Paramagnetic-Resonance Determination of a Nuclear Quadrupole Interaction by Means of Forbidden Hyperfine Transitions—$CaWO_4:Mn^{2+}$

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The $2I$ forbidden hyperfine doublets $[M(\frac{1}{2}\rightleftarrows-\frac{1}{2}),\ \Delta m=\pm1]$ in the EPR spectrum of Mn^{2+} impurity ions in $CaWO_4$ have been observed. The doublet separations at $\theta=0°$ are in excellent agreement with a perturbation-theoretic calculation which includes fourth order terms. The angular variation of the separations agrees very well with the angular variation calculated by computer diagonalization of the spin Hamiltonian. The doublet separations for general θ have been computed to order $(h\nu)^{-3}$ using perturbation theory and qualitative agreement with the exact angular variation has been obtained. The value obtained for the nuclear quadrupole coupling constant is $Q'=-(0.30\pm0.03)\times10^{-4}$ cm^{-1}.

I. INTRODUCTION

THE electron paramagnetic-resonance spectra of S-state ions usually consist of $(2S)(2I+1)$ so-called allowed ($\Delta M=\pm1$, $\Delta m=0$) lines when either the crystal field is negligible or the external field is parallel to a symmetry axis.[1] Otherwise many forbidden ($\Delta m=\pm1, \pm2\cdots$) lines may also be observable.[2–15]

The fields for resonance for the forbidden transitions $|Mm\rangle\rightleftarrows|M-1, m-1\rangle$ and $|M, m-1\rangle\rightleftarrows|M-1, m\rangle$ differ by first-order nuclear Zeeman and nuclear quadrupole effects, and by higher order effects of the crystal field and hyperfine interactions. As a result, the above pair of transitions gives rise to a so-called forbidden hyperfine doublet labeled (Mm) for each $I\geqslant m>-I$. We are concerned in this paper with the five $(\frac{1}{2},m)$ doublets in the EPR spectrum of Mn^{2+} impurity ions.

We have observed such doublets in the spectrum of Mn^{2+} ions in $CaWO_4$. The doublet separations $\Delta H(m)$ as functions of the angle θ between the external field H and the c axis of the crystal have been measured. From these data and the position of the allowed transitions, we have determined the quadrupole coupling constant and other spin-Hamiltonian constants. Excellent fit to the data was obtained by computer diagonalization of the 36×36 spin Hamiltonian. The value $Q'=-0.30\pm0.03$ (10^{-4} cm^{-1}) was obtained.

II. EXPERIMENTAL PROCEDURE

The EPR experiments were performed at liquid-nitrogen temperatures (80°K) and at X-band frequencies using a spectrometer described previously.[16] Although x-ray orientation was employed, the final means of orientation after the sample was placed in the microwave cavity was to observe the forbidden hyperfine spectrum for which the intensity vanishes at $\theta=0°$ and 90°.

$CaWO_4$ crystals intentionally doped with Mn were not the best samples to use for these experiments because of excessive linewidths and the presence of overlapping fine structure groups. It was found that the Mn spectrum observed in Fe^{3+}-doped crystals was ideally suited for the following reasons:

(a) The low Mn^{2+} concentration yielded linewidths less than 0.5 G for the central $\frac{1}{2}\rightleftarrows-\frac{1}{2}$ fine structure transition.
(b) The $(\pm\frac{1}{2}\rightleftarrows\pm\frac{3}{2})$ and $(\pm\frac{3}{2}\rightleftarrows\pm\frac{5}{2})$ transitions were strain broadened such that they either were not observable or could be easily distinguished from the resonances of interest.
(c) The Fe^{3+}-induced crystal strain does not alter the spin-Hamiltonian parameters of Mn^{2+}.
(d) The isoelectronic Fe^{3+} ion is not observed in the same spectral region as Mn because it has a g' value of 4.3.[16]

Forbidden and allowed hyperfine transitions within the $(\frac{1}{2}\rightleftarrows-\frac{1}{2})$ fine structure group which can be observed under the proper conditions are shown in Fig. 1(a). Derivative EPR spectra observed at several angles for Mn^{2+} impurity ions in $CaWO_4$ are shown in Fig. 1(b). Both $\Delta m=\pm1$ and $\Delta m=\pm2$ forbidden hyperfine transitions are present at large angles between the applied field and the c axis. The broadened $(\pm\frac{1}{2}\rightleftarrows\pm\frac{3}{2})$ resonances are barely visible in the top spectrum ($\theta=90°$) and do not complicate the spectrum at other angles.

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III. DISCUSSION

When the electronic states are mixed by competing Zeeman and crystal-field energies, the direction of the effective field acting on the nuclear moment depends on the electronic state. This direction may diverge considerably from the direction of the external field for even relatively small crystal fields, especially for nominally $M=\frac{1}{2}$ electronic states. As a result, forbidden $\Delta m\neq 0$ transitions can acquire large intensities. Bleaney and Rubins[6] have explained, in this way, the apparently abnormal intensity of Mn^{2+} forbidden hyperfine transitions observed in $ZnSiF_6\cdot 6H_2O$ and $(NH_4)_2Zn(SO_4)_2\cdot 6H_2O$. These authors used "conventional" perturbation theory starting with zero-order wave functions $|\mathrm{Mn}\rangle$ quantized along the external field direction. In their theory, cross-product terms in the crystal-field and hyperfine interactions give rise to large intensities for the forbidden transitions, when H is neither along nor orthogonal to the c axis.

Conventional perturbation theory for the forbidden intensities is known to be quantitatively unsatisfactory except for quite small angles θ between H and the symmetry axis. An improved treatment of this problem has been given by Bir.[17] In this theory, which gives excellent agreement with experiment, the zero-order states are again product states; but the electronic part is nearly an exact eigenfunction of the Zeeman term plus the crystal field, and, together with the hyperfine interaction tensor, it defines the axis of quantization for the nuclear states. The doublet separation to sufficient accuracy for large θ has not yet been computed using this technique, however.

The spin Hamiltonian appropriate to a Mn^{2+} ion in a crystal field with axial symmetry is[18]

$$\begin{aligned}\mathcal{H}=&g_{\parallel}\beta H_z S_z+g_{\perp}\beta(H_x S_x+H_y S_y)+D\left(S_z^2-\frac{35}{12}\right)\\&+\frac{7b_4^0}{12}\left(S_z^4-\frac{95}{14}S_z^2+\frac{81}{16}\right)+\frac{b_4^4}{120}(S_+^4+S_-^4)\\&+AS_zI_z+B(S_xI_x+S_yI_y)\\&-\gamma\beta_N\mathbf{H}\cdot\mathbf{I}+Q'\left(I_z^2-\frac{35}{12}\right).\end{aligned}\quad(1)$$

The forbidden hyperfine-doublet separation,

$$\begin{aligned}\Delta H=&H_{\mathrm{res}}(|\tfrac{1}{2},m\rangle\rightleftarrows|-\tfrac{1}{2},m-1\rangle)\\&-H_{\mathrm{res}}(|\tfrac{1}{2},m-1\rangle\rightleftarrows|-\tfrac{1}{2},m\rangle),\end{aligned}\quad(2)$$

where H_{res} is the field for resonance of the indicated transition, can be computed using conventional perturbation theory.[8] If we neglect b_4^0 and b_4^4 for the moment, the result to second order in $1/H_0$ (found by *third*-order perturbation theory) is

$$\begin{aligned}\Delta H=&\frac{17B_0^2}{H_0}+\frac{4D(A-B)\sin^2 2\theta}{H_0}\\&+\left(\frac{\gamma\beta_N}{g\beta}\right)[2H_0-A(2m-1)]-\frac{25A^3}{2H_0^2}(2m-1)\\&-\left[Q'-\frac{4A^2D}{H_0^2}\right](3\cos^2\theta-1)(2m-1),\end{aligned}\quad(3)$$

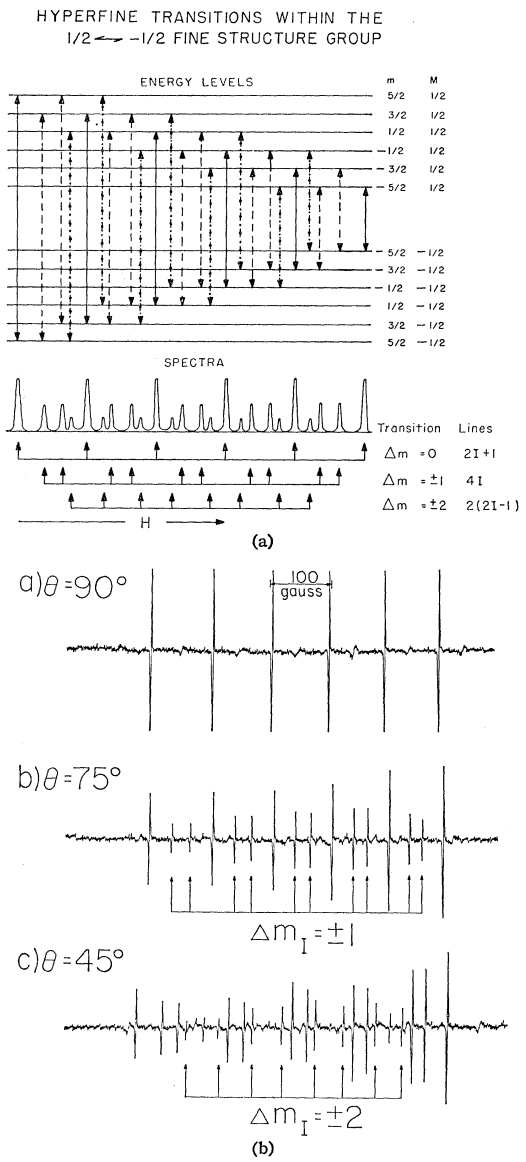

FIG. 1(a). Schematic representation of the number and relative positions of allowed and forbidden hyperfine transitions in the $M(\frac{1}{2}\rightleftarrows-\frac{1}{2})$ fine-structure group for an ion having a nuclear spin $I=\frac{5}{2}$. (b). Derivative EPR spectra of Mn^{2+} impurity ions in $CaWO_4$ obtained at 80°K and X-band frequencies are shown for several angles (θ) between the applied field and the crystalline C axis. The positions of the $\Delta m=\pm 1$ and $\Delta m=\pm 2$ forbidden hyperfine transitions are indicated by the arrows at $\theta=75°$ and $\theta=45°$, respectively.

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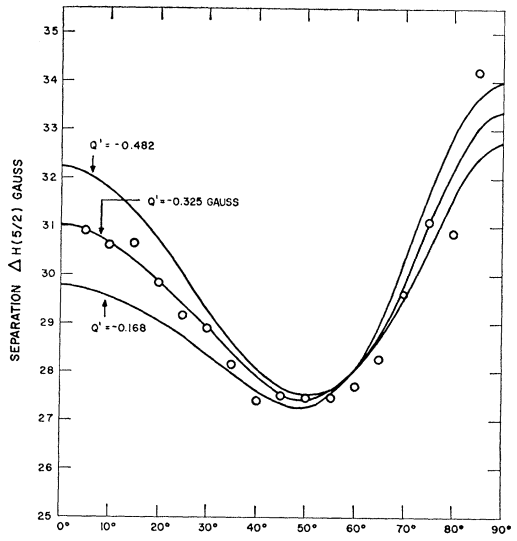
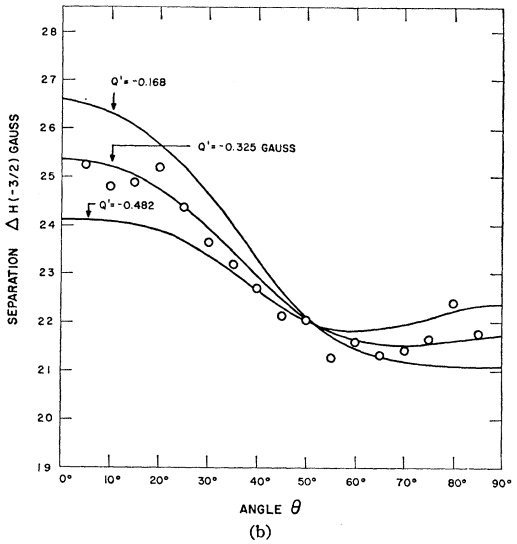


(a)



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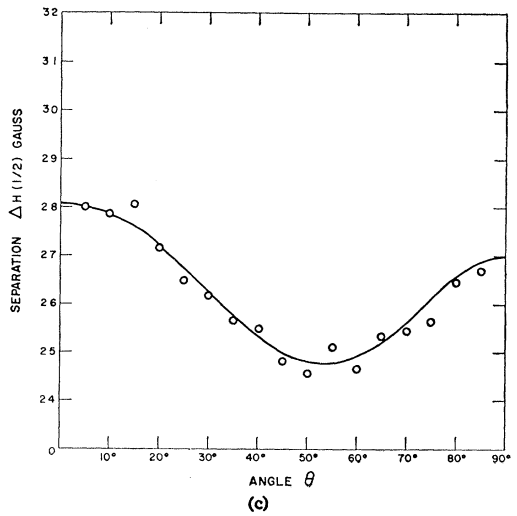

(c)

where

$$B_0=\tfrac{1}{2}(B+B\cos^2\theta+A\sin^2\theta). \tag{3a}$$

Here $H_0=h\nu/g\beta$, and the parameters are in units of gauss. Some very small terms in β_N and Q' have been neglected. We have made the approximation $g_{\parallel}=g_{\perp}=g$, and, for the second-order (in $1/H_0$) corrections, assumed $A=B$.

According to Eq. (3), the doublet separation varies as $(3\cos^2\theta-1)$ for $A\simeq B$ and is independent of angle for the central $(m=\frac{1}{2})$ doublet. Both of these predictions are at variance with the experimental results and the exact theory, as can be seen from Fig. 2.

In order to obtain agreement to within 0.1 G with the exact results obtained by computer diagonalization of Eq. (1) for $\theta=0°$, it is necessary to include the second-order contribution from b_4^0 and also third-order terms in $1/H_0$. The modified ΔH at $\theta=0°$ is then

$$\Delta H=\frac{17B^2}{2H_0}+\left(\frac{\gamma\beta_N}{g\beta}\right)[2H_0-A(2m-1)]-2(2m-1)Q'$$
$$-\frac{(2m-1)A^2}{H_0^2}\left[\frac{25A}{2}-8D+20b_4^0\right]$$
$$-\frac{A^2}{H_0^3}\left\{A^2\left[\frac{2107}{16}+\frac{131}{4}(m-m^2)\right]\right.$$
$$\left.-16D^2-16DA\left[\frac{27}{4}+m-m^2\right]\right\}. \tag{4}$$

The third-order correction is about $\frac{1}{3}$ G and is necessary for agreement to within the experimental error.

We have also computed the corrections to Eq. (3) for general θ, to order $1/H_0^3$. In this calculation, which is outlined in the Appendix, it was necessary to include terms of fifth and sixth order from the perturbation series, as the energy denominators for some nth order terms are of order $H_0^{n/2}A^{(n/2)-1}$ (for even n). The results of this calculation are shown in Fig. 3. We see nearly perfect agreement with the exact calculation for angles near 0° and 90°, but discrepancies of the order of 2 G around 45° remain. The next order would not improve the agreement for the central $m=\frac{1}{2}$ doublet, as this splitting depends only on odd powers of H_0. However, the discrepancy between the perturbation-theory result and the exact calculation is very frequency sensitive, so that a mere 15% increase in the operating frequency

FIG. 2. (a) Angular dependence of the high-field forbidden ($\Delta m=\pm1$) hyperfine doublet separation $\Delta H(\frac{5}{2})$. The open circles are the data points. The continuous curves are obtained from the exact calculations using the three different values of Q' shown on the figure. (b) Same as in (a) except that the open circles are data points obtained from the angular dependence of the low-field forbidden hyperfine doublet separation $\Delta H(-\frac{3}{2})$. The best fit to the data in (a) and (b) is obtained from the exact calculation by choosing $Q'=-0.325$ G. (c) Angular dependence of the center doublet separation $\Delta H(\frac{1}{2})$. The solid curve obtained from the exact calculation is independent of Q'.

would reduce the maximum discrepancy by a factor of 2.

IV. RESULTS

For X-band frequencies, the spin-Hamiltonian parameter cannot be calculated to sufficient accuracy by second-order perturbation theory. The parameters were obtained instead by computer diagonalization of the 36×36 spin Hamiltonian. A least-squares analysis of the fit to 18 allowed transitions yielded the parameters and errors given in Table I. Agreement with the work of Bowers and Hempstead is extremely good.

The greatest differences between second-order theory and exact theory parameters occur for A and B. Second-order theory predicts $A>B$, but the 1% decrease in A and the 1% increase in B obtained from the exact calculation reverses the inequality because of the near isotropy ($A\approx B$). An error of 1% in B would lead to an error of $\frac{1}{2}$ G in the first term of (4) and would preclude a fit to both $\Delta H(\frac{5}{2})$ and $\Delta H(-\frac{3}{2})$ to within experimental error.

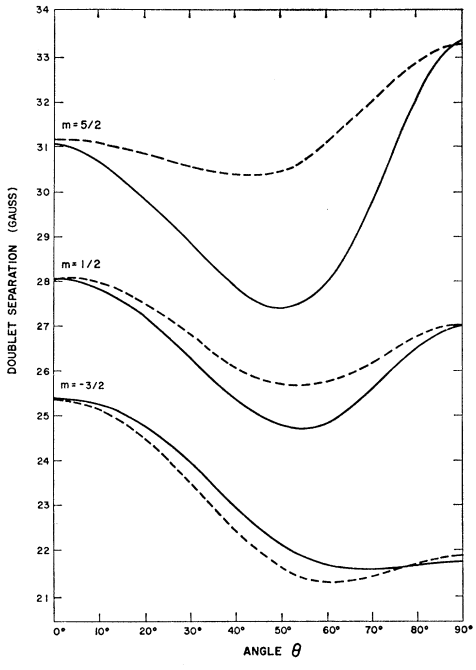

FIG. 3. Comparison between the high field $\Delta H(\frac{5}{2})$, low field $\Delta H(-\frac{3}{2})$, and center $\Delta H(\frac{1}{2})$ angular-dependent doublet separations (solid curves) and those obtained from the perturbation-theory calculation outlined in the Appendix (dashed curves).

The quadrupole constant Q' was determined from the forbidden hyperfine doublet separations. Using the other spin Hamiltonian parameters already determined from the allowed transitions, the value of Q' was adjusted to give best fit to the angularly dependent outer ($m=\frac{5}{2}$, $-\frac{3}{2}$) doublet separations. This is shown in Figs. 2(a) and 2(b). The best value, $Q'=-0.325$ G, can also be obtained, to within 0.01 G, from the perturbation-theory formula (4). Using both outer doublet separations, it is not necessary to consider the odd terms in H_0 (such as the fourth-order term) in Eq. (4), since they are the same for m and $1-m$. Thus,

$$\Delta H(\tfrac{5}{2})-\Delta H(-\tfrac{3}{2}) = -8(\gamma\beta_N/g\beta)A-16Q' - (8A^2/H_0^2)[25A/2-8D+20b_4^0]. \quad (5)$$

The terms even in H_0 change sign so that

$$\Delta H(\tfrac{5}{2})+\Delta H(-\tfrac{3}{2}) = 17B^2/H_0+4H_0(\gamma\beta_N/g\beta) - (A^2/H_0^3)\{17A^2-32D^2-96DA\}. \quad (6)$$

We suggest that the most convenient way to determine Q' accurately from the doublet separations is to use Eq. (5). This determination will be relatively insensitive to the exact values of the crystal-field and hyperfine interactions. Using the exact doublet separation machine-computed from Eqs. (1) and (2), taking $Q'=-0.325$ G and the other parameters from Table I, this method leads to $Q'=-0.32$ G. The numerical values of various terms in (5) are given in Table II, where it is seen that Eq. (5) is accurate to 0.1 G.

We have also checked Eq. (6) and the results are given in Table III. We conclude from Tables I and II that Eq. (4) is accurate to about 0.1 G, under our experimental conditions.

We note from Table III that inclusion of the third-order correction in $1/H_0$ in the work of Schneider and Sircar[8] would have spoiled their previous agreement between theory and experiment. However, in their theory, they assumed that the hyperfine interaction was isotropic, $B=A=-79.3$ G. If we relax this condition

TABLE I. Spin-Hamiltonian parameters for Mn^{2+} in $CaWO_4$.

$g_{\parallel}$	2.00006±0.00005
$g_{\perp}$	2.00079±0.00006
D	$(-137.6\pm0.05)\times10^{-4}$ cm^{-1}
$b_4^0=\frac{9}{2}+\frac{1}{2}F$	$(-1.43\pm0.07)\times10^{-4}$ cm^{-1}
$b_4^4=5a/2$	$(-)(10.43\pm0.06)\times10^{-4}$ cm^{-1}
A	$(-88.93\pm0.06)\times10^{-4}$ cm^{-1}
B	$(-89.60\pm0.06)\times10^{-4}$ cm^{-1}

TABLE II. Contributions to $\Delta H(\frac{5}{2})-\Delta H(-\frac{3}{2})$.

Crystal	$-8\left(\frac{\gamma\beta_N}{g\beta}\right)A$	$-16Q'$	2nd-order terms[a]	$\Delta H(\frac{5}{2})-\Delta H(-\frac{3}{2})$ Eq. (7)	Exact or expt.
$CaWO_4$	0.28	5.20	0.32	5.80	5.70[e]
ZnO[b]	0.0[b]	−2.72[c]	−4.70[b]	−7.42	−7.4[f]
ZnO[d]	0.24	−2.61	−5.04	−7.41	−7.4[f]

[a] See Eq. (5). Terms proportional to $1/H_0^2$.
[b] Follows computation of Ref. 8, in which correction to Zeeman energy (1st term) and contribution to 2nd-order term from b_4^0 is omitted. The contributions nearly cancel.
[c] Using $Q'=0.17$ G of Ref. 8.
[d] Using $Q'=0.163$, with terms omitted under (b) included.
[e] Exact value using quoted parameters and computer calculation.
[f] Experimental value from Ref. 8.

TABLE III. Contributions to $\Delta H(\frac{5}{2})+\Delta H(-\frac{3}{2})$.

Crystal	$\frac{17B^2}{H_0}$	$4H_0\left(\frac{\gamma\beta_N}{g\beta}\right)$	3rd-order terms[a]	$\Delta H(\frac{5}{2})+\Delta H(-\frac{3}{2})$ Eq. (6)	Exact or expt.
$CaWO_4$	51.24	4.61	0.63	56.48	56.40[d]
ZnO[b]	32.20[b]	5.00	0.0[b]	37.26	37.2[e]
ZnO[c]	31.54	5.00	0.66	37.20	37.2[e]

[a] See Eq. (6). Terms proportional to $1/H_0^3$.
[b] Follows computation of Ref. 8, in which $B=-79.3$ G and 4th-order terms are omitted.
[c] Using $B=-78.5$ G and including 3rd-order terms. See text.
[d] Exact value using quoted parameters and computer calculation.
[e] Experimental value from Ref. 8.

and take $B=-78.5$ G, agreement is restored. Note that in the present computation of Q' for ZnO, as given in Table II, we have included some additional terms omitted in Ref. 8. These, however, nearly cancel, leaving Q' almost unchanged.

V. CONCLUSION

The nuclear quadrupole coupling constant for Mn^{2+} in $CaWO_4$ is $Q'=-0.30\pm0.03(10^{-4}\ \mathrm{cm}^{-1})$. To obtain the quoted accuracy from a perturbation-theoretic analysis of the data, it is necessary to include terms of order $1/H_0^3$, where $H_0=h\nu/g\beta$.

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APPENDIX: CALCULATION OF DOUBLET SPLITTING

Using conventional perturbation theory, the energy of the state $|\frac{1}{2},m\rangle$ may be expressed as a series in inverse powers of the applied field H. It is convenient, when performing this calculation, to include all diagonal terms in the zero-order Hamiltonian, as this reduces the number of terms that must be computed. Then the energy denominators may be expanded in powers of $1/H$. The result may be written

$$E(\tfrac{1}{2},m)=\tfrac{1}{2}H-(8/3)D_0+2b_0-\rho Hm+Q_0(m^2-35/12)+\tfrac{1}{2}A_0m+\textstyle\sum_{n=1}R_n(m)H^{-n}, \quad \text{(A1)}$$

where

$$D_0=\tfrac{1}{2}(3\cos^2\theta-1)D, \quad \text{(A2a)}$$

$$Q_0=\tfrac{1}{2}(3\cos^2\theta-1)Q', \quad \text{(A2b)}$$

$$b_0=\tfrac{1}{8}(35\cos^4\theta-30\cos^2\theta+3)b_4^0+\tfrac{1}{8}\cos(4\phi)\sin^4\theta b_4^4, \quad \text{(A2c)}$$

$$A_0=A\cos^2\theta+B\sin^2\theta, \quad \text{(A2d)}$$

$$\rho=\gamma\beta_N/g\beta. \quad \text{(A2e)}$$

All energies are in units of gauss.

We note that the energy of the state $|-\frac{1}{2},m-1\rangle$ may be simply obtained from Eq. (A1) by substituting $1-m$ for m, and changing the sign of H since, in general,

$$E(-M,-m|-H)=E(Mn|H), \quad \text{(A3)}$$

in an obvious notation. If we set

$$E(\tfrac{1}{2},m)-E(-\tfrac{1}{2},m-1)=H_0, \quad \text{(A4)}$$

where $H_0=h\nu/g\beta$, we obtain from Eqs. (A1)–(A4) the resonance energy H_0 as a series in the field for resonance. By a standard iteration technique, starting with $H=H_0$, the series may be inverted, giving the field for resonance as a series in powers of H_0. In the same way, the field for resonance for the transition $|\frac{1}{2},m-1\rangle\rightleftarrows|-\frac{1}{2},m\rangle$ may also be expressed as a series. Subtracting the two series gives the doublet splitting,

$$\Delta H=H_{\mathrm{res}}(|\tfrac{1}{2},m\rangle\rightleftarrows|-\tfrac{1}{2},m-1\rangle)-H_{\mathrm{res}}(|\tfrac{1}{2},m-1\rangle\rightleftarrows|-\tfrac{1}{2},m\rangle)=-2(2m-1)Q_0+\rho[2H_0-A(2m-1)]+\textstyle\sum_{n=1}F_nH_0^{-n}, \quad \text{(A5)}$$

where

$$F_1=G_1, \quad \text{(A6a)}$$

$$F_2=G_2+A(m-\tfrac{1}{2})G_1, \quad \text{(A6b)}$$

$$F_3=G_3+A(2m-1)G_2+A^2(m-\tfrac{1}{2})^2G_1+[R_1(m-1)+R_1(-m)]^2-[R_1(m)+R_1(1-m)]^2, \quad \text{(A6c)}$$

with

$$G_n=R_n(m-1)-R_n(m)+(-)^n[R_n(1-m)-R_n(-m)]. \quad \text{(A7)}$$

Essentially identical calculations have been carried out previously up to terms of order $1/H_0^2$. That the computation of the next order is considerably more complicated may be seen from Eq. (A6c) and, even more importantly, from the fact that certain fifth- and sixth-order terms in the perturbation series as well as lower order terms contribute to F_3. For example, the sixth-order term

$$-\tfrac{1}{16}(D\cos\theta\sin\theta)^2A^4\frac{\langle\frac{1}{2},m|S_-(S_z-\frac{1}{2})\times S_+I_-\times S_-I_+\times S_+I_-\times S_-I_+\times S_+(S_z+\frac{1}{2})|\frac{1}{2},m\rangle}{H_0(A/2)H_0(A/2)H_0} \quad \text{(A8)}$$

contributes to $R_3(m)$, as can be seen from the energy denominators, and hence to G_3 and F_3.

After a tedious calculation, and neglecting terms contributing obviously less than 0.1 G for our experimental conditions, we get

$$R_1(m)=-\tfrac{1}{4}A^2(-35/4+m^2+17m) -8D_1^2+16D_2^2, \quad \text{(A9)}$$

$$G_1=(17/2)B_0^2+16D_1\Delta, \quad \text{(A10a)}$$

$$G_2=A^2(2m-1)[8D_0-(67/4)A-20b_0], \quad \text{(A10b)}$$

$$G_3=\tfrac{1}{4}A^4[-2995/4+399(m^2-m)]+578AD_1^2D_0 +1088AD_1^2D_2-1508A^2D_1^2+192A^2D_2^2 +16A^2D_0^2+16A^3D_0(25/4-3m^2+3m) +384AD_0D_2^2, \quad \text{(A10c)}$$

where

$$D_1=D\sin\theta\cos\theta, \quad \text{(A11a)}$$

$$D_2=\tfrac{1}{4}D\sin^2\theta, \quad \text{(A11b)}$$

$$B_0=\tfrac{1}{2}(A\sin^2\theta+B+B\cos^2\theta), \quad \text{(A11c)}$$

$$\Delta=(A-B)\sin\theta\cos\theta. \quad \text{(A11d)}$$

We have taken $g_\perp=g_\parallel=g$, and neglected the small difference between A and B except in (A10a) where this difference contributes 0.2 G at $\theta=90°$. Substituting Eqs. (A9) and (A10) in (A6c) and neglecting terms in $A-B$ and b_0, we finally obtain the contribution third order in $1/H_0$ to ΔH:

$$F_3=A^4[-2107/16+131/4(m^2-m)] -D_0A^3[-108+16(m^2-m)]-1508A^2D_1^2 +578AD_0D_1^2+1088AD_1^2D_2+192A^2D_2^2 +16A^2D_0^2+384AD_0D_2^2. \quad \text{(A12)}$$

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Ranges of Ba^{126} and Ba^{128} Recoil Fragments in Aluminum*

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Using thin-target, thin-catcher recoil techniques, we have measured the average ranges and range straggling of 97-min Ba^{126} and 2.4-day Ba^{128} in Al. The Ba ions were produced with initial energies from 3 to 14 MeV by bombardment of Sn^{120}, Sb^{121}, and Sb^{123} targets with B^{10}, B^{11}, C^{12}, and N^{14} ion beams. The assumption of a compound-nucleus mechanism in the nuclear reactions leads to a smooth relationship between average range and recoil energy. Theoretical predictions are in good agreement with the observed ranges, but relatively small (<10%) systematic deviations become apparent at the higher energies. The range distributions are consistent with a Gaussian representation, and yield straggling parameters which are substantially larger than theoretical estimates of straggling inherent in the stopping process. Possible sources of the discrepancy are discussed. In one experiment, the product Ba ion is believed to be formed in a reaction involving emission of an alpha particle, and an abnormally large straggling parameter is observed.

I. INTRODUCTION

THE stopping of energetic heavy ions in matter has been of considerable interest in recent years.[1–5] The theoretical description of the energy-loss processes has attained a degree of sophistication which allows a reasonable attempt at predicting stopping properties over a wide region of initial velocities.[1] To determine the accuracy of the theory, it is important that comparisons be made with experimental data for a variety of ions moving in various stopping media. A number of such studies have been reported,[6] and general agreement between theory and experiment has been found for range-energy relationships, with somewhat less satisfactory agreement for range straggling. However, as the measurements have become more refined, systematic deviations have appeared which may require a more critical examination of the theoretical approximations.[4]

In this paper we report measurements of average ranges and range straggling in Al of 2.8 to 14.2-MeV Ba^{126} and Ba^{128} ions (22 to 113 keV/amu). These energies correspond to a velocity region where the contribution of electronic interactions to the stopping process becomes dominating, and the theoretical energy dependences of the stopping properties are quite sensitive to the details of the theory. The energetic Ba ions were

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