

with the experimental value, it is found that  $(6.3 \pm 1.4)\%$  of the magnetization is effectively perpendicular to the direction of the external field. Including this effect as well as the finite solid angles of both counters in the calculation of the intensity ratio of the coincidence lines as a function of  $\delta$ , the solid line in Fig. 1 is obtained. The experimental value of this ratio was determined from a least-squares fit of the sum of all coincidence runs to Lorentzian distributions. The results are  $I_1^e/I_2^e = 0.91 \pm 0.035$  and  $I_6^e/I_6^c = 0.89 \pm 0.035$ . The average of these values, divided by the average intensity ratio of the single count lines and corrected for chance coincidences, is also indicated in Fig. 1, together with its statistical limits of error. From the intersections

with the theoretical curve (solid line), values of  $\delta = -0.15 \pm 0.035$  or  $\delta = -2.58 \pm 0.024$  are found. The first value yields an enhancement factor of about 4 for the  $E2$  part of the 123-keV transition; the second value may be discarded because it would give a much too large  $E2$  transition probability.

The value of  $\delta$  found from the present experiment is in good accordance with a value previously determined by Bishop *et al.*<sup>4</sup> from a study of the directional distribution and the polarization of 123-keV  $\gamma$  rays emitted by oriented  $\text{Co}^{57}$  nuclei.

<sup>4</sup>G. R. Bishop, M. A. Grace, C. E. Johnson, A. C. Knipper, H. R. Lemmer, J. Perez y Jorba, and R. G. Surlock, *Phil. Mag.* **46**, 951 (1955).

## Nuclear Moments and Hyperfine Structure of 13-Year $\text{Eu}^{152*}$

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The magnetic dipole interaction constant  $a$  and the electric quadrupole interaction constant  $b$  for  $\text{Eu}^{152}$  (13 yr) were measured by the method of atomic beams. These values are  $a = \pm 9.345 \pm 0.006$  Mc/sec and  $b = \mp 1.930 \pm 0.165$  Mc/sec. By comparison with the known moment of  $\text{Eu}^{151}$ , the nuclear dipole moment of  $\text{Eu}^{152}$  was found to be  $\mu = \pm 1.912 \pm 0.004$  nm. The sign of this moment cannot be inferred from the experimental results. The zero-field hyperfine separations between levels of different total angular momentum were directly measured.

### INTRODUCTION

IN recent years much work has been done on the isotopes of europium ( $4f^7 5s^2 5p^6 6s^2$ ,  $^8S_{7/2}$ ). Pichanick *et al.* directly determined the magnetic dipole moment of stable  $\text{Eu}^{151}$  in an atomic beam experiment using three rf loops.<sup>1</sup> Sandars and Woodgate, also using the atomic beam method and mass-spectrographic detection, determined the interaction constants for the stable europium isotopes.<sup>2</sup> By use of the results of these experiments, it is possible by means of comparison to determine the nuclear magnetic dipole moment for all the other europium isotopes for which the interaction constants can be measured in the free atom.

Since there are seventeen isotopes of europium with atomic weights in the range 144 to 159, it would seem that the validity of the collective model which is generally taken to hold in the region  $150 < A < 190$  could be checked or modified with knowledge of the nuclear moments of many of the isotopes of europium.

Abraham *et al.*, working with divalent europium ions

bound in crystalline KCl, have performed electron paramagnetic resonance experiments on  $\text{Eu}^{151}$ ,  $\text{Eu}^{152}$ ,  $\text{Eu}^{153}$ , and  $\text{Eu}^{154}$  and measured the hyperfine interaction constants of these species in ionic form.<sup>3</sup> The spin of  $\text{Eu}^{152}$  was found to be  $3\hbar$ . Similarly, Baker and Williams measured the hyperfine interaction in ionic  $\text{Eu}^{151}$  and  $\text{Eu}^{153}$  bound in crystalline  $\text{CaF}_2$  by means of the electron nuclear double resonance (ENDOR) technique.<sup>4</sup> When the results relating to the crystalline ionized Eu isotopes are compared with similar results derived for the atomic state by means of the atomic beam method, significant differences are seen in the magnetic dipole interaction constants. This, when subjected to the theoretical analysis, may furnish useful information about the electronic wave function of atomic and doubly ionized europium.

### THEORY

In the free atom there generally exists an angle-dependent interaction between the nucleus and the surrounding electrons. This interaction can be represented in the nuclear Hamiltonian by a series of terms of which only the first two are ordinarily significant.

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<sup>1</sup>F. M. Pichanick, P. G. H. Sandars, and G. K. Woodgate, *Proc. Roy. Soc. (London)* **A257**, 277 (1960).

<sup>2</sup>P. G. H. Sandars and G. K. Woodgate, *Proc. Roy. Soc. (London)* **A257**, 269 (1960).

<sup>3</sup>M. Abraham, R. Kedzie, and C. D. Jeffries, *Phys. Rev.* **108**, 58, (1957).

<sup>4</sup>J. M. Baker and F. I. B. Williams, *Proc. Roy. Soc. (London)* **A267**, 283 (1962).

The Hamiltonian is written in the form

$$\mathcal{H} = a\mathbf{I} \cdot \mathbf{J} + bQ_{op}, \quad (1)$$

where  $a$  and  $b$  are the magnetic-dipole and electric-quadrupole interaction constants, respectively;  $\mathbf{I}$  is the nuclear spin;  $\mathbf{J}$  is the electronic angular momentum; and  $Q_{op}$  is given by<sup>5</sup>

$$Q_{op} = \frac{3(\mathbf{I} \cdot \mathbf{J})^2 + \frac{3}{2}(\mathbf{I} \cdot \mathbf{J}) - I(I+1)J(J+1)}{2I(2I-1)J(2J-1)}. \quad (2)$$

In the absence of an externally applied magnetic field, the total angular momentum  $\mathbf{F} = \mathbf{I} + \mathbf{J}$  is a constant of the motion. In a representation in which  $F^2$  and  $F_z$  are diagonal matrices, the operators  $\mathbf{I} \cdot \mathbf{J}$  and  $Q_{op}$  are also diagonal, and the solution of Eq. (1) can be written as

$$W_F = C_1(F, I, J)a + C_2(F, I, J)b, \quad (3)$$

where  $C_1(F, I, J)$  and  $C_2(F, I, J)$  are constants depending only upon the  $F$ ,  $I$ , and  $J$  quantum numbers; and  $W_F$  is the energy, usually stated in units of frequency. The total angular momentum  $F$  assumes different integral or half-integral values running from a maximum of  $F = I + J$  to a minimum of  $F = |I - J|$  for any given values of  $I$  and  $J$ .

When an external magnetic field,  $H_0$ , is present, the Hamiltonian (1) becomes

$$\mathcal{H} = a\mathbf{I} \cdot \mathbf{J} + bQ_{op} - g_J(\mu_0/h)\mathbf{J} \cdot \mathbf{H}_0 - g_I(\mu_0/h)\mathbf{I} \cdot \mathbf{H}_0. \quad (4)$$

The symbols  $g_J$  and  $g_I$  are the electronic and nuclear  $g$  factors defined by the relations  $g_J = \mu_J/J$  and  $g_I = \mu_I/I$ , where  $\mu_J$  and  $\mu_I$  are the electronic and nuclear dipole moments in terms of  $\mu_0$ , the Bohr magneton. The electronic  $g$  factor,  $g_J$ , has been measured in stable Eu<sup>151</sup> and Eu<sup>153</sup> and has the value  $g_J = -1.9935 \pm 0.0003$ .<sup>2</sup> For small values of the magnetic field  $H_0$ —i.e., for  $|g_J(\mu_0/h)\mathbf{J} \cdot \mathbf{H}_0| \ll |a\mathbf{I} \cdot \mathbf{J}|$ —the separation in terms of frequency between adjacent magnetic sublevels of a given value of  $F$  can be written as

$$\nu \approx g_F(\mu_0 H_0/h), \quad (5)$$

where  $g_F$  is defined by

$$g_F \approx g_J \frac{F(F+1) + J(J+1) - I(I+1)}{2F(F+1)}. \quad (6)$$

In Eq. (6) a small term proportional to  $g_I$  has been omitted.

During the course of the experiment the transitions labeled  $\alpha$ ,  $\beta$ , and  $\gamma$  in the schematic energy level diagram (Fig. 1) are observed, first at low fields, where their field dependence is given by Eq. (5), and then at higher and higher fields, where this dependence is determined by an exact solution of the Hamiltonian (4) and in particular by the values of  $a$  and  $b$ . A computer program

<sup>5</sup> N. F. Ramsey, *Molecular Beams* (Oxford University Press, New York, 1956), Chap. 9.

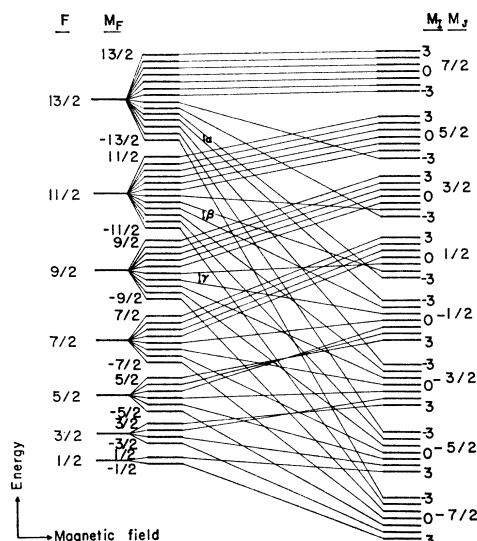


FIG. 1. Schematic energy-level diagram for Eu<sup>152</sup> ( $I=3$ ,  $J=7/2$ ) assuming  $\mu_I$  positive.

is used to solve the Hamiltonian (4) as a function of magnetic field. The input data are the observed transition frequencies, the associated magnetic field, and their uncertainties; the output is the best values of  $a$  and  $b$  obtained by a least-squares fit of Eq. (4) to the data. With these values of  $a$  and  $b$ , a second computer program is used to calculate transition frequencies at higher fields and a search is made for these new resonances. When they are found, the new data are inserted into the first-mentioned program and the process continued until  $a$  and  $b$  are known sufficiently accurately to permit a search to be made for the direct hyperfine transitions ( $\Delta F = \pm 1$ ) at low fields. The fit of the Hamiltonian (4) to the data depends directly upon the choice of the sign of  $g_I$ . First the magnitude of  $g_I$  is estimated by using the known moment and magnetic interaction constant of Eu<sup>152</sup> (as discussed later in this paper). The value of  $g_I$  is first assumed positive and then negative. The data are processed for both choices of sign and the “goodness of fit” is determined by the  $\chi^2$  test of significance.<sup>6</sup> In this way the sign of the nuclear moment can be determined if the precision of observation justifies this. These programs have been described elsewhere.<sup>7,8</sup>

## METHOD

The method used is the atomic beam “flop-in” resonance method first proposed by Zacharias.<sup>9,10</sup> The apparatus is of conventional design utilizing an oven

<sup>6</sup> R. A. Fisher, *Statistical Methods for Research Workers* (Oliver and Boyd, Ltd., Edinburgh, 1946), 10th ed.

<sup>7</sup> H. L. Garvin, T. M. Green, E. Lipworth, and W. A. Nierenberg, *Phys. Rev.* **116**, 393 (1959).

<sup>8</sup> R. Marrus, W. A. Nierenberg, and J. Winocur, *Phys. Rev.* **120**, 1429 (1960).

<sup>9</sup> J. R. Zacharias, *Phys. Rev.* **61**, 270 (1942).

<sup>10</sup> L. Davis, D. E. Nagel, and J. R. Zacharias, *Phys. Rev.* **76**, 1068 (1949).

arrangement particularly convenient for handling materials with high radiation levels. Both the apparatus and oven arrangements have been discussed elsewhere.<sup>11,12</sup>

In this experiment the source material, 13-yr  $\text{Eu}^{152}$ , was produced by irradiation with thermal neutrons. The target material, natural metallic europium, was put into a nuclear reactor operating at a flux of  $9 \times 10^{13}$  neutrons per  $\text{cm}^2\text{-sec}$  for 96 h. As a result of the large thermal-neutron cross section (7200 b) for the reaction  $\text{Eu}^{151}(n,\gamma)\text{Eu}^{152}$  (13 yr), it was possible to produce reasonable specific activities of the 13-yr  $\text{Eu}^{152}$ , of the order of 15.0 mC/mg. Before the irradiated material was used in a run, at least a full week was allowed to elapse so that all the 9.2-h  $\text{Eu}^{152}$ , which is also produced by an  $(n,\gamma)$  reaction, would decay away.

The decay scheme of 13-yr  $\text{Eu}^{152}$  is known and has been summarized by Strominger *et al.*<sup>13</sup> The active isotope decays both with  $K$ -electron capture (approximately 80%) and  $\beta$  decay (approximately 20%). It is known that the former process gives rise to several strong  $\gamma$  rays with energies between 0.9 and 1.5 MeV.<sup>13</sup> For this reason, heavy lead shielding was required, and, as much as possible, loading procedures were carried out remotely.

In the first few attempts at beam production, the sample was introduced into a sharp-lipped tantalum crucible which was then put into a tantalum oven. The whole assembly was heated slowly by electron bombardment. At temperatures of about 1200°K there was a marked burst of activity, after which little activity remained in the oven. This behavior is thought to be due to a thin film of high-melting  $\text{Eu}_2\text{O}_3$ , which ultimately breaks and allows the volatile europium metal to escape quickly. This problem was surmounted by introducing the active sample into a carbon crucible half filled with fine carbon powder. The oven was heated slowly. At temperatures on the order of 2000°K, a stable beam was produced. It is thought that the carbide of europium is formed at low temperatures and then is dissociated at the higher operational temperature. Beam stability was adequate; the intensity fell off uniformly at a rate of about a factor of 2 every hour.

Beam collection was tested on cold, clean surfaces of sulfur, silver, and freshly flamed platinum. All these materials showed comparable collection efficiencies. Platinum foils were used throughout the experiment for collection purposes. Counting was done in small-volume methane counters.

The beam intensity was measured after each resonance exposure for purposes of normalization. This was done by taking a short exposure with all beam barriers—i.e., stop wires—removed but with the magnetic fields

still on. It was noted by this method that the beam consisted almost entirely of atoms.

The magnetic field was determined from observations of the resonant frequency of potassium-39 between the levels  $F=2, M_F=-1$ , and  $F=2, M_F=-2$ , where  $F$  is the total angular momentum quantum number and  $M_F$  designates the projection of the total angular momentum vector along the direction of quantization, i.e., the magnetic field direction. The potassium-39 beam was detected by surface ionization from a hot platinum wire.

## RESULTS

A total of eleven resolved resonances was observed, representing eight different types of transitions. The results are displayed in Table I. Under the heading “transition type” in Table I there appear the subheadings  $F_1, M_1$  and  $F_2, M_2$ , which indicate the levels between which the observed transition occurs. The last column in Table I gives the difference between the observed transition frequency and the frequency calculated from the diagonalization of the Hamiltonian (4) by using the values of  $a$  and  $b$  resulting from the best fit of the data. The uncertainty in the magnetic field is estimated from the width of the calibrating isotope resonance. We have taken this uncertainty to be  $\frac{1}{3}$  the  $\text{K}^{39}$  resonance linewidth. The uncertainty in the  $\text{Eu}^{152}$  resonances is taken as  $\frac{1}{2}$  of their linewidth.

The eleven observed resonances listed in Table I were used as input data along with the accurately known value of  $g_I$  for the least-squares fit program. First  $g_I$  was assumed positive and convergence was obtained. The assumption was then made that  $g_I$  was negative and the process was repeated. The results are shown in Table II. The last column of Table II shows the appropriate value of  $\chi^2$ , the “goodness of fit” parameter, which is defined as

$$\chi^2 = \sum_i (f_i^{\text{obs}} - f_i^{\text{calc}})^2 / (\Delta\nu_i)^2, \quad (7)$$

where  $(f_i^{\text{obs}} - f_i^{\text{calc}})$  is the difference between the observed and calculated frequencies for the  $i$ th resonance and  $\Delta\nu_i$  is the combined error consisting of contributions from both the uncertainty in the calibrating resonance and that in the  $\text{Eu}^{152}$  resonance. It is readily seen from Table II that the assumption of either positive or negative values of  $g_I$  does not affect the resulting values of  $a$  and  $b$ . It is also seen that there is no significant difference in the  $\chi^2$ 's resulting from either sign assignment; that is, the data are equally well fitted under the assumption of either positive or negative  $g_I$ . Because there is no significant difference between the values of  $\chi^2$  for the assumption of both  $g_I > 0$  and  $g_I < 0$ , no statement concerning the sign of  $g_I$  is warranted.

Positive identification is assured in several ways. Bombarding natural europium with neutrons gives rise to isotopes of europium other than  $\text{Eu}^{152}$ . Simple analysis shows that the only other isotope that can

<sup>11</sup> G. O. Brink, thesis, University of California Radiation Laboratory Report UCRL-3642, 1957 (unpublished).

<sup>12</sup> J. C. Hubbs, R. Marrus, W. A. Nierenberg, and J. L. Worcester, *Phys. Rev.* **109**, 390 (1958).

<sup>13</sup> D. Strominger, J. M. Hollander, and G. T. Seaborg, *Rev. Mod. Phys.* **30**, 585 (1958).

TABLE I. Observed resolved resonances in Eu<sup>152</sup>;  $I=3, J=7/2$ .<sup>a</sup>

$F_1$	Transition type		$M_2$	Potassium frequency and uncertainty (Mc/sec)		Magnetic field and uncertainty (G)		Observed resonance frequency and uncertainty (Mc/sec)		$f_{\text{obs}}^{\text{less}}$ (Mc/sec)
	$M_1$	$F_2$								
13/2	-5/2	11/2	-5/2	0.704	0.020	1.000	0.028	59.950	0.075	+0.006
11/2	-3/2	9/2	-3/2	0.704	0.020	1.000	0.028	51.325	0.035	-0.002
9/2	-1/2	7/2	-1/2	0.704	0.026	1.000	0.037	42.350	0.063	+0.005
7/2	5/2	5/2	3/2	7.334	0.027	10.001	0.035	49.400	0.150	+0.051
7/2	3/2	5/2	5/2	7.334	0.027	10.001	0.035	48.350	0.175	-0.107
	$\alpha$			6.000	0.028	8.248	0.037	13.570	0.050	+0.035
	$\alpha$			12.065	0.017	16.001	0.021	28.485	0.240	-0.079
	$\alpha$			20.542	0.050	26.001	0.056	51.360	0.125	+0.074
	$\alpha$			35.777	0.037	42.007	0.036	92.430	0.150	+0.003
	$\beta$			6.000	0.028	8.248	0.037	14.400	0.130	+0.008
	$\gamma$			26.006	0.059	32.004	0.063	83.875	0.163	+0.139

<sup>a</sup> The symbols  $\alpha$ ,  $\beta$ , and  $\gamma$  denote the transitions of the type:  $\alpha$ , ( $F=13/2, M_F=-5/2 \leftrightarrow F=13/2, M_F=-7/2$ );  $\beta$ , ( $F=11/2, M_F=-3/2 \leftrightarrow F=11/2, M_F=-5/2$ );  $\gamma$ , ( $F=9/2, M_F=-1/2 \leftrightarrow F=9/2, M_F=-3/2$ ).

possibly be confused with Eu<sup>152</sup> is Eu<sup>154</sup> and that this isotope is produced in small amounts. The ratio of produced Eu<sup>152</sup> to Eu<sup>154</sup> is 21:1. Since the background level is usually about  $\frac{1}{10}$  of a resonance maximum, any effects due to Eu<sup>154</sup> are small compared with the background. Comparison of the magnetic dipole interaction constants for Eu<sup>152</sup> as determined in this experiment with the value determined by Sandars and Woodgate<sup>2</sup> for Eu<sup>151</sup> gives the same results as found in a paramagnetic resonance experiment by Abraham *et al.*<sup>3</sup> This is discussed in a later section. Our identification is consistent with the results found by these other researchers. Lastly, use of a RCL 256-channel analyzer showed nine definite peaks in the  $\gamma$ -ray spectrum of a source sample, all of which agreed within 1% with the known  $\gamma$ -ray energies of Eu<sup>152</sup> as listed by Strominger *et al.*<sup>13</sup> No peaks were observed that could not be identified as a definite member of the Eu<sup>152</sup> spectrum. All these means of identification give unambiguous evidence that Eu<sup>152</sup> was the isotope studied in this experiment.

MAGNETIC DIPOLE MOMENT

The magnetic moment of stable Eu<sup>151</sup> was measured directly by Pichanick *et al.*<sup>1</sup> by means of an atomic-beam method utilizing three rf loops. The diamagnetically corrected value that these researchers found for the moment of Eu<sup>151</sup> was  $\mu_{151} = 3.419(4)$  nm. The nuclear magnetic dipole moment of Eu<sup>152</sup> is related to that of Eu<sup>151</sup> by the relation

$$\mu^{152}/\mu^{151} = (a^{152}/a^{151})(I^{152}/I^{151}), \tag{8}$$

where the superscripts indicate to which nuclear species the symbol refers and the symbols themselves have already been defined. Absolute values are taken in the application of Eq. (8) because of the inherent difficulty of the atomic-beam method in determining the absolute sign of the interaction constants. The value of  $a^{151}$ , the magnetic dipole interaction constant for Eu<sup>151</sup>, has been determined by Sandars and Woodgate<sup>2</sup> as

$a^{151} = -20.0523(2)$  Mc/sec. Using the appropriate values in Eq. (8), we determine

$$(\mu_I^{152})_{\text{corr}} = \pm 1.912 \pm 0.004 \text{ nm.} \tag{9}$$

Since comparison is made to a diamagnetically corrected moment, the value (9) can be considered as diamagnetically corrected. The diamagnetically uncorrected value is  $(\mu_I^{152})_{\text{uncorr}} = \pm 1.899 \pm 0.004$  nm.

It is known that the individual-particle model is invalid in the region  $150 < A < 190$ , where large nuclear deformations are known to occur. It is in this region that the collective model has its greatest utilization.<sup>14,15</sup> In the case of Eu<sup>152</sup>, where  $Z=63$  and  $N=89$ , we are dealing with an odd-odd nucleus, subject to the coupling rules proposed by Gallagher and Moszkowski.<sup>16</sup> These rules state that for strongly deformed nuclei described by the asymptotic quantum numbers  $N, n_z, \Lambda$ , and  $\Sigma$  stated in the order  $(N, n_z, \Lambda, \Sigma)$ , where  $N$  is the total harmonic oscillator quantum number,  $n_z$  is the number of oscillator quanta along a spatial axis,  $\Lambda$  is the projected orbital angular momentum of the odd nucleon along the axis of nuclear symmetry, and  $\Sigma$  is the projected spin angular momentum of the odd nucleon along the axis of nuclear symmetry, the following relations

TABLE II. Results of the computer program using  $g_J = -1.9935(3), I=3$ , and  $J=7/2$ .

Assumption on sign of $g_I$	Magnetic dipole interaction constant (Mc/sec)	Electric quadrupole interaction constant (Mc/sec)	$\chi^2$
$g_I > 0$	$\pm 9.345 \pm 0.006$	$\mp 1.930 \pm 0.165$	1.29
$g_I < 0$	$\mp 9.345 \pm 0.006$	$\pm 1.930 \pm 0.165$	1.14

<sup>14</sup> S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. **29**, No. 16 (1955).

<sup>15</sup> B. R. Mottelson and S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Skrifter **1**, No. 8 (1959).

<sup>16</sup> C. J. Gallagher, Jr., and S. A. Moszkowski, Phys. Rev. **111**, 1282 (1958).

hold:

$$\begin{aligned} I &= \Omega_p + \Omega_n \quad \text{for } \Omega_p = \Lambda_p \pm \frac{1}{2} \text{ and } \Omega_n = \Lambda_n \pm \frac{1}{2}, \\ I &= |\Omega_p - \Omega_n| \quad \text{for } \Omega_p = \Lambda_p \pm \frac{1}{2} \text{ and } \Omega_n = \Lambda_n \mp \frac{1}{2}. \end{aligned} \quad (10)$$

Here  $\Omega$  equals  $\Lambda + \Sigma$ , and is the total angular momentum of an odd nucleon along the axis of nuclear symmetry; the subscript  $p$  or  $n$  refers to the odd proton or neutron, respectively. Using the collective model,<sup>14,15</sup> Gallagher and Moszkowski<sup>16</sup> have assumed a configuration of [411+] for the proton part and either [521+] or [651+] for the neutron part. This configuration assignment is consistent with the first of the two rules stated above, i.e.,  $I = \Lambda_p + \Sigma_p + \Lambda_n + \Sigma_n = 1 + \frac{1}{2} + 1 + \frac{1}{2} = 3$ , which was experimentally observed. Gallagher and Moszkowski further state the relation derived from the collective model

$$\mu = (I/I+1)[\pm(\Lambda_p + 5.6\Sigma_p) \mp 3.8\Sigma_n + Z/A], \quad (11)$$

where the signs of the two terms of the expression are the same as the signs of  $\Omega_p$  and  $\Omega_n$  appearing in the coupling rules (10). By use of expression (11), which makes use of the Schmidt values for the gyromagnetic ratios of the odd nuclei (i.e., no quenching), the value for the moment is derived as

$$(\mu_I^{152})_{\text{calc}} = +1.73 \text{ nm}. \quad (12)$$

This value compares favorably in magnitude to the experimentally observed value of  $(\mu_I^{152})_{\text{exp}} = \pm 1.912(4)$  nm. This seems to imply that the asymptotic quantum-number nuclear configuration has been correctly assumed, and gives further support to the collective model in this region.

#### ELECTRIC QUADRUPOLE MOMENT

The electric quadrupole interaction constant,  $b$ , is related to the quadrupole moment,  $Q$ , by the expression

$$hb = -e^2 Q \langle 1/r^3 \rangle \langle LSJJ | 3 \cos^2\theta - 1 | LSJJ \rangle. \quad (13)$$

This cannot be evaluated directly because the ground-state electronic wave function is not known for europium. It is known that there is a definite departure from pure Russell-Saunders coupling, which predicts a value of  $g_J = -2.0023$  and also the absence of any hyperfine interaction for the Hund's-rule ground level of  $^8S_{7/2}$ . Judd and Lindgren have shown that the experimental value of  $g_J = -1.9935 \pm 0.003^{(2)}$  is in agreement with the simple Landé formula if corrections are made for the departure from the pure Russell-Saunders coupling and for relativistic and diamagnetic effects.<sup>17</sup> As yet, there are no adequate theoretical calculations to explain quantitatively the existence of the hyperfine interaction in the europium isotopes.

Although the quadrupole moment cannot currently be calculated, it is known that for the same electronic wave function—i.e., the same chemical element—the

following relation holds for various isotopes

$$Q^{(1)}/Q^{(2)} = b^{(1)}/b^{(2)}, \quad (14)$$

where the superscripts are used to indicate different nuclei. In using Eq. (14) absolute values are taken, for the reason indicated previously.

Using Eq. (14) and the results of Sandars and Woodgate,<sup>2</sup> we have

$$\begin{aligned} |Q^{152}/Q^{151}| &= 2.75 \pm 0.24 \\ \text{and} \\ |Q^{152}/Q^{153}| &= 1.08 \pm 0.09. \end{aligned} \quad (15)$$

Although the atomic-beam method is ill suited for the absolute determination of the signs of the interaction constants, the relative signs of the interaction constants can readily be determined; hence, we display our results with those of Sandars and Woodgate<sup>2</sup>:

$$\begin{aligned} \text{Eu}^{151}: \quad b/a &= +0.03497(18); \\ \text{Eu}^{152}: \quad b/a &= -0.207(18); \\ \text{Eu}^{153}: \quad b/a &= +0.2016(4). \end{aligned} \quad (16)$$

#### HYPERFINE STRUCTURE

Solution of the Hamiltonian (1) gives the zero-field separation in energy levels characterized by different  $F$  values. These values are

$$\begin{aligned} \Delta\nu_{13/2,11/2} &= 59.848 \pm 0.086 \text{ Mc/sec}, \\ \Delta\nu_{11/2,9/2} &= 51.246 \pm 0.035 \text{ Mc/sec}, \\ \Delta\nu_{9/2,7/2} &= 42.343 \pm 0.037 \text{ Mc/sec}, \\ \Delta\nu_{7/2,5/2} &= 33.191 \pm 0.048 \text{ Mc/sec}, \end{aligned} \quad (17)$$

where  $\Delta\nu_{13/2,11/2}$  is the zero-field separation between the  $F=13/2$  and  $F=11/2$  levels, and similarly for the other separations. The relative ordering of the  $F$  levels was found to be normal although no statement can be made as to whether  $F=13/2$  or  $F=11/2$  lies highest in the energy-level diagram.

#### DISCUSSION

The ground state of both the europium atom ( $4f^7 5s^2 5p^6 6s^2$ ) and the divalent europium ion ( $4f^7 5s^2 5p^6$ ) is  $^8S_{7/2}$ . Since this is a spherically symmetric state, no hyperfine structure should be evident. The presence of hyperfine effects probably results from admixture of the other levels of the  $f^7$  configuration.

An interesting feature is revealed by comparison of the measured values of the magnetic dipole interaction constant determined by the atomic-beam technique on the one hand and by the paramagnetic resonance and ENDOR techniques on the other hand. By means of the atomic-beam method it is possible to measure the electron-nuclear interaction in the free atom, whereas the paramagnetic resonance and ENDOR techniques are used to measure the electron-nuclear interaction of the  $\text{Eu}^{++}$  ion bound in a suitable crystal. Abraham, Kedzie, and Jeffries measured the spin of  $\text{Eu}^{152}$  and

<sup>17</sup> B. R. Judd and I. Lindgren, Phys. Rev. **122**, 1802 (1961).

Eu<sup>154</sup> in a paramagnetic resonance experiment and also the magnetic dipole interaction constants of Eu<sup>151</sup>, Eu<sup>152</sup>, and Eu<sup>153</sup> in the doubly ionized form bound in crystalline KCl.<sup>3</sup> Baker and Williams, employing the ENDOR technique, measured the hyperfine interaction constants for doubly ionized Eu<sup>151</sup> and Eu<sup>153</sup> bound in crystalline CaF<sub>2</sub>.<sup>4</sup> The results of these researchers are indicated in Table III along with our results. It is seen that the value of the magnetic dipole interaction constant for the KCl-crystalline bound Eu<sup>++</sup> is 4.87 times that for the free atom. The value for the CaF<sub>2</sub>-crystalline bound Eu<sup>++</sup> is about 5.14 times that for the free atom. The difference in these two ratios, which amounts to about 5%, is presumably directly connected to the structural differences between the KCl and CaF<sub>2</sub> crystals.

The magnetic dipole interaction constant is defined as

$$ha = -\langle II | \mathbf{y} | II \rangle \langle JJ | \mathbf{H} | JJ \rangle / IJ, \quad (18)$$

where the first set of brackets indicates the expectation value of the magnetic moment operator  $\mathbf{y}$  for the nuclear states with  $M_I = I$ , and the second set of brackets indicates the expectation value of the magnetic field operator,  $\mathbf{H}$ , for electronic states with  $M_J = J$ . The magnetic field operator,  $\mathbf{H}$ , is defined as

$$\mathbf{H} = -2\mu_0 \left\{ \sum_k \left[ \frac{\mathbf{l}_k - \mathbf{s}_k}{r_k^3} + \frac{3(\mathbf{r}_k \cdot \mathbf{s}_k)\mathbf{r}_k}{r_k^5} \right] + \frac{8\pi\delta(\mathbf{r}_k)}{3} \mathbf{s}_k \right\}, \quad (19)$$

where  $\mu_0$  is the Bohr magneton. The subscript  $k$  refers to the  $k$ th electron of the system;  $\mathbf{l}_k$ ,  $\mathbf{s}_k$ , and  $\mathbf{r}_k$  denote the orbital angular momentum, spin angular momentum, and position of the  $k$ th electron, respectively. The term appearing in the square brackets in Eq. (19) corresponds to classical dipole-dipole interaction. The second term, first hypothesized by Fermi,<sup>18</sup> denotes the contact interaction of the  $s$  electrons with the nuclear spin.

Since the value of the magnetic dipole interaction constant for the Eu<sup>++</sup> ion in both the KCl and CaF<sub>2</sub> crystals is about five times that of the free atom, and since the expectation value for the nuclear dipole moment must be the same in both the crystalline-bound ion and the free atom, as also must the values of  $I$  and  $J$ , we conclude from Eq. (18) that the expectation value of the magnetic field at the nucleus is correspondingly about five times as large for the Eu<sup>++</sup> ion in the crystal as in the free atom. The theoretical explanation for the large difference in the expectation value of the operator  $\mathbf{H}$  of Eq. (19) is not readily apparent. Neglecting small effects from the crystalline field, one might at first assume that the removal of two  $6s$  electrons in

TABLE III. Values of the magnetic dipole interaction constant.

Isotope	Value from paramagnetic resonance in KCl, $ a_{PR} $ (Mc/sec)	Value from atomic beams, $ a_{AB} $ (Mc/sec)	Ratio of paramagnetic resonance value to atomic beam value, $ a_{PR}/a_{AB} $
Eu <sup>151</sup>	97.61 (18) <sup>a</sup>	20.0523 (2) <sup>b</sup>	4.868 (9)
Eu <sup>152</sup>	45.33 (45) <sup>a</sup>	9.345 (6) <sup>c</sup>	4.851 (49)
Eu <sup>153</sup>	43.11 (9) <sup>a</sup>	8.8532 (2) <sup>b</sup>	4.869 (9)

Isotope	Value from ENDOR in CaF <sub>2</sub> , $ a_{ENDOR} $ (Mc/sec)	Value from atomic beams, $ a_{AB} $ (Mc/sec)	Ratio of ENDOR value to atomic beam value, $ a_{ENDOR}/a_{AB} $
Eu <sup>151</sup>	102.9069 (13) <sup>d</sup>	20.0523 (2) <sup>b</sup>	5.13193 (8)
Eu <sup>153</sup>	45.6730 (25) <sup>d</sup>	8.8532 (2) <sup>b</sup>	5.15893 (30)

<sup>a</sup> See reference 3.

<sup>b</sup> See reference 2.

<sup>c</sup> (this paper).

<sup>d</sup> See reference 4.

the divalent ion would have little, if any, effect on the magnetic field at the nucleus, since the total electron spin density of these two electrons taken together is zero, and hence the Fermi or contact term in Eq. (19) would make no contribution to the field. Work of Heine<sup>19</sup> has indicated, however, that there is an  $s$ -electron effect even when there are no unpaired  $s$  electrons. His explanation for this is based on electron exchange between the  $s$  electrons and electrons from other subshells, resulting in a net polarization of the  $s$  electron, and thus making possible a contribution from the Fermi term in Eq. (19). Abragam *et al.* have hypothesized  $s$ -electron promotion in ions<sup>20</sup> to explain effects such as seen in this experiment. By "promotion" is meant admixture with the ground ionic electronic state ( $6f^7$ ) of electronic configurations of the type  $ns6f^7rs$ , where  $n < 6$  and  $r \geq 6$ . Such a mechanism might possibly allow for such effects as seen in this experiment. Unfortunately, calculations based on this mechanism are difficult and have not been made.

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<sup>19</sup> V. Heine, Phys. Rev. **107**, 1002 (1957).

<sup>20</sup> A. Abragam, J. Horowitz, and M. H. L. Pryce, Proc. Roy. Soc. (London) **A 230**, 169 (1955).

<sup>18</sup> E. Fermi, Z. Physik **60**, 320 (1930).