

Spin and Magnetic Moment of Mn<sup>53†</sup>W. DOBROWOLSKI, R. V. JONES, AND C. D. JEFFRIES  
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The microwave paramagnetic resonance hyperfine structure of Mn<sup>53</sup> has been observed in a powdered sample of MnCl<sub>2</sub> diluted with SrCl<sub>2</sub> at 3 cm wavelength and 20°K. From the number of hfs lines the nuclear spin is found to be  $I(\text{Mn}^{53})=7/2$ ; from their spacing relative to those of Mn<sup>55</sup>, the magnetic moment is found to be  $|\mu(\text{Mn}^{53})|=5.050\pm 0.007$  nuclear magnetons. These results are discussed in relation to the nuclear shell model.

## I. INTRODUCTION

THE method of microwave paramagnetic resonance absorption<sup>1</sup> is sufficiently sensitive and simple to be of considerable utility in the determination of nuclear moments of radioactive nuclei of the transition elements. In addition to its application to several long-lived transuranic nuclei,<sup>2</sup> this technique has recently been used to determine the moments of the radionuclides Co<sup>56</sup>,<sup>3</sup> Co<sup>57</sup>,<sup>4</sup> Co<sup>58</sup>,<sup>5</sup> and Co<sup>60</sup>.<sup>6</sup> This paper describes the extension of the work on radiocobalt to Mn<sup>53</sup>, for which a long-lived activity has been reported.<sup>7</sup>

## II. THEORY

In the Mn<sup>++</sup> ion the five 3*d* electrons just half-fill the shell and the resultant orbital angular momentum is zero. The ground state of the free ion is <sup>6</sup>S, which is split through higher order interactions into a doublet and a quadruplet by crystalline electric fields of cubic symmetry.<sup>8,9</sup> The spectroscopic splitting factor *g* is expected to be isotropic and close to the free spin value *g*=2. In fields of lower symmetry, a dipole-dipole interaction as suggested by Abragam and Pryce<sup>10</sup> may be dominant in producing further and larger splittings. These expectations are borne out by the microwave paramagnetic resonance studies of Bleaney and Ingram,<sup>11</sup> who investigated the spectra of Mn<sup>++</sup> in magnetically dilute Tutton salts and fluorosilicates. An isotropic hyperfine structure due to the 100% abundant stable isotope Mn<sup>55</sup> is also observed, the

relatively wide splitting being explained by Abragam<sup>12,10</sup> as due to a small admixture of a 4*s* electronic state.

A suitable Hamiltonian for a magnetically dilute crystal containing Mn<sup>++</sup> ions in an external magnetic field *H* is<sup>10,11</sup>:

$$\mathcal{H} = g\beta[H_x S_x + H_y S_y + H_z S_z] + F + D[S_z^2 - \frac{1}{3}S(S+1)] \\ + E[S_x^2 - S_y^2] + A'[S_x I_x + S_y I_y + S_z I_z] + N + Q, \quad (1)$$

where *S* is the effective electronic spin and *I* the nuclear spin. At microwave frequencies the first (Zeeman) term is much larger than the others; experimentally it is found that *g*=2.00 and *S*=5/2 for most crystal structures. The term *F* represents the higher order splitting by the cubic component of the crystalline field and is found to be very small ( $\sim 10^{-4}$  cm<sup>-1</sup>). The terms *D* and *E* represent the additional splittings in axial and lower field symmetries. The term *A'* represents the magnetic hyperfine interaction, and for Mn<sup>55</sup> has the typical magnitude  $\sim 10^{-2}$  cm<sup>-1</sup>. The terms *N* and *Q* represent the direct nuclear magnetic dipole and electric quadrupole interactions with the external magnetic field and the crystalline field gradient, respectively, and are negligible. For electronic magnetic dipole transitions this Hamiltonian yields a spectrum of 2*S* fine structure lines, each split into 2*I*+1 hfs lines, as observed experimentally.

In addition to the original work on Tutton salts and fluorosilicates, the spectrum of Mn<sup>++</sup> has been extensively investigated in other single crystals<sup>2,13</sup> in powdered cubic structures,<sup>14,15</sup> and in aqueous solution.<sup>16</sup> In a cubic environment *D*, *E*, and *F* are usually small compared to *A* and the simpler Hamiltonian is suitable for the present purpose of determining nuclear moments in powdered cubic structures:

$$\mathcal{H} = \beta[g(H_x S_x + H_y S_y + H_z S_z) \\ + A(S_x I_x + S_y I_y + S_z I_z)]. \quad (2)$$

The energy levels may be obtained by a perturbation calculation which yields, to terms in second order, the following expression for the magnetic fields *H* for which magnetic dipole transitions ( $\Delta m_s = \pm 1$ ,  $\Delta m = 0$ ;  $S_z = m_s$ ,

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<sup>1</sup> See, for example, B. Bleaney and K. W. H. Stevens, Repts. Progr. in Phys. **16**, 108 (1953).

<sup>2</sup> For a recent review of results see, K. D. Bowers and J. Owen, Repts. Progr. in Phys. **18**, 305 (1955).

<sup>3</sup> Jones, Dobrowolski, and Jeffries, Phys. Rev. **102**, 786 (1956); Baker, Bleaney, Llewellyn, and Shaw, Proc. Phys. Soc. (London) **A69**, 353 (1956).

<sup>4</sup> Baker, Bleaney, Bowers, Shaw, and Trenam, Proc. Phys. Soc. (London) **A66**, 305 (1953).

<sup>5</sup> W. Dobrowolski and C. D. Jeffries (to be published).

<sup>6</sup> Dobrowolski, Jones, and Jeffries, Phys. Rev. **101**, 180 (1956).

<sup>7</sup> J. R. Wilkinson and R. K. Sheline, Phys. Rev. **99**, 752 (1955).

<sup>8</sup> H. Bethe, Ann. Physik **3**, 133 (1929).

<sup>9</sup> J. H. Van Vleck and W. G. Penney, Phil. Mag. **17**, 96 (1934).

<sup>10</sup> A. Abragam and M. H. L. Pryce, Proc. Roy. Soc. (London) **A205**, 135 (1951).

<sup>11</sup> B. Bleaney and D. J. E. Ingram, Proc. Roy. Soc. (London) **A205**, 336 (1951).

<sup>12</sup> A. Abragam, Phys. Rev. **79**, 534 (1950).

<sup>13</sup> W. Low, Phys. Rev. **101**, 1827 (1956).

<sup>14</sup> E. E. Schneider and T. S. England, Physica **17**, 221 (1951).

<sup>15</sup> W. D. Hershberger and H. N. Leifer, Phys. Rev. **88**, 714 (1952).

<sup>16</sup> Tinkham, Weinstein, and Kip, Phys. Rev. **84**, 848 (1951).

$I_z = m$ ) occur for a constant applied microwave frequency  $\nu = g\beta H_0/h$ :

$$H(m) = H_0 - Am - [A^2/2(H_0 - Am)] \cdot [I(I+1) - m^2]. \quad (3)$$

The spectrum thus consists of  $2I+1$  hfs lines approximately uniformly spaced about  $H_0$ , the lines at high fields being slightly more widely spaced because of second order effects.

### III. EXPERIMENTAL PROCEDURES AND RESULTS

With the view in mind of utilizing paramagnetic resonance to determine the nuclear moments of radio-manganese, powdered samples of  $Mn^{55}Cl_2$  diluted 1:1000 with  $SrCl_2$  were prepared.<sup>17</sup>  $SrCl_2$  is cubic and, as expected, the observed spectrum could be fitted to Eq. (3) very well, the maximum discrepancy between calculated and measured resonance field values being about 1 gauss out of 3000. The line half-width at half-maximum was 5 gauss at 20°K. In addition to the  $2I+1$  hfs lines predicted by Eq. (3), there were weak lines in pairs between the hfs lines. These disappeared upon heating the sample and are perhaps due to  $Mn^{++}$  ions in interstitial lattice positions.

$Mn^{53}$  was prepared by bombarding a purified chromium-plated target with 18-Mev deuterons from the 60-in. Crocker Cyclotron, utilizing the reactions  $Cr^{52}(d,n)Mn^{53}$  and  $Cr^{53}(d,2n)Mn^{53}$ ; 300-day  $Mn^{54}$  and 6-day  $Mn^{52}$  were also produced by the reactions  $Cr^{53}(d,n)Mn^{54}$ ,  $Cr^{54}(d,2n)Mn^{54}$ , and  $Cr^{52}(d,n)Mn^{52}$ . The target was cooled a month to allow for  $Mn^{52}$  decay, and then processed by the procedure of Haymond *et al.*<sup>18</sup> The carrier-free  $Mn^{53}$  thus obtained was incorporated as  $Mn^{++}$  in a powdered sample of  $SrCl_2$  and the paramagnetic resonance spectrum observed at a frequency of  $\sim 8900$  Mc/sec at 20°K using the apparatus previously described.<sup>6</sup> Two distinct sets of hfs lines were observed, as shown schematically in Fig. 1. The six strong lines corresponding to a spin  $I=5/2$  are due to  $Mn^{55}$ , a trace of which was present in the chromium target. The eight weaker lines (two are not quite resolved from the stronger  $Mn^{55}$  lines) corresponding to a spin  $I=7/2$  are due to another odd manganese isotope.  $Mn^{49}$ ,  $Mn^{51}$ , and  $Mn^{57}$  are all definitely ruled out because of their short half-lives, leaving  $Mn^{53}$  as the only possibility. These eight lines maintained constant intensity relative to those of  $Mn^{55}$  over the period of observation, which was several weeks, indicating that the half-life is greater than several months. This is in agreement with the result of Wilkinson and Sheline,<sup>7</sup> who find a half-life of 140 years for  $Mn^{53}$ . We thus conclude from our spectrum that the spin is

$$I(Mn^{53}) = 7/2. \quad (4)$$

<sup>17</sup> The utility of  $SrCl_2$  as a cubic host for  $Mn^{++}$  was kindly suggested by Dr. Dieter Gruen.

<sup>18</sup> Haymond, Garrison, and Hamilton, *J. Chem. Phys.* **19**, 382 (1951).

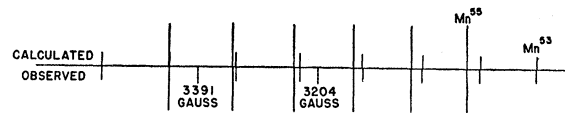


Fig. 1. Observed and calculated paramagnetic resonance hyperfine spectrum of  $Mn^{55}$  (six stronger lines) and  $Mn^{53}$  (eight weaker lines).

In Fig. 1, both sets of hfs lines are compared to calculated values from Eq. (3), taking for  $Mn^{55}$ :  $I=5/2$ ,  $A=93.6$  gauss,  $H_0=3204$  gauss, and for  $Mn^{53}$ :  $I=7/2$ ,  $A=97.4$  gauss, and  $H_0=3204$  gauss. The fact that excellent agreement is obtained for both sets of hfs lines using the same spectroscopic splitting factor  $g$  (contained in  $H_0$ ) indicates that they are indeed both due to the same ion,  $Mn^{++}$ . In a separate experiment we have shown that the hfs lines attributed to  $Mn^{53}$  could not be due to an impurity of  $V^{51}$ , which also has a spin  $7/2$ ; the  $g$  factor and also the hfs coupling constant  $A$  are clearly different.

Since the hyperfine coupling constant  $A$  is proportional to the nuclear gyromagnetic ratio, we see from Eq. (3) that the separation  $\Delta H$  between the two extreme hfs lines ( $m = \pm I$ ) is simply proportional to the nuclear magnetic moment  $\mu$  to a very good approximation. For a series of 17 runs, we thus find for the ratio of the magnetic moments

$$\frac{|\mu(Mn^{53})|}{|\mu(Mn^{55})|} = \frac{\Delta H(Mn^{53})}{\Delta H(Mn^{55})} = 1.455 \pm 0.002, \quad (5)$$

which yields

$$|\mu(Mn^{53})| = 5.050 \pm 0.007 \text{ nm}, \quad (6)$$

including diamagnetic correction. We have used in this calculation the value  $\mu(Mn^{55}) = 3.468$  nm.<sup>19</sup>

In addition to the  $Mn^{53}$  and  $Mn^{55}$  hfs lines described above and shown in Fig. 1, the spectrum also included a line corresponding to  $g=2.00$  very near the field marker  $H_0=3204$  gauss. The intensity of this line increased with time, indicating that it is most likely due to the buildup of paramagnetic resonance centers induced by the radiation damage in the sample by the residual  $Mn^{52}$  and  $Mn^{54}$  activity.

The hfs of  $Mn^{53}$  together with that of  $Mn^{55}$  was also observed in a sample of  $10^{-4}$  molar aqueous solution of  $Mn^{++}$  ions. However, the line width was too large to allow for resolution of the two sets of hfs lines, except for the two extreme  $Mn^{53}$  lines. This experiment yielded a value  $|\mu(Mn^{53})| = 5.04$  nm, in essential agreement with that of Eq. (6); the spin could not be determined with this sample.

### IV. DISCUSSION

From the nuclear shell model viewpoint,  $^{25}Mn_{28}^{53}$  has a neutron configuration  $(f_{7/2})^8$ , i.e., a closed shell, and

<sup>19</sup> W. G. Proctor and F. C. Yu, *Phys. Rev.* **81**, 20 (1951); J. E. Mack, *Revs. Modern Phys.* **22**, 64 (1950).

a proton configuration ( $f_{7/2}$ )<sup>5</sup>. In the extreme single-particle model, a spin  $I=7/2$  and a magnetic moment given by the Schmidt limit  $\mu=+5.79$  nm would be expected. Although we have not determined the sign of the magnetic moment in our experiment, the fairly good agreement in magnitude between the Schmidt limit value and our measured value, Eq. (6), indicates that it is positive. Other nuclei with 25 odd nucleons,  $^{25}\text{Mn}_{30}$ <sup>55</sup> and  $^{22}\text{Ti}_{25}$ <sup>47</sup> and, presumably,  $^{10}\text{Ne}_{11}$ <sup>21</sup>, represent some of the few cases where the single-particle model clearly breaks down, as was pointed out by Mayer.<sup>20</sup> In these cases the assumption of a simple  $jj$  coupling of all the odd particles in the unfilled shell to a resultant spin  $j-1$  gives approximate agreement with the empirical magnetic moment and spin. In the present case it is possible to check this assumption more closely for  $\text{Mn}^{55}$  by noting that identical nucleons in  $jj$  coupling yield a resultant  $g$  factor which is the same as that of the single nucleon.<sup>21</sup> The empirical value of the proton  $g$  factor from our measurements on  $\text{Mn}^{55}$ ,  $g_p=\mu/I=1.443$ , corresponds rather well with the corresponding empirical value  $g_p=1.385$  for  $\text{Mn}^{55}$ .

<sup>20</sup> M. G. Mayer, Phys. Rev. **78**, 16 (1950).

<sup>21</sup> See, for example, A. de-Shalit, Phys. Rev. **90**, 83 (1953).

A long-lived manganese radioactivity attributed to  $\text{Mn}^{53}$  has recently been established by Wilkinson and Sheline,<sup>7</sup> who bombarded enriched  $\text{Cr}^{53}$  with 9.5-Mev protons:  $\text{Cr}^{53}(p,n)\text{Mn}^{53}$ . From the measured activity and an estimated cross section for this reaction, they estimate the half-life to be  $\sim 140$  years. There is good evidence that the decay is by electron capture to the  $p_{3/2}$  ground state of  $\text{Cr}^{53}$  and that there is no gamma radiation. For a disintegration energy of 0.6 Mev obtained from  $\text{Cr}^{53}(p,n)\text{Mn}^{53}$  threshold measurements,<sup>22</sup> this leads to a  $\log ft$  value of about 8.5. On this basis they suggested that the ground state of  $\text{Mn}^{53}$  is  $f_{5/2}$ , as is that of  $\text{Mn}^{55}$ . Our measurements show that the  $\text{Mn}^{53}$  ground state is  $f_{7/2}$ , so that a rather high  $\log ft$  value ( $\sim 12$ ) for the  $f_{7/2} \rightarrow p_{3/2}$  transition would be normally expected. This would seem to indicate that the  $\text{Mn}^{53}$  half-life is longer than 140 years.

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<sup>22</sup> P. H. Stelson and W. M. Preston, Phys. Rev. **86**, 807 (1952).

## Spins of Cesium-127, Cesium-129, and Cesium-130\*

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The spins of three neutron-deficient radioactive isotopes of cesium have been measured by atomic beam methods. The results are: for 6-hour  $\text{Cs}^{127}$ ,  $I=1/2$ ; for 31-hour  $\text{Cs}^{129}$ ,  $I=1/2$ ; for 30-minute  $\text{Cs}^{130}$ ,  $I=1$ .

### EXPERIMENTAL METHOD

THE isotopes  $\text{Cs}^{127}$  (6 hour),<sup>1,2</sup>  $\text{Cs}^{129}$  (31 hour),<sup>1</sup> and  $\text{Cs}^{130}$  (30 minutes)<sup>2,3,4</sup> were produced in the Berkeley 60-inch cyclotron by  $(\alpha, kn)$  reactions on iodine. About 5 mg of  $\text{CsI}$  carrier was added to a solution of the  $\text{BaI}_2$  target material; then the barium was

TABLE I. Counting rates for  $\text{Cs}^{130}$ , integral spin values.

Spin value	0	1	2	3	4
Counting rate (arbitrary units)	$-14 \pm 7$	$100 \pm 13$	$7 \pm 5$	$14 \pm 6$	$9 \pm 3$

\* Research supported jointly by the U. S. Atomic Energy Commission and the Office of Naval Research.

<sup>1</sup> Fink, Reynolds, and Templeton, Phys. Rev. **77**, 614 (1950).

<sup>2</sup> H. B. Mathur and E. K. Hyde, Phys. Rev. **95**, 708 (1954).

<sup>3</sup> J. R. Risser and R. N. Smith, private communication from Lark-Horowitz cited by Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953).

<sup>4</sup> Smith, Mitchell, and Caird, Phys. Rev. **87**, 454 (1952).

eliminated by precipitation of its carbonate, and the excess iodine was sublimed off as  $\text{NH}_4\text{I}$ . The resulting  $\text{CsI}$  was transferred to an atomic-beam oven and reduced by heating with calcium to form a beam of atomic cesium. The beam could be obtained in a little more than an hour after the end of the bombardment.

The  $\text{I}^{127}(\alpha, n)\text{Cs}^{130}$  reaction could be relatively enhanced by using short bombardments and placing aluminum foils in front of the target material to cut the  $\alpha$ -beam energy to a point where the  $\text{I}^{127}(\alpha, 2n)\text{Cs}^{129}$  cross section would be low but the  $\text{Cs}^{130}$  yield would be reasonably high.

TABLE II. Counting rates for  $\text{Cs}^{127}$  and  $\text{Cs}^{129}$ , half-integral spin values.

Spin value	1/2	3/2	5/2	7/2	9/2
Counting rate (arbitrary units)	$100 \pm 1.3$	$1.4 \pm 0.2$	$1.9 \pm 0.2$	$1.8 \pm 0.2$	$1.4 \pm 0.2$