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Nuclear Orientation of Iron-59 in Rare-Earth Double-Nitrate Crystals*

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Anisotropic emission of 1.10- and 1.29-MeV γ rays from Fe^{59} oriented in clear single crystals of Ce-Zn and Nd-Zn nitrates cooled by adiabatic demagnetization has been studied as a function of temperature in fields of 0 and 250 Oe. From the temperature dependence of the γ anisotropy is extracted an estimate of the nuclear magnetic dipole moment of the ground state of Fe^{59} of $(1.1 \pm 0.2)\mu_N$. Analysis of the data in terms of the currently accepted spin sequence $\frac{3}{2}(\beta)\frac{3}{2}(\gamma)\frac{7}{2}$ for both γ rays, assuming the fraction of Fe^{59} aligned in zero field is 0.30 ± 0.03, requires either (i) the hypothesis of Fermi contributions in excess of 30% (contrary to conserved-vector-current theory) or, alternatively, (ii) enhancement factors $Q_2 = 5.0 \pm 2.6$ and 6.7 ± 2.4 for intermediate-state reorientation in the 1.10- and 1.29-MeV levels, respectively. Admixtures of $M3$ radiation of the order 1–4% may also be involved in the interpretation.

I. INTRODUCTION

The angular distribution of γ radiation from oriented radioactive nuclei provides independent information on spin assignments, β-decay matrix elements, and γ-ray multipolarities which supplements that obtained from internal-conversion coefficients, γ-γ angular correlations, and β-γ circular polarization correlations.[1] Thus, despite the relatively large number of experimental studies[2-17] of the Fe^{59} decay, the parameters assigned the two principal decay channels *via* the 1.10- and 1.29-MeV levels in Co^{59} have undergone continual evolution and modification in the past 18 years as new information became available, and so we decided to carry out an experiment to orient Fe^{59} by the method suggested by Culvahouse and Olsen.[18] This method is reviewed and the theoretical framework set up in Sec. II, after which the experimental data are presented in Sec. III and their implications discussed in Sec. IV.

II. NUCLEAR-ORIENTATION METHOD

Spin-Hamiltonian coefficients corresponding to sharp-line electron paramagnetic resonance (EPR) spectra of Fe^{3+} ions in two sites in La-Zn nitrate (LZN) and Ce-Zn nitrate (CZN) have been measured by Culvahouse and Olsen.[18] The essential feature of these data is that D, the coefficient of the second-degree axial crystal field term $D(S_z^2 - \frac{35}{12})$, is negative and much larger in magnitude than either the fourth-degree term or the isotropic hfs coefficient A of Fe^{57}, just the condition discussed by Bleaney[19] for nuclear orientation described, as a result of the $M_s = \pm\frac{5}{2}$ states lying lowest, by an effective spin Hamiltonian

$$H = g'_{\parallel}\beta H_z S'_z + g'_{\perp}\beta(H_x S'_x + H_y S'_y) + A' I_z S'_z + B'(I_x S'_x + I_y S'_y)\,, \tag{1}$$

with

$S' = \frac{1}{2}, \quad g'_{\parallel} = 5g \gg g'_{\perp}, \quad g = 2.003(2);$

$I = \frac{3}{2}, \quad A' = 5A \gg B';$

$A/k = 1.57(4)$ mK for $Fe^{57}(I = \frac{1}{2})$. The z axis is also the c axis of the double nitrate crystal structure.[20] When $H_z = 0$, the lowest-lying hfs levels are four doubly-degenerate states equally spaced by $\frac{1}{2}|A'|$, and if the temperature T is low enough to populate just the ground state with $m_I = \pm\frac{3}{2}$, axial nuclear alignment of Fe^{59} is produced. Application of a few hundred oersteds along the z axis gives rise to nuclear polarization at virtually the same T, with very little change in the degree of alignment.

Because Fe^{3+} ions do not easily enter lattice sites in the double-nitrate crystal structure, a significant fraction of ferric ions containing Fe^{59} will be incorporated in small pockets of frozen solution within the crystal.[21] These might be expected to be approximately randomly oriented in zero field, but some nuclear polarization should be produced by a sufficiently strong H_z.[18] Unfortunately, the complexity of the Fe^{3+} spectrum precludes an accurate determination by EPR of Fe partitioning in CZN, as was possible for Cr^{3+} partitioning in Ce-Mg nitrate (CMN), [22-25] though Culvahouse and Olsen[18] suggest it is of like magnitude. However, in view of the near equality of chromic and ferric ion radii and of the similar partitioning effects for Cr^{3+} in LZN and LMN,[25] we may reasonably expect the fraction f of ferric ions incorporated in crystal sites in CZN crystals of good visual clarity to be comparable to corresponding fractions for chromic ions in CMN, i.e., $0.26 < f < 0.32$.[24,25] Cloudy crystals containing Fe^{59} do not exhibit detectable γ-ray anisotropy,[26] presumably because of the smaller value of f and the difficulty of producing sufficient nuclear polarization with modest fields.[18]

We represent the γ-radiation pattern $W(\theta)$ by an expansion in Legendre polynomials, $1 + A_2P_2(\cos\theta)$. No terms beyond P_2 are expected, because the spin of Fe^{59} is[13] $\frac{3}{2}$. The experimental value of A_2 should be corrected for the detector solid angle[27] by a factor estimated from the graphs of Stanford and Rivers[28] to be 0.94 for 1.2-MeV radiation incident on our 1-in.-diam × 1-in.-thick NaI(Tl) detectors 3.5 cm from the center of the source.

The theoretical coefficient A_2 is composed of a number of factors[29]:

$$A_2 = fB_2^s U_2 Q_2^s F_2 [1 + (1-f) B_2^p Q_2^p / f B_2^s Q_2^s]. \tag{2}$$

As mentioned above, $f = N_s/N$ stands for the fraction of Fe^{59} nuclei oriented in crystal sites, and we have assumed $f = (0.3 \pm 10\%)$ is a reasonable value. Then $(1-f)$ is the fraction of Fe^{59} in frozen solution pockets which are presumed to contribute to the γ-ray anisotropy only when $H_z \neq 0$, for which we must add to A_2 the second term on the right-hand side of Eq. (2). B_2^s and B_2^p are functions of $A'/2kT$ characterizing the nuclear orientation in sites and pockets, respectively; for either, $B_2 = \sum m^2 N_m/N - \frac{5}{4}$ with $m = \pm\frac{3}{2}, \pm\frac{1}{2}$, and N_m is the thermal equilibrium population of the substate $|m\rangle = |I_z\rangle$.

Throughout our discussion we shall assume spin-spin interaction[30] has negligible influence on the nuclear orientation of Fe^{59}. This is certainly a good assumption for Fe^{59} nuclei in lattice sites,[18] since these correspond effectively to the case[19] $g'_{\parallel} \gg g'_{\perp}$, $A' \gg B'$. However, such interaction would clearly have to be included in any detailed consideration of Fe^{59} in frozen solution pockets.

Factors U_2 and F_2 represent the β and γ transitions, respectively.[29,31] For a decay sequence $I_0(i_\beta)I_1(L_\gamma)I_2 = \frac{3}{2}^-(0,1)\frac{3}{2}^-(2,3)\frac{7}{2}^-$ appropriate[15,17] to either principal decay channel of Fe^{59}, $U_2 = (0.200 + y^2)/(1 + y^2)$ with $y = C_V M_F / C_A M_{GT}$ and $F_2 = (-0.143 + 0.926\delta + 0.500\delta^2)/(1 + \delta^2)$ with $\delta = \pm(M3/E2)^{1/2}$. For pure Ganow-Teller β decay and $E2$ γ rays, $U_2F_2 = 0.0286$.

Finally, Q_2 represents possible intermediate state reorientation. Though several past attempts in this laboratory to detect reorientation ($Q_2 \neq 1$) of states with nanosecond half-lives in Fe^{57}, La^{139}, and Pr^{141} have not revealed any large effects,[32,33] we retain this factor in anticipation of the discussion below. Also, we note that Agarwal, Baba, and Bhattacherjee[15] found perturbation of the 0.14–1.29-MeV γ-γ directional correlation in a metallic iron source, giving $Q_2 = 0.33$ relative to a dilute chloride solution.

Putting all these factors together for the decay scheme $\frac{3}{2}(GT)\frac{3}{2}(E2)\frac{7}{2}$, we should observe an A_2 no larger than -0.008, with $B_2 = 1$, $Q_2 = 1$, and no contribution from solution pockets. This would provide a severe test of the counter stability, especially in view of the unfavorable partitioning effect on Fe^{3+} and the consequent low counting rates. Surprisingly, the data presented in the next section show values of A_2 four to six times larger than this.

III. EXPERIMENTS

A. Samples

Trivalent ions containing the isotope Fe^{59} were incorporated into natural-shaped double-nitrate crystals grown from saturated aqueous solutions containing stoichiometric portions of high-purity (99.999%) Lindsay rare-earth nitrates and Fisher "Certified" reagent-grade zinc nitrate. The ratio of iron to rare earth in solution was about 10^{-4}. High specific activity Fe^{59}, obtained from Tracerlab in the form of ferric chloride in 1N HCl, was converted to the nitrate by heating the original solution to dryness and redissolving the residue in 1N HNO_3. Without adding carrier, the activity ($Fe^{59}/Fe = 2\times10^{-4}$) was added into the growing solution.

In crystals grown from aqueous solutions, a portion of the crystal volume is occupied by minute pockets of the solution, the total volume of pockets depending on the conditions under which a crystal was grown. The crystals we studied were grown slowly over a period of several days in a refrigerator and were very clear, minimizing the percentage of the crystal volume consisting of solution pockets for the reason mentioned in Sec. II. NMR measurements on similar clear double-nitrate crystals have given an estimate of less than 2% of the crystal volume occupied by solution pockets.[22,25] The fact that ferric ions are not readily assimilated into the double-nitrate crystal structure was quite apparent; for example, in the first crystal 0.5% of the activity in the growing solution was incorporated into a crystal that contained 12% of the total solute.

We experimented with two crystals containing Fe^{59}. The first, more extensively studied one, was a 1.18-g CZN crystal containing initially 3 μCi of Fe^{59}; the other was a 1.28-g neodymium zinc nitrate (NZN) crystal in which were 5 μCi of Fe^{59}. With the CZN crystal we were able to investigate the orientation of Fe^{59} nuclei at values of $1/T$ up to[34-36] approximately 650 K^{-1}. The NZN crystal was used to study the orientation for smaller $1/T$ values of about[37] 100 K^{-1}, since the presence of 20% odd-A Nd isotopes contributes a large hyperfine heat capacity which limits the lowest temperature attainable.

B. Measurements

Fe^{59} nuclei were oriented by the mechanism discussed in Sec. II in CZN and NZN natural single crystals cooled by adiabatic demagnetization from initial field and bath conditions up to 21.7 kOe/K, using repeated demagnetizations from a limited selection of fields to build statistics. The demagnetization field was applied perpendicular to the crystal axis, in which direction the Ce^{3+} and Nd^{3+} g factors are largest. After cooling, the crystal was in zero magnetic field for nuclear alignment, or placed in magnetic fields up to 250 Oe parallel to the crystal axis for nuclear polarization.

Measurements carried out on the 1.10- and 1.29-MeV γ rays included their angular distributions $W(\theta)$ at the lowest temperature attainable with each salt and the temperature dependence of the anisotropy with $H_z = 0$ and 250 Oe. For a description of the general experimental techniques used in our laboratory, see Refs. 30 and 32. The major variation in procedure involved our use of a 400-channel analyzer (TMC model 404) operated in the "multichannel mode." A complete spectrum was accumulated for each of two counters in 100 channels, so that values of $W(\theta)$ could be determined for any portion of the spectrum. The spectra were accumulated for four minutes of live time immediately following magnetic cooling; when the crystal had returned to 1.1 K (15–30 min), a normalizing spectrum was then accumulated for the same period of live time. All counts in channels lying between the half-maximum points on the photopeak were summed. The effect of extrapolating the counting rates back to time of demagnetization was estimated to be negligible.

Having the entire γ-ray spectrum permitted correction of the apparent 1.10-MeV anisotropy because of the Compton distribution of the 1.29-MeV γ ray lying under the 1.10-MeV peak. This Compton distribution was constructed from the spectrum

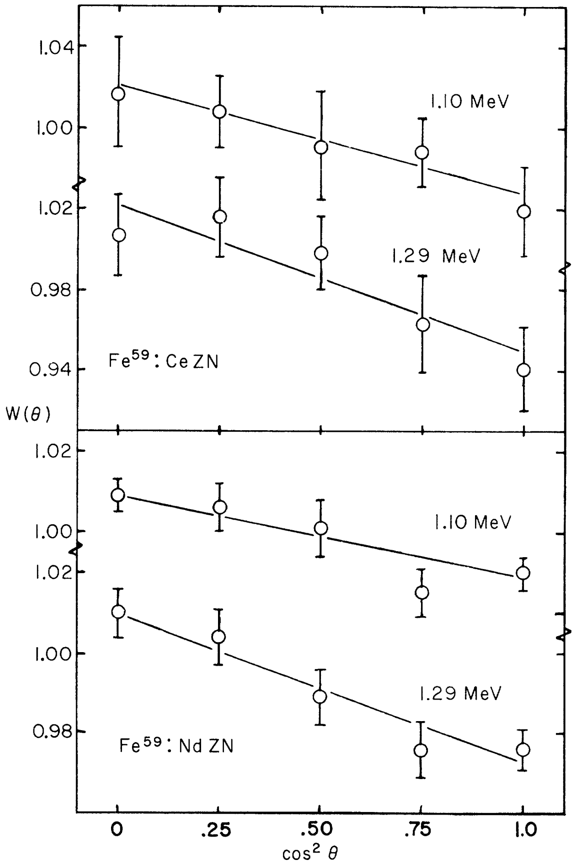

FIG. 1. Radiation patterns $W(\theta)$ versus $\cos^2\theta$ of 1.10- and 1.29-MeV γ rays from Fe^{59} aligned in Ce-Zn (top) and Nd-Zn (bottom) nitrate single crystals cooled by adiabatic demagnetization from 21.7 kOe/K. The straight lines represent best computer fits to $A_0 + A_2P_2(\cos\theta)$.

of 1.275-MeV γ rays from a Na^{22} source. In experiments[37] with Co^{60} in NZN, we verified that the anisotropy of the upper portion of the Compton distribution is the same as for the photopeak. A background correction of about 2% was also applied to the counting rate. An independent check on the functioning of the entire apparatus was afforded by the Co^{60} measurements.[37]

C. Angular Distributions

Radiation patterns obtained at five angles from 0 to 90° with respect to the crystal axis for demagnetizations from 21.7 kOe/K to a final field of zero are given in Fig. 1. No counter solid-angle correction has been made. Computer fits of $W(\theta)$ are listed in Table I. All patterns (except one) are linear in $\cos^2\theta$, conforming to the expectation of a $P_2(\cos\theta)$ pattern for spin $\frac{3}{2}$ (Sec. II); addition of a $P_4(\cos\theta)$ term did not significantly improve the normalized χ^2 of the fit, nor did the coefficients A_4 differ from zero by more than one standard deviation. The exceptional pattern is that of 1.29-MeV radiation from the CZN sample, for which the low point at 90° favors $A_2 = -0.040 \pm 0.019$, $A_4 = -0.022 \pm 0.019$ with $\chi^2 = 0.17$. We do not know of any way in which a nonzero A_4 could arise for this decay scheme other than by experimental error in the 90° point, and have therefore preferred to use the value of A_2 listed in Table I.

Note that all patterns have $A_2 < 0$, as expected for axial alignment and predominantly $E2$ radiations. However, the magnitude of anisotropy is about four to six times larger than anticipated in Sec. II for pure GT and $E2$ transitions.

D. Temperature Dependence of the γ-Ray Anisotropy

The theoretical temperature dependence of the γ anisotropy is contained entirely in the orientation parameter B_2 (in the absence of temperature-dependent intermediate-state reorientation). Since B_2 is a function of the ratio $5A/2kT$, we used the temperature variation of A_2 to estimate the isotropic hfs constant A. The same scintillation detectors and procedures described above were used for this measurement. With one detector at 0° and the other at 90°, both $W(0°)$ and $W(90°)$ could be obtained simultaneously, and by rotating the table on which the detectors were mounted we could interchange their roles. These data for Fe^{59} in CZN are presented in Fig. 2, plotted as $-A_2 = 2[W(90) - W(0)]/3$ versus $1/T$ on the scale of Hudson and Kaeser.[35] Again, no counter solid-angle correction was applied.

Because of the faster warmups of CZN from higher temperatures, spectra from demagnetizations from less than 14 kOe/K were accumulated for only one minute of live time. The consequent larger uncertainties made these data points rather im-

TABLE I. Best-fit coefficients in the experimental angular distribution $W(\theta) = A_0 + A_2 P_2(\cos\theta)$ of γ rays from decay of Fe^{59} aligned in rare-earth double-nitrate crystals adiabatically demagnetized from 21.7 kOe/K to zero field. No counter solid-angle corrections have been applied.

Crystal	E_γ (MeV)	A_0	A_2	χ^2
Ce-Zn nitrate	1.10	1.003 ±0.011	−0.036 ±0.019	0.12
	1.29	0.998 ±0.010	-0.048_5 $\pm0.017_5$	0.53
Nd-Zn nitrate	1.10	1.0021 ±0.0024	−0.0142 ±0.0036	0.80
	1.29	0.9976 ±0.0032	−0.0243 ±0.0048	0.50

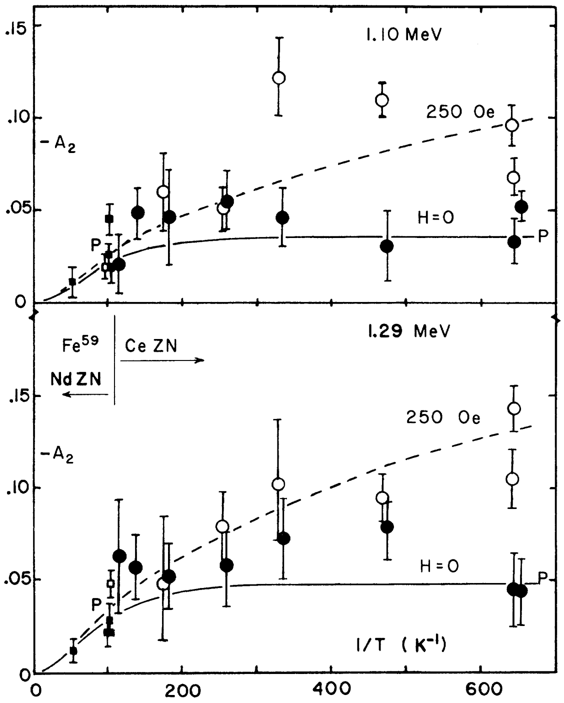

FIG. 2. Anisotropy parameter ($-A_2$) for 1.10- and 1.29-MeV γ rays from Fe^{59} oriented in Ce-Zn (circles) and Nd-Zn (squares) nitrate single crystals cooled by adiabatic demagnetization to final fields of zero (solid symbols) and 250 Oe (open symbols), the field being parallel to the crystal axis. Inverse temperatures are from Hudson and Kaeser's calorimetric scale for Ce-Mg and from a Co^{60} scale for Nd-Zn nitrate. Curves represent theoretical dependences adjusted to pass through low- and high-temperature points, with hyperfine constant $A/k = 6.2 \pm 1.1$ mK. (P indicates a full pattern was taken at this temperature.)

precise. To investigate this temperature range more carefully and to provide an independent confirmation of the surprisingly large anisotropy we were observing, we put Fe^{59} in NZN for our second sample. However, in order to correlate such data with those for CZN, we needed a temperature scale for NZN. Cooke, Meyer, and Wolf[38] had studied Nd-Mg nitrate up to 9.5 kOe/K, and in another experiment had demonstrated equality of T and $T^{\otimes}$ for $T > 0.05$ K. Neither of these efforts extended to sufficiently low temperatures for our purposes, so we generated a provisional scale, using the alignment of Co^{60} as a convenient thermometer. Because the spectrum of Co^{60} was so intense and is so similar to that of Fe^{59}, the thermometry had to be performed on another NZN crystal, to all appearances identical with the first. The details of the Co^{60} in NZN experiments and the resulting temperature scale are presented in Ref. 37. The largest value of $1/T$ attained for NZN demagnetized from 21.7 kOe/K was 105 ± 5.

Data points for Fe^{59} in NZN cooled from four initial fields to a final field of zero are included in Fig. 2. Though low counting rates again limit severely the precision, these anisotropies fit rather well into theoretical $B_2(T)$ trends toward the low-temperature CZN data, but fall well below the high-temperature CZN points.

Fitting of the solid curves in Fig. 2 to the temperature dependence of zero-field anisotropy data for Fe^{59} in NZN and CZN permits a rough but useful first estimate of the nuclear magnetic dipole moment μ of the ground state of Fe^{59}. Those curves correspond to a fit assuming the T dependence of A_2 arises solely from $B_2(T)$. If $B_2 = 0.50$ at $5A/2kT = 1.32$ corresponds to $1/T = 85 \pm 15$, we obtain $|A|/k = 6.2 \pm 1.1$ mK for Fe^{59}. Culvahouse and Olsen[18] measured $|A|/k$ for Fe^{57} in double-nitrate sites to be 1.57 ± 0.04 mK; also, $\mu(Fe^{57}) = 0.0903\mu_N$.[39] Using the proportionality between A and (μ/I) for the two isotopes enables us to calculate that

$$|\mu(Fe^{59})| = 1.1 \pm 0.2\mu_N .$$

This may be compared with the Schmidt limit of $1.9\mu_N$ for a neutron hole in a $p_{3/2}$ single-particle state; the empirical moments lie between 0.5 and $1.1\mu_N$.[40]

E. Nuclear Polarization

γ-ray anisotropies were also measured with the external magnetic field of 250 Oe applied along the crystal axis. The data in this case were taken as before, except that it was not possible to interchange the positions of the two counters since the counter and field coil assembly formed a rigid unit. Light pipes and extra magnetic shielding were added to the counter assemblies for the experiments with a magnetic field on.[30]

Anisotropies at several temperatures with 250-Oe field applied to CZN and NZN are shown in Fig. 2. The magnitude of anisotropy is larger, presumably because of contributions from Fe^{59} in pockets, and its variation with temperature is again largely what is expected for $B_2(T)$. (An exception is the lowest-temperature point associated with each γ ray. There is an apparent sharp rise for 1.29 MeV, reflected through the Compton correction mentioned above in a possibly spurious depression of the 1.10-MeV point.) The curves drawn in Fig. 2 represent fits to the lowest-temperature experimental data with a temperature variation corresponding to contributions of the form $B_2(5A/2kT)$ from $(Fe^{59})^{3+}$ in sites and $B_2(A/2kT)$ from pockets of frozen solution. This form of T dependence, though not rigorously justifiable, provided a reasonable approximation to the behavior of Cr^{51} in Ce-Mg nitrate.[25]

The relative contributions of Fe^{59} in pockets can be deduced from the increase in anisotropy upon application of the 250-Oe field. Within rather broad limits, these contributions appear to be approximately the same for each γ ray. A weighted average yields $B_2^p Q_2^p(H = 250)/B_2^s Q_2^s(H = 250) = 0.63 \pm 0.32$, which may be compared, if $Q_2^p \cong Q_2^s$, with $B_2^p/B_2^s \cong B_2^s(A/2kT)/B_2^s(5A/2kT) = 0.73$.

IV. RESULTS AND DISCUSSION

In Table II we summarize the values of the anisotropy parameter A_2 for various conditions, consistent with the results presented in Figs. 1 and 2 and Table I. The third column includes the correction for counter solid angle; the fourth column shows the further corrections for cross feeds in

TABLE II. Coefficients of the P_2 term in the angular distribution of γ rays from the decay of Fe^{59} oriented in rare-earth double-nitrate crystals.

E_γ (MeV)	H_z (Oe)	A_2	$A_2/0.98B_2$	$A_2/0.98B_2f$
1.10	0	-0.038[a]	-0.042_5[b]	-0.142[c]
		± 0.020	± 0.022	± 0.075
	250	-0.104	-0.107	-0.356
		± 0.023	± 0.024	± 0.080
1.29	0	-0.051_5	-0.058	-0.192
		± 0.019	± 0.021	± 0.069
	250	-0.138	-0.142	-0.472
		± 0.018	± 0.019	± 0.063

[a] Corrected for counter solid angle by dividing A_2(exp) by 0.94.

[b] $B_2^s = 0.99$; correction for cross feeds from other decay branches = 0.98.

[c] Using $f = 0.30 \pm 0.03$.

the decay scheme, and for a nearly saturated B_2^s.

It is perhaps of interest at this point to note that such anisotropies would be easy to account for with a $\frac{3}{2}$-$\frac{5}{2}$-$\frac{7}{2}$ decay scheme, since $U_2 = 0.748$ and $F_2 = (0.134 + 1.289\delta + 0.325\delta^2)/(1+\delta^2)$. Solutions with $\delta(1.10\text{ MeV}) = (E2/M1)^{1/2} = -0.23$ and $\delta(1.29\text{ MeV}) = -0.32$ would have been tempting were it not for the work of Blair and Armstrong[12] which ruled out the $\frac{5}{2}$ spin assignments.

As Eq. (2) shows, the γ-ray anisotropy only gives the product $U_2F_2Q_2^s(H=0)$. To make further progress, we must disentangle these three factors, which requires the consideration of alternative assumptions about the possibility of intermediate-state reorientation (ISR) in sites.

A. No Intermediate-State Reorientation

If we assume $Q_2 = 1$, on the basis of some earlier experience with cases involving subnanosecond lifetime states,[32,33] the products U_2F_2 for each γ ray follow immediately from the first and third entries in the last column of Table II. In combination with the β-γ circular polarization-direction correlations of Mann *et al.*,[11] the Fermi contribution and $M3$ mixing can in principle be determined.

Figure 3 presents graphically the ranges (shaded areas) of the parameters $y = C_VM_F/C_AM_{\text{GT}}$ and $\delta = (M3/E2)^{1/2}$, which simultaneously satisfy the requirements of our nuclear-orientation data, the results of Ref. 11, and the internal-conversion coefficients of Ref. 2. Only the branches corresponding to $y < 0$ are shown. The two curves adjacent, on either side, to the one labeled $Q_2 = 1$ define the ranges permitted by the relatively large experimental errors in our nuclear-orientation data. Likewise, the lines labeled IC define the extreme limits of $|\delta|$ consistent with Metzger's internal-conversion coefficients,[2] including experimental errors.

Inspection of Fig. 3 makes it clear that for either γ ray, no $M3$ admixture can alone harmonize all the data. On the other hand, for either β ray the required range of Fermi contributions is 30–100%, disturbingly outside the limits of a few percent[41-44] imposed by the conserved-vector-current (CVC) theory of β decay.[45,46] Therefore, we must consider the possibility of ISR.

B. Intermediate-State Reorientation

Consider entries one and three in column four of Table II under the assumption that the β-γ cascade is in both cases GT-$E2$, for which $U_2F_2 = -0.0286$. Then $Q_2^s = 5.0 \pm 2.6$ for the 1.10 and 6.7 ± 2.4 for the 1.29-MeV γ ray. (Even in the unlikely limit $f = 1$, $Q_2^s = 1.5 \pm 0.8$ and 2.0 ± 0.7, respectively.) The similar magnitudes of these two values are somewhat surprising in view of the gross disparity of the half-lives of the intermediate states; for the 1.29-MeV level, $T_{1/2} = 0.58$ nsec,[14-16] while for the 1.10-MeV state, $2.9 < T_{1/2} < 14$ psec.[16,17] (Discussion of the nature of these, and other, excited states of Co^{59} may be found in Refs. 15, 17, and 47.) Such a result seems not quite impossible, however, if the enhancement were to be produced by a large but very short-lived coherent perturbation induced by β emission. Possible mechanisms include recoil and "shake-off" effects leading to large dynamic magnetic and electric hyperfine interactions,

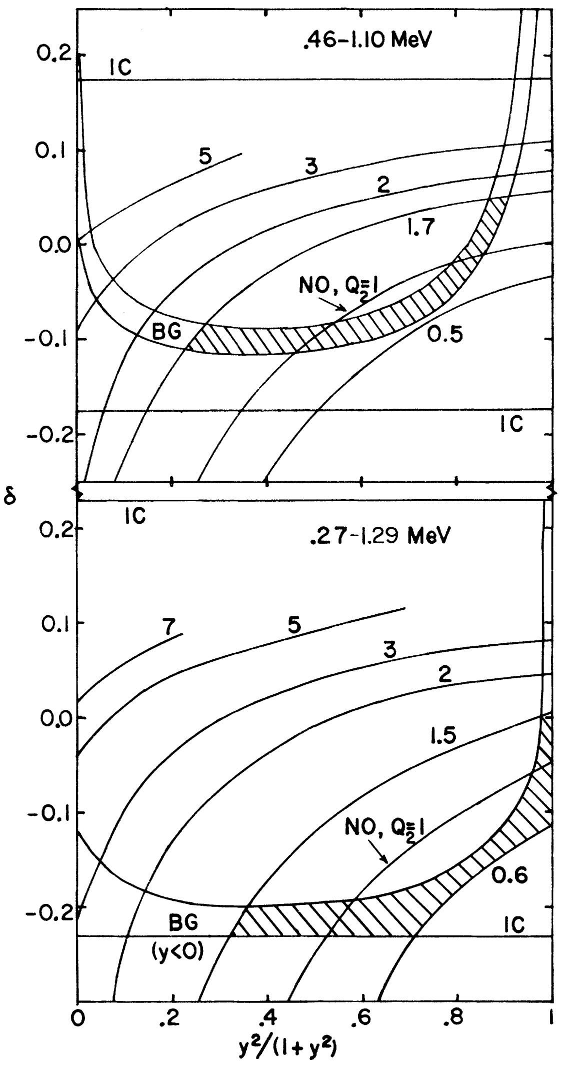

FIG. 3. γ-ray mixing amplitude $\delta = \pm(M3/E2)^{1/2}$ versus fractional Fermi contribution $y^2/(1+y^2)$ to the β transtion for the 0.46–1.10- and 0.27–1.29-MeV β-γ decay chains of Fe^{59}; $y = C_VM_F/C_AM_{\text{GT}} < 0$. The shaded areas represent values which are consistent with nuclear orientation (NO), β-direction γ-circular polarization correlation (BG), and with internal conversion (IC). The various NO curves are labeled with values of Q_2, assuming one third of Fe^{59} is in lattice sites.

but unfortunately no theoretical treatments of the possible influence of such dynamic effects in solids which are precisely relevant to the case at hand have yet been published. Most treatments[48-51] of ISR focus on static magnetic hfs interaction, to which it is more appropriate to refer in connection with long-lived electronic hole states in insulators after electron-capture decay. It is very difficult to see how such static perturbations could lead to substantial *enhancements* at low temperatures, since *attenuation* is their normal consequence.[52] Small enhancements can arise in static hfs perturbation of nuclear orientation at low temperatures through the assumption of coherence between electronic and nuclear intermediate states for particular combinations of the sign of hfs coupling and of initial and intermediate-state spins.[48, 50, 51]

Very recently, Misra[53] has presented the first theoretical treatment of the effect of recoil following β emission. His calculations were necessarily limited to a specific model case, that of $Fe^{2+}(S=2)$ in a noncentrosymmetric site and a hypothetical nuclide with $I=1$, but as neither are far from the present case of $S=\frac{5}{2}$, $I=\frac{3}{2}$ his results may be thought to be a reasonable guide for our considerations. On the other hand, the site symmetry and anisotropy of the terms in the perturbing Hamiltonian which couples the electron spin to the vibrating surroundings play crucial roles in his results, and because the nature of the sites occupied by Fe^{3+} ions in CZN is not known,[18] it is practically impossible to evaluate the relevance of this recoil theory. We do note, however, that both attenuation ($Q_2 < 1$) and enhancement ($Q_2 > 1$) are found by Misra.

C. Magnetic Octupole Admixture

Of course, the preceding subsections have considered extreme possibilities, and one may properly inquire as to whether some intermediate combinations of parameters may provide a reasonable explanation. Inspection of Fig. 3 in the limited range of not more than a few percent Fermi contribution to the β transitions, taking into account the experimental error in the nuclear alignment data, suggests compatibility of all results with $M3$ admixtures of about 1% for the 1.10 and 4% for the 1.29-MeV γ radiation, if we accept Q_2 of the order 5–7 without further attempting at present to delineate the ISR mechanisms. Such γ admixtures are easily accommodated to the angular correlations of Heath *et al.*[5] with acceptable $E2/M1$ contributions of approximately 2% in the 192- and 1% in the 145-keV transitions. However, such $M3$ admixtures in the 1.10- and 1.29-MeV transitions would require not only $E2$ retardation factors of the order 10^{-2} but also $M3$ enhancement factors of at least 10^5, relative to Weisskopf single-particle transition probabilities; such $M3$ enhancements are at least 2 orders of magnitude greater than would reasonably be expected on the basis of experiments to date.[54]

It is clearly necessary to have further independent measurements to reduce the ambiguities in the above interpretation. Perhaps the most fruitful set of experiments could be carried out on Fe^{59} polarized in an iron foil[55] at temperatures near 10 mK. Investigation of the β-ray asymmetry should determine the Fermi contribution, and hence U_2, unambiguously; simultaneous study of the γ-ray anisotropy will yield an F_2Q_2 product of each transition, bearing in mind that we should anticipate the possibility of perturbation in the 1.29-MeV state with a metallic iron source.[15] Development of a He^3 dilution refrigerator[56] for these experiments is now in progress in our laboratory, but in view of the considerable delay expected, we present these results now.

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