>26,32,33,35</sup> may need to be considered, particularly for the 467- and 84-keV cases. This may become feasible with the advent of more refined theoretical treatments of the  $f$ - $p$  shell<sup>36</sup> combined with more experimental data on one-particle transfer reactions in this region.

It should be noted that our  $\beta$  asymmetry measurement is in principle sensitive to the *sign* of  $\mu$ , whereas the  $\gamma$  directional distribution and  $\beta$ - $\gamma$  circular polarization correlation are not. Though our observed effect is small, as was remarked earlier, it is consistent with Refs. 20 and 21 *only* for  $\mu < 0$ . Thus some support is provided by our result for the expected negative sign of  $\mu$ , a question of increased importance in view of the drastic reduction in the magnitude of the measured magnetic moment of this nuclide, well outside the trend of the empirical moments ( $\mu \sim \mu_N$ ). In order to examine the question of the sign of  $\mu$  theoret-

cally, we carried out some model calculations of various types,<sup>37-40</sup> with the numerical results quoted in Table II. For the rotational model the relation<sup>40</sup>

$$\mu = g_R J + (g_K - g_R) K^2 / (J + 1) \quad (4)$$

was used with  $K = j = J$ . The admixtures present in Larner's wave function<sup>32</sup> turn out to have very little effect on the magnetic moment. Only the configuration mixing approach of Noya, Arima, and Horie<sup>39</sup> yielded moments as small as  $-0.29^{20}$  with a weak dependence on the singlet nucleon interaction strength parameter  $C$  when chosen in a range appropriate to the model potential. However, in view of the approximations in Ref. 39 such estimates<sup>20</sup> must be considered uncertain within at least a few tenths  $\mu_N$ .

We wish to acknowledge useful advice on the dilution refrigerator design from O. E. Vilches and B. M. Abraham; the loan of electronic equipment by R. W. Krone; discussions of the nuclear properties of  $^{59}\text{Fe}$  and  $^{59}\text{Co}$  with P. Goldhammer and

TABLE II. Magnetic moment estimates for  $^{59}\text{Fe}$  on the basis of various nuclear models, assuming a ground configuration  $\pi(1f_{7/2})^6 \nu(2p_{3/2})^3 (1f_{5/2})^2$ .

Model	Ref.		$\mu(^{59}\text{Fe})/\mu_N$
Extreme single particle	37	Free neutron $g_l, g_s$	-1.913
Independent particle, configuration mixing	38	$A_{3/2}^1 = -3.00, \xi_1 = -0.38$	-0.77
$jj$ coupling, configuration mixing	39	Harmonic oscillator $C=25$ MeV	-0.39
		$C=40$ MeV	-0.30
		Square well $C=60$ MeV	-0.51
		$C=90$ MeV	-0.30
Rotational, deformed core	26	$\delta = 0.14, K = 3/2, g_R = Z/A$	-0.88
Vibrational, spherical core, intermediate coupling	32	Ground state $\psi$ admixtures	-1.76
Experimental	20	$y, \delta$ from $\beta$ - $\gamma$ cp and $\gamma$ - $\gamma$ correlations	-0.29 $\pm 0.03$

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correspondence with S. S. Rosenblum. We are especially indebted to D. A. Larner and to K. W. C. Stewart for generously providing the unpublished details of their wave functions.

\*Assisted by Office of Naval Research Contract N00014-72-C-0219 and initially by National Science Foundation Grant GP-8133, as well as by the University of Kansas Graduate Research Fund. K. P. B. was the holder of a Graduate School Honors Fellowship.

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