Nuclear Orientation of Mn^{54} and Mn^{52m} *†

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The spins of the nuclides Mn^{54} (290 days) and Mn^{52m} (21 min), incorporated in the lattice of cerium magnesium nitrate and nickel fluosilicate crystals, have been oriented at low temperatures. The measurement of the angular distribution of the gamma rays from Mn^{54} indicate the spin of Mn^{54} to be 3 or 2; in the latter case, the beta decay must be predominantly of the Fermi type with an upper limit of 10% of a possible Gamow-Teller admixture. From a simultaneous measurement of the angular distribution of the gamma rays from Mn^{54} and 5.7-day Mn^{52} the ratio of the nuclear g values of the two isotopes is found, which yields a magnetic moment for Mn^{54} of 2.55±0.21 nm (for spin 3) or 2.16±0.26 nm (for spin 2). The circular polarization measurements of the gamma rays from Mn^{54} determine this moment to be positive. Similar angular distribution experiments on Mn^{52m}, assuming spin 2, yield a magnetic moment of 1.04±0.16 nm if the beta decay is predominantly a Gamow-Teller transition, or 0.72±0.16 nm for a pure Fermi transition. These results indicate that the nuclear g values of the 21-min and the 5.7-day states of Mn^{52} are about the same.

I. INTRODUCTION

EXPERIMENTS by several groups have shown that manganese isotopes incorporated in the lattice of paramagnetic cooling salts can be oriented by the low-temperature method. Using cerium magnesium nitrate with an external field of several hundred gauss, this technique (Gorter-Rose method) was used to produce nuclear polarization of Mn^{54} by Grace and co-workers,[1] of 5.7-day Mn^{52} by Huiskamp and collaborators,[2] and of the relatively short-lived 2.6-hr Mn^{56} by Bauer and Deutsch.[3] Recently Dagley *et al.*[4] worked with nickel fluosilicate to align Mn^{56} utilizing the internal crystalline field to produce nuclear orientation (Bleaney method).

The purpose of the present work was to extend the magnetic hfs orientation methods to the investigation of the 21-min excited state of Mn^{52} (subsequently referred to as Mn^{52m}). Since the short lifetime of this nuclide made it impractical to incorporate it directly into the coolant crystals, we utilized the fact that Mn^{52m} can be derived by radioactive decay from an 8-hr parent, Fe^{52}, which was incorporated in the cooling salt. The magnetic moment of Mn^{52m} was obtained from a gamma-ray angular distribution comparison measurement with Mn^{54}, while the magnetic moment of the latter was found in turn by a comparison with 5.7-day Mn^{52}. The sign of the moment of Mn^{54} was measured by the observation of the circular polarization of the gamma rays.

II. DECAY SCHEMES AND FORMALISM

The decay chain $Fe^{52} \rightarrow Mn^{52} \rightarrow Cr^{52}$ has been the subject of many investigations in the past and the principal features of the decay schemes of Fe^{52} and Mn^{52} are well established (see, for instance, Strominger *et al.*[5] and Juliano *et al.*[6]); Fig. 1(a) is based on this together with the results of the present experiments.

A word may be said concerning the assignment of spin 2 to Mn^{52m}. The allowed beta decay to the 2+ level in Cr^{52} determines the value as $I=1$, 2, or 3. The allowed beta decay of Fe^{52} to the 555-kev level in Mn^{52} implies a spin of 0 or 1 for the latter. We have attempted

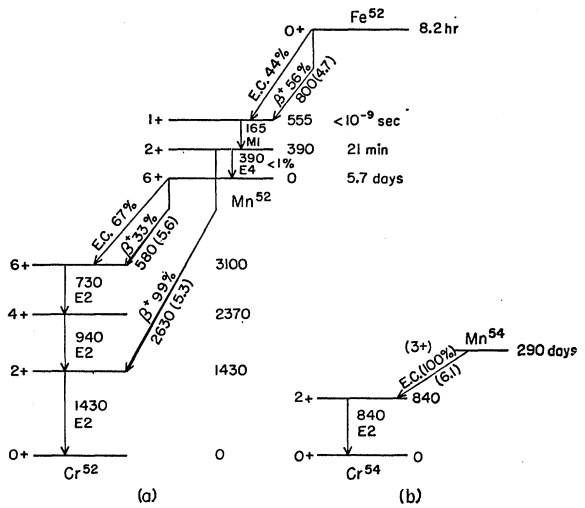

FIG. 1. Decay schemes of (a) $Fe^{52} \rightarrow Mn^{52} \rightarrow Cr^{52}$ and (b) Mn^{54}. The energies are given in kev, the intensities in percent of disintegration. For the beta transitions the logft values are indicated.

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to measure the lifetime of this level by the observation of delayed coincidences and find an upper limit of 5×10^{-10} sec in contradiction to earlier reports.[6] The 165-kev gamma-ray transition is thus almost certainly predominantly of $M1$ character. This determines the spin of Mn^{52m} as $I=0$, 1, or 2. The decision between the values $I=1$ and $I=2$ may be made from the observation by Osborne and Deutsch[7] of internal electrons due to the isomeric transition, which implies $I=2$, but their experiment has never been repeated. One may also argue that the absence of beta transitions from Mn^{52m} to the ground state of Cr^{52} strongly favors the assignment $I=2$. The results of our nuclear orientation experiment, however, do not allow us to eliminate spin 1 since anisotropy effects of only 4% have been observed (see Sec. V). But we can conclude that the spin of Mn^{52m} is almost certainly 2, with $I=1$ not completely excluded.

The decay scheme of 5.7-day Mn^{52} with all the spin assignments is well established. The Fermi-Gamow-Teller mixture in the beta decay has been measured[8]; the magnetic moment of Mn^{52} is known to be 3.08 nm from paramagnetic resonance experiments.[9] In our experiments we have used Mn^{52} as our "thermometric" reference isotope for the measurements of the magnetic moments of Mn^{54} and Mn^{52m}.

The decay of Mn^{54} takes place through electron capture followed by a single gamma ray. The decay scheme is given in Fig. 1(b). The 840-kev gamma ray is known to be pure $E2$ from angular distribution[1] and linear polarization measurements.[10] The spin and the magnetic moment of Mn^{54} have not been measured previously.

Expressions for the angular distribution $W(\theta)$ and the degree of polarization P of gamma rays emitted from a system of oriented nuclei with rotational symmetry have been given in a previous paper.[3] The anisotropy of the angular distribution is defined by

$$\epsilon=[W(\pi/2)-W(0)]/W(\pi/2),$$

where the angles are measured with respect to the axis of orientation of the spin system.

III. APPARATUS AND EXPERIMENTAL PROCEDURES

In our experiments the radioactive ions were incorporated in the lattice of two types of cooling salts: cerium magnesium nitrate $Ce_2Mg_3(NO_3)_{12}\cdot 24H_2O$ and magnetically diluted nickel fluosilicate (90% Zn, 10% Ni) $\cdot SiF_6\cdot 6H_2O$. Both substances possess an axially symmetric crystal structure and can incorporate divalent ions of the iron group in their lattices. Discussions of the nuclear orientation mechanism of Mn ions in these salts have been presented by several authors.[1,2,4] With our cryogenic facilities, by means of adiabatic demagnetization of these salts, temperatures of about 0.010°K or below were reached, thus giving rise to a large degree of nuclear orientation. The magnetically cooled double-nitrate crystals placed in a magnetic field of several hundred gauss along their trigonal axis yield polarization of Mn nuclei; the cooled fluosilicate crystals in a field-free region produce alignment of the Mn nuclei along the symmetry axis of the crystals due to interaction of the internal electric fields with the Mn ions.

The "thermometric" reference isotope Mn^{52} was produced at the MIT cyclotron by a $(d,2n)$ reaction on a chromium probe; a chemical separation of Mn was made, and none of the other Mn isotopes produced were abundant enough to disturb our measurements. The yield ratio of Mn^{54}/Mn^{52} was found to be less than 0.01. Mn^{54} was obtained from the Nuclear Science and Engineering Corporation, Pittsburgh, Pennsylvania. The isotope was produced by proton bombardment on chromium. Mn^{52m} was derived from Fe^{52} by radioactive decay. The latter was produced at the MIT cyclotron by an $(\alpha,2n)$ reaction on a chromium probe. Fe^{53} and Mn^{52} were produced in abundance in the same bombardment. After Fe^{53} had decayed, the active iron was extracted into nitrobenzene. Repeated back-extractions reduced the contamination of Mn^{52} to less than 0.01%.

The cerium magnesium nitrate and nickel fluosilicate crystals containing Mn^{52} and Mn^{54} were prepared in the conventional manner by growth from radioactive solutions. Since only ferrous ions can be incorporated in these crystals at the proper lattice sites, about 1 mg of the carrier was added to 300 μC of Fe^{52}, and the iron was reduced with sulfur dioxide. The crystals were grown in a desiccator filled with carbon dioxide or under reduced pressure. The latter method is of advantage especially when the crystals had to be prepared within a few hours because of the short-lived radioactivity.

The low-temperature apparatus, the source mounting, the experimental arrangements for the measurement of the angular distribution and of the circular polarization gamma rays emitted by oriented nuclei, and the methods of data accumulation using the multichannel pulse sorter at the L.N.S. Data Center have been described in previous papers.[3,11]

IV. EXPERIMENTAL RESULTS ON Mn^{54}

Experiments were performed with sources containing Mn^{54}, with about 50 μC of activity in 2 to 5 grams of crystals. Polarization in the double nitrates together with an external field of 450 gauss and alignment in the fluosilicates with no polarizing field were applied in separate experiments. The results of the observed angular distribution anisotropy of the 0.84-Mev gamma ray are given in Fig. 2, where the normalized counting

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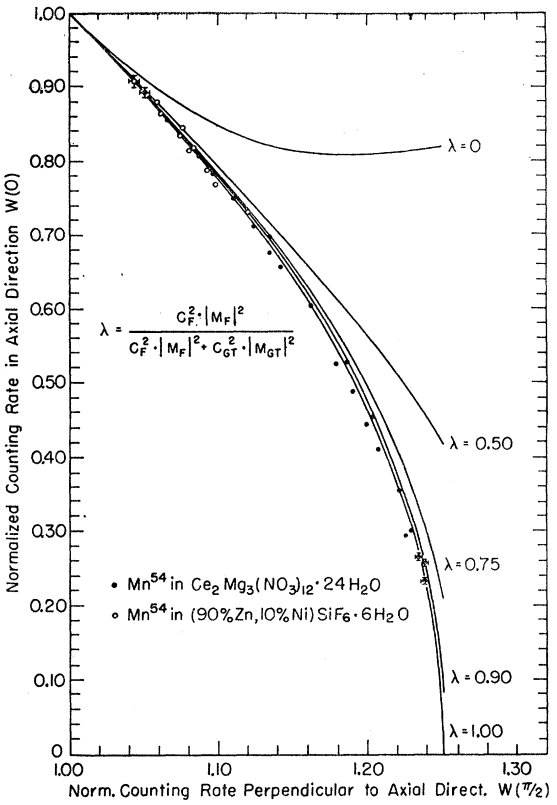

FIG. 2. The counting rate along the axis $W(0)$ is plotted against that in the equatorial plane $W(\pi/2)$ for the 0.84-Mev gamma ray from oriented Mn^{54}. The curves show the dependence expected for different values of the beta-transition mixing parameter λ assuming a $2 \rightarrow 2$ transition. Statistical errors are indicated. The expected dependence for a $3 \rightarrow$ beta transition is identical with that of the $2 \rightarrow 2$ beta transition with $\lambda = 1.00$.

rate in the axial direction $W(0)$ is plotted against that in the plane perpendicular to the axial direction $W(\pi/2)$. The value 1.00 corresponds to an isotropic distribution. The solid curves show the expected dependence for different values of the beta-transition mixing parameter λ assuming a $2 \rightarrow 2$ transition, where $\lambda = 0$ corresponds to a pure Gamow-Teller and $\lambda = 1$ to a pure Fermi transition. The expected dependence for a $3 \rightarrow 2$ beta transition is identical with that of the $2 \rightarrow 2$ beta transition with $\lambda = 1$ (Fermi) where no disorientation in the beta decay occurs. For a $1 \rightarrow 2$ beta transition the expected dependence is a straight line with extreme values of $W(0) = 0.75$ and $W(\pi/2) = 1.12$, thus a maximum anisotropy ϵ of 33%, for complete nuclear orientation.

Table I. Alternate values of the nuclear g values and magnetic moments of Mn^{54}, obtained from the nuclear orientation comparison experiments with 5.7-day Mn^{52}, for the possible spin assignments of Mn^{54} and the various types of beta transitions. The nuclear g value of Mn^{52} is taken to be 0.514.

Mn^{54} spin	Beta decay of Mn^{54}	g_{54}	μ_{54}
3	only G-T possible	0.85±0.07	2.55±0.21
2	F (gives lower limit)	1.03±0.08	2.06±0.16
2	90% F, 10% G-T (upper limit)	1.13±0.08	2.26±0.16
2	not specified	1.08±0.13	2.16±0.26

The degree of orientation of Mn^{54} achieved in the double nitrates was about the same as that reported by the Oxford group.[1] Our measurements also indicate a spin assignment of 3 for Mn^{54}. As seen from Fig. 2, the spin of 2 is not excluded; but in this case the almost complete absence of disorientation in the beta decay would require the Fermi-type transition to be predominant. The possible Gamow-Teller admixture would have to be less than 10%.

Similar sources containing an additional amount of 5.7-day Mn^{52} activity (about 5 μC) were used in the simultaneous measurement of the anisotropies of the 0.84-Mev gamma ray of Mn^{54} and of the 1.43-Mev gamma ray of Mn^{52}. Since both activities were grown into the same crystal, these experiments yield a ratio of the nuclear g values of Mn^{54} and Mn^{52}. The experimental results of these comparison experiments performed with both double nitrate and fluosilicate are given in Fig. 3. The solid curves show the expected dependence for different values of the ratio R of the g value of Mn^{52} to that of Mn^{54}, using the spin assignments given in Fig. 1. With $g = 0.514$ for Mn^{52} (see Sec. II), we get from Fig. 3 a g value of 0.85±0.07 for Mn^{54}, assuming spin 3. Curves similar to the ones in Fig. 3 can be drawn for the case of spin 2 for Mn^{54},

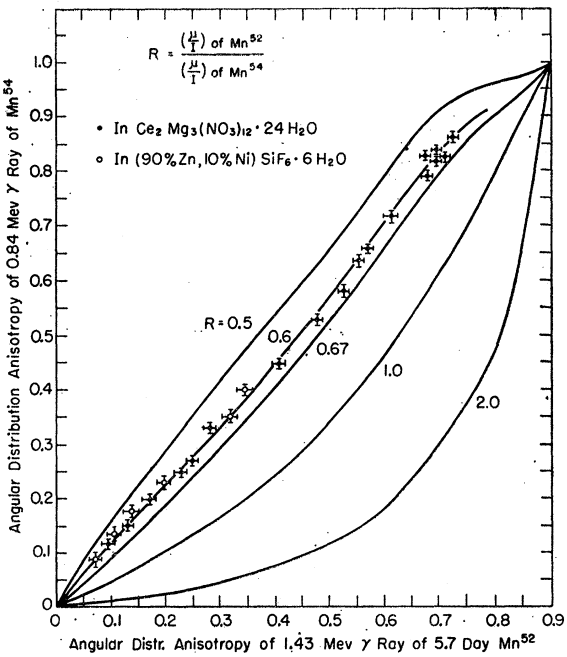

FIG. 3. The simultaneously observed anisotropies of the gamma rays from Mn^{54} and Mn^{52}. The curves show the dependence expected for different values of the ratio R of the nuclear g value of Mn^{52} to that of Mn^{54} using the spin assignments of Fig. 1. Statistical errors are shown.

taking various values of the beta-transition mixing parameter λ. Table I gives the alternate values of the nuclear g values and magnetic moments of Mn^{54}.

The sign of the magnetic moment of Mn^{54} was determined by the measurement of the circular polarization of the 0.84-Mev gamma ray. With sources up to 100 μC of Mn^{54} in cerium magnesium nitrate, the nuclei were polarized using an external field of H_p of about 450 gauss. The gamma polarization was analyzed by forward Compton scattering of the circularly polarized gamma ray with polarized electrons (in magnetically saturated iron). The geometry of the experiment and the efficiency of the polarization analyzer magnet were described in an earlier paper.[3] Figure 4 gives the results of the scattering experiments. The counting rate of the temperature monitor counter $W(90°)$ is given together with the counting rate in the Compton-scattering peak, designated as $W(15°)$. The circular polarization effect E was found to be $+(0.045\pm0.007)$ at the lowest temperatures achieved; thus the counting rates in the polarization detection counter were larger when the polarizing field H_p and the analyzing field H_a are opposite in direction, than in the case of the fields being in the same direction. From the analysis of the angular momenta in the decay of Mn^{54}, the sign of the magnetic

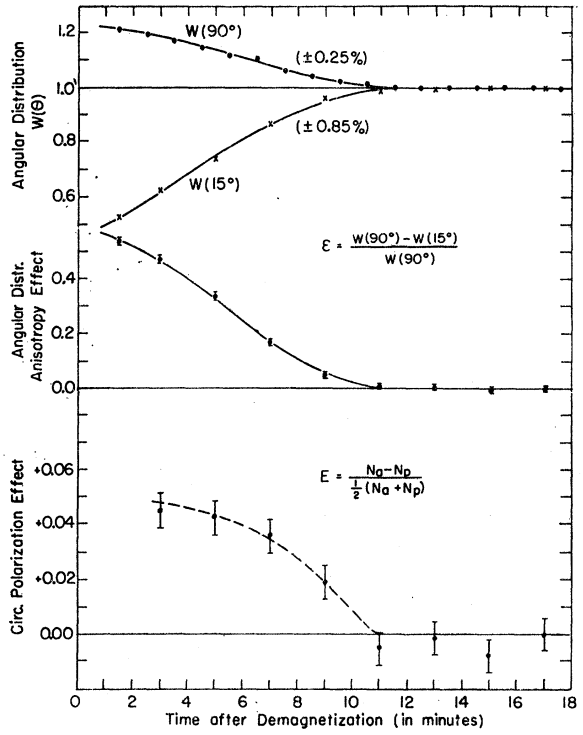

FIG. 4. The angular distribution $W(90°)$ and $W(15°)$, the anisotropy effect, and the polarization effect E, observed on the 0.84-Mev gamma ray emitted from Mn^{54}, as a function of the warm-up time. N_a and N_p are the counting rates in the polarization detection counter when the polarizing and analyzing magnetic fields are in opposite and in the same directions, respectively. Statistical errors are shown.

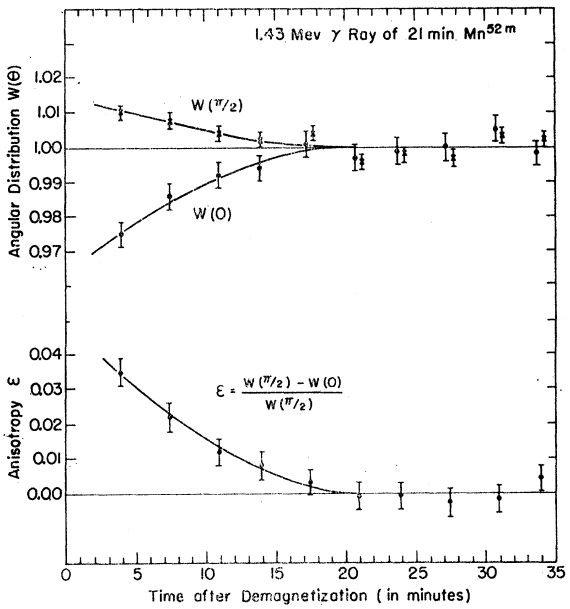

FIG. 5. The observed angular distribution anisotropy of the 1.43-Mev gamma ray of Mn^{52m} aligned in nickel fluosilicate as a function of the warm-up time. The curves are those corresponding to a quadrupole transition. The observed anisotropy is not large enough to determine disorientation effects in the preceding beta decay. Statistical errors are shown.

hyperfine coupling constant of the Mn ions, and the sign of the circular polarization-dependent part of the Compton-scattering cross section, we get a positive magnetic moment for Mn^{54}.

V. EXPERIMENTAL RESULTS ON Mn^{52m}

Experiments were performed with sources containing about 10 μC of Fe^{52} in 2 to 5 grams of crystals. Due to the short half-life of Mn^{52m}, crystals were prepared containing Fe^{52} from which Mn^{52m} is derived in radioactive equilibrium in the crystalline lattice.

We found that no nuclear orientation of Mn^{52} was observed if Fe^{52} was grown into cerium magnesium nitrate crystals. An explanation of this fact can be that divalent iron does not replace magnesium in the crystal lattice, but that iron is bound at interstitial positions. If, however, divalent Fe^{52} was incorporated in the fluosilicates, the Mn^{52m} nuclei, derived from it by beta and gamma decay at lattice sites, were oriented at low temperatures. Figure 5 gives the angular distribution observed for the 1.43-Mev gamma ray of Mn^{52m}. The measured anisotropy was about 4%, which unfortunately was not large enough to allow a determination of the Fermi and Gamow-Teller matrix elements for the beta decay of Mn^{52m} which is believed to be a $2\rightarrow 2$ transition. The observed anisotropy also was not large enough in order to exclude the assignment of spin 1 for Mn^{52m} completely, which was pointed out in Sec. II.

Fluosilicate crystals containing about 5 μC Mn^{52m} and 50 μC Mn^{54} were used in the simultaneous measurement

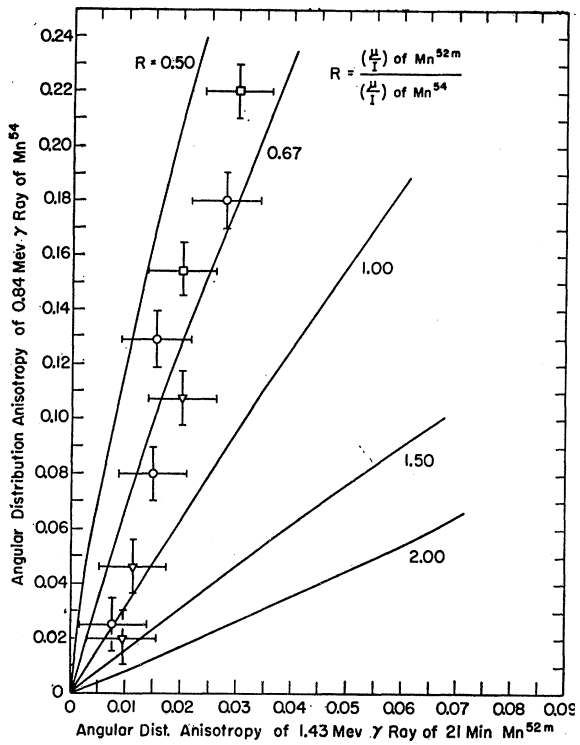

FIG. 6. The simultaneously observed anisotropies of the gamma rays from Mn^{54} and Mn^{52m}. The curves show the dependence expected for different values of the ratio R of the nuclear g value of Mn^{52m} to that of Mn^{54}, using the spin assignment of Fig. 1 and assuming the beta decay of Mn^{52m} to be a pure G-T transition. Statistical errors are shown.

of the anisotropies of the 1.43-Mev gamma ray of Mn^{52m} and of the 0.84-Mev gamma ray of Mn^{54}, which allows a determination of the ratio of the nuclear g values of Mn^{52m} and Mn^{54}, provided the Mn^{52m} atoms resulting from the decay of Fe^{52} are distributed at lattice sites in the same manner as the Mn^{54} atoms incorporated there during the crystal growth. The experimental results of these comparison experiments are given in Fig. 6. The solid curves show the dependence expected for different values of the ratio R of the g value of Mn^{52m} to that of Mn^{54}, using the spin assignments given in Fig. 1 and assuming the beta decay of Mn^{52m} to be a pure Gamow-Teller transition. With $g=0.85\pm0.07$ for Mn^{54} (see Sec. IV), from Fig. 6 we get a g value of 0.52 ± 0.08 for Mn^{52m}, assuming a Gamow-Teller beta transition. Curves similar to the ones in Fig. 6 can be drawn for other beta-decay matrix elements. Table II gives the alternate values of the nuclear g values and magnetic moments of Mn^{52m}.

VI. DISCUSSION AND CONCLUSION

It is shown that a substantial degree of alignment of Mn^{52m}, a nucleus with a half-life of 21 min, can be obtained by cooling a crystal of nickel zinc fluosilicate in which the radioactive parent Fe^{52} has been incorporated and where the isotope under investigation has been produced by radioactive decay. By comparing the gamma-ray anisotropies from Mn^{52m} and Mn^{54}, the ratio of the nuclear g values of the two isotopes is found. The g value of Mn^{54} is determined in terms of that of 5.7-day Mn^{52} which is known from paramagnetic resonance experiments.

The comparison of the Mn^{52m} and Mn^{54} g values by alignment in fluosilicate crystals is independent of temperature but it depends critically on the assumption that the two manganese species are distributed identically in the lattice. This assumption would be invalid either if the parent iron atoms are not incorporated into the crystal in the same manner as is manganese or if the beta decay of Fe^{52} dislocates a significant fraction of the daughter atoms sufficiently to prevent their occupying the appropriate sites even after a mean time of 30 minutes. Ferric iron, which would occupy interstitial positions, is practically not included at all in the crystals as was shown in a separate experiment. A slightly displaced position of the ferrous ion would not disturb our experiment as long as there is no barrier preventing the daughter manganese from finding its equilibrium position. We cannot completely exclude the possibility that a certain fraction of the ions occupy interstitial positions or that the bulk distribution of the two elements in the crystals is slightly different, giving rise to somewhat different average temperatures. On the other hand, general experience would suggest that the behavior of various related ionic species is very similar if they are accommodated by the lattice at all. Ferrous fluosilicate and manganous fluosilicate have been prepared separately and it is found that both have about the same molecular weight indicating that the two salts constitute members of the extensive isomorphous series of fluosilicates. It also seems quite unlikely that a large fraction of the relatively low-energy Fe^{52} beta-decay processes results in dislocating the daughter atom. Recent experiments, utilizing recoilless gamma-ray emission, concerning the interaction of crystalline media with Fe^{57} and Sn^{114} nuclei produced by beta decay *in situ* support this view.

Another phenomenon which may cause the Mn^{52m} nuclei to have a higher average temperature than the Mn^{54} nuclei is connected with the time required to establish thermal equilibrium between the lattice and nuclear spins. Experiments by Grace *et al.*[1] with Mn^{54} and by

TABLE II. Alternate values of the nuclear g values and magnetic moments of Mn^{52m}, obtained from the nuclear orientation comparison experiments with Mn^{54}, for various beta-decay matrix elements in the $2 \rightarrow 2$ transition of Mn^{52m}. A comparison is made with the 5.7-day state of Mn^{52}, whose g value is taken to be 0.514.

Beta decay of Mn^{52m}	g_{52m}	μ_{52m}	g_{52m}/g_{52}
pure G-T	0.52 ± 0.08	1.04 ± 0.16	1.02 ± 0.16
pure F	0.36 ± 0.08	0.72 ± 0.16	0.70 ± 0.16
not specified	0.44 ± 0.16	0.88 ± 0.32	0.86 ± 0.32

Huiskamp *et al.*[2] with Mn^{52} in cerium magnesium nitrate indicate that this time may be several minutes at very low temperatures. The Mn^{52m} nuclei are formed continuously during the experiment, effectively at an infinite temperature since they grow from a parent with $I=0$. Since their mean life at the time of observation is 30 minutes, we can estimate the fraction of Mn^{52m} nuclei which have a temperature much higher than the lattice as $\tau/30$ where τ is the mean time, in minutes, to reach lattice temperature. Thus at most a few percent of the nuclei are affected.

All of the phenomena discussed will tend to reduce the degree of nuclear alignment for Mn^{52m} compared with Mn^{54}, but their effect is probably no larger than the experimental probable errors. A conservative approach would consider our value of g_{52m}/g_{54} as a lower limit only, but it is not likely to be exceeded by much more than the error indicated.

We find the ratio of the g values of the 21-min state to that of the 5.7-day state of Mn^{52} to be of the order of unity. This is to be expected from shell-model arguments[12] if the 21-min state is derived from the same configuration as the ground state, since both the odd proton and the odd neutron in Mn^{52} are in the $f_{7/2}$ shell.

As pointed out in Sec. IV, the spin of Mn^{54} has not been established uniquely. Earlier tabulations (Table IV of the review by Blin-Stoyle[12]) give a spin of 2 and a magnetic moment of about 5 nm. Beta-decay arguments favor spin 3 over spin 2. The decay of $Mn^{54} \rightarrow Cr^{54}$ is closely related to that of $Fe^{55} \rightarrow Mn^{55}$, both taking place by electron capture where an $f_{7/2}$ proton decays to a $p_{3/2}$ neutron and both having a log ft value of about 6. But the Fe^{55} decay is believed to be an allowed beta transition with $\Delta I=1$; this argument carried over to the neighboring Mn^{54} indicates a spin of 3 for this nucleus.

The rather small magnetic moment found by us for Mn^{54} has some interesting implications in the light of a strict j-j coupling model. We note that Mn^{53}, with a closed shell of 28 neutrons, shows the "normal" $\frac{7}{2}^-$ ground state of the $(f_{7/2})^5$ proton configuration, while Mn^{55}, with 30 neutrons, has an anomalous $\frac{5}{2}^-$ ground state. Both isotopes show a gyromagnetic ratio of about 1.4, quite close to the Schmidt limit of 1.65 for the $f_{7/2}$ shell. The configuration of the 29 neutrons in Mn^{54} is certainly $p_{3/2}$, in agreement with the $\frac{3}{2}^-$ ground state of Cr^{53} although the g value of this nucleus is -0.3, much smaller than the Schmidt limit of -1.27.

If we assume that the $(f_{7/2})^5$ configuration for the protons is maintained and coupled separately to the $p_{3/2}$ neutron, we can calculate the g value of Mn^{54} on the assumption of $j_p=\frac{7}{2}$ for the protons. One finds, for $I=3$, $g=g_p$; and for $I=2$, $g=(3/2)g_p-(1/2)g_n$. This coupling scheme therefore predicts too large a moment, whether we choose the Schmidt limit of 1.64 for g_p or the empirical value of 1.4. Thus the protons must contribute a smaller fraction of the angular momentum than $j_p=\frac{7}{2}$, provided j-j coupling holds at all. If the $(f_{7/2})^5$ configuration is coupled to $j_p=\frac{5}{2}$, as in Mn^{55}, we get, for $I=3$, $g=(17/24)g_p+(7/24)g_n$; and for $I=2$, $g=(11/12)g_p+(1/12)g_n$. The latter expression gives a value $1.16<g<1.5$, depending on the values of g_p and g_n chosen, with the Schmidt values and the empirical values from Mn^{55} and Cr^{53} used to obtain the limits. This is substantially greater than our experimental result. For $I=3$, however, we get $0.62<g<1.0$ which is in the right range. In particular, if we choose $g_p=1.4$ as indicated by Mn^{53} and Mn^{55} and $g_n=-0.3$, as in Cr^{53}, we get $g=0.90$ in excellent agreement with experiment. Although this coupling scheme cannot be taken very seriously, especially in view of the small moment of Cr^{53}, the result may help in understanding the moments of other $f_{7/2}$-shell ground states.

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