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DIRECT MEASUREMENT OF THE MAGNETIC MOMENT OF ERBIUM-169

Walter M. Doyle and Richard Marrus

September 28, 1962
About thirty-five nuclear moments in the rare earth region have been
determined from paramagnetic resonance and atomic beam data. With the
exception of europium, these determinations rest entirely on hyperfine-structure
data and on assumptions made concerning the electronic fields at the nucleus.
These assumptions can be summarized briefly as follows: (a) For the free atom,
that the angular part of the wave function is amply described by assuming pure
Russell-Saunders coupling among the electrons of the configuration \((4f)^n\) to the
Hund's Rule ground term. (b) That the degree of configurational admixture is
small, and its effect is less than about 5%. (c) The value of \(\langle 1/r^3 \rangle\) for \(4f\)
electrons.

The evidence for assumption (a) comes from the observation that the free-
atom \(g_J\) values lie very close to the pure Russell-Saunders values.\(^1\) The theoretical
analysis of the measured \(g_J\) values has yielded a set of parameters that permits
accurate calculation of the correction to the hyperfine fields due to the breakdown
of Russell-Saunders coupling.\(^2\) These calculations have been performed for
praseodymium\(^3\) and promethium,\(^4\) and (by us) for erbium. In all cases, the
correction is of order 1%. Lindgren has neglected this effect in a recent recal-
culation of rare-earth moments.\(^5\)

The effect of configurations involving unpaired \(s\) electrons on the hyperfine
fields of triply ionized rare earths has been investigated by Bleaney.\(^6\) He esti-
mates that the correction for all rare earths is small, the maximum being about
6% for terbium. Lindgren believes that the effect should be even smaller for neutral rare earth atoms, and assigns an error of 5% in his calculations for this effect.

There is disagreement in the literature concerning the correct values of \( \langle 1/r^3 \rangle \). Bleaney has derived values for the triply ionized rare earths on the basis of hydrogenic functions with an estimated uncertainty of about 5%. Values have since been obtained for both the triply ionized and neutral atoms, by use of modified hydrogenic functions. These values differ from Bleaney's results by about 15 to 25%, and are also estimated to be uncertain to within about 5%. Hence the nuclear moments of an isotope determined from hyperfine-structure data can differ by as much as 25%, depending on whose values are used. The most recently reported work is that of Freeman and Watson, who have made nonrelativistic Hartree-Fock calculations for rare earth ions. They report values lying within 5% of Bleaney's, but they conclude that rather substantial uncertainties are to be associated with the moments derived from any theoretical \( \langle 1/r^3 \rangle \) values.

To obtain experimental information about these questions we have undertaken to measure by atomic beam magnetic resonance the hyperfine-structure constant, \( A \), and the magnetic moment, \( \mu_1 \), for Er\(^{169}\). We find \( A = 725.46(31) \text{Mc} \) and, for the directly measured moment, \( \mu_1 = 0.513(25) \text{nm} \), where the nuclear moment has been corrected for diamagnetic shielding. From the \( A \) constant and the value of \( \langle 1/r^3 \rangle \) derived from modified hydrogenic functions, we obtain \( \mu_1 = 0.504(50) \text{nm} \). This agreement with the directly measured value is excellent, and is interpreted by us as an important verification of Bleaney's idea that configuration admixing is not appreciable in rare earths, and of the \( \langle 1/r^3 \rangle \) values of Judd and Lindgren.

Prior work on erbium-169 had determined the ground-state spin (\( I = 1/2 \))
and the electronic angular momentum \( J = 6 \). The hyperfine structure of such a system is indicated in Fig. 1, and the energy levels are described by the Breit-Rabi Hamiltonian:

\[
\mathcal{H} = \mathbf{A} \cdot \mathbf{J} - g_J \mu_0 \mathbf{J} \cdot \mathbf{\gamma} - B_I \mu_0 \mathbf{I} \cdot \mathbf{I}.
\]

The effect of the term in \( g_1 \) on the erbium transition frequencies is estimated to be a few parts in \( 10^5 \), but is not negligible with an apparatus of sufficient precision. Our ability to determine \( g_1 \) for erbium rests on the good line width of our apparatus and on the ability to form very stable erbium beams which give highly reproducible data with good signal-to-noise ratio. A sample resonance curve is shown in Fig. 2.

Data were taken on the two transitions, which are observable by the flip-in method and are indicated in Fig. 1. The magnetic field was calibrated by observing the flip-in potassium resonance. It was found that, with this method of calibrating, the pulsating magnetic field of the nearby Bevatron caused small frequency shifts. All our data were obtained when the Bevatron was off.

The best fit to our data was obtained by a least-squares calculation on the IBM 709. The parameters best fitting our data are: \( A = 725.46(31) \text{Mc} \), \( g_J = -1.1638(1) \), and \( g_1 = 5.55(7) \times 10^{-4} \), where the error given for \( A \) and \( g_J \) is twice the standard deviation. Frequencies calculated by use of these parameters are compared with the observed frequencies in Table I.

The observed \( g_1 \) value must be increased by a factor 1.0078 to correct for diamagnetic shielding.\(^8\) This gives the true nuclear moment \( \mu_1 = 0.513(25) \text{nm} \).

We have investigated the possibility that there are contributions to the measured \( g_1 \) value that arise from other sources than the true nuclear moment. Let us write, for the nuclear moment term, \( g_1 (1 + \alpha) \mathbf{I} \cdot \mathbf{R} \), where \( g_1 \) is the true
nuclear gyromagnetic ratio and a arises from possible perturbations. The possible contributions to a that we have considered are (1) systematic errors, which cause the true magnetic field to be different from the measured field with an error proportional to the field (such an error arises, for example, if the field seen by the calibrating isotope differs from the field traversed by the erbium beam by an amount proportional to the field); (2) mixing of the fine-structure levels of the $^3\Pi$ term by the hyperfine structure of erbium in such a way as to introduce a pseudo $^1\Pi$ term in the Hamiltonian.

The possibility of systematic errors in the field has been investigated by us and is found to be completely negligible (i.e., $<10^{-6}$). We have calculated fine-structure mixing and find it to be less than 1%. Hence to within 1% our measured $g_\Pi$ is felt to be the true value.

From the $A$ constant, the magnetic moment can be inferred from the relation

$$\mu_\Pi = -\frac{1}{2}JA/\langle \hat{H} \rangle, m_J = J,$$

where $\langle \hat{H} \rangle$ is the expectation value for the magnetic field at the nucleus and is given by

$$\Pi = -2\mu_0 \left( \frac{1}{r^3} \right) \left( \sum \frac{1}{i} \left( \vec{r} - \vec{g} - \frac{3\vec{r}(\vec{g} \cdot \vec{r})}{r^2} \right) \right).$$

For $\langle 1/r^3 \rangle$ we have used the value of 9.84 atomic units given in reference 5. The evaluation of $\sum_i \xi_i$ depends on the assumptions made about the angular part of the wave function. If the ground configuration is assumed to be $(4f)^{12}$, only $^3\Pi$ and $^1\Pi$ can give states with $J = 6$, and the ground-state wave function will be of the form
\[ \psi = (1-a^2)^{1/2} |3H_6\rangle + a |1I_6\rangle . \] The value of \( a \) can be determined by diagonalizing the Coulomb and spin-orbit energies for \( J = 6 \). The Coulomb energy will be characterized by three Slater radial integrals, \( F_2, F_4, \) and \( F_6 \), while the spin-orbit energy is characterized by the parameter \( a_{4f} \). These parameters have been reliably evaluated for erbium by Judd and Lindgren, 2 and we use their results.

We obtain for \( a \) the value -0.094. The contributions to \( \langle \Sigma \rangle \) come from three terms:

\[ \langle \Sigma \rangle = (1-a^2) \langle 3H_6 | \Sigma | 3H_6 \rangle + 2a \langle 3H_6 | \Sigma | 1I_6 \rangle + a^2 \langle 1I_6 | \Sigma | 1I_6 \rangle . \]

The first term represents the contribution to the field from assumption (a) of the first paragraph lowered by an amount proportional to \( a^2 \). The other terms give the corrections due to the breakdown of Russell-Saunders coupling. The net value of \( \langle \Sigma \rangle \) is only about 1% different from that obtained from assumption (a), owing to fortuitous canceling in the last two terms. Therefore the moment inferred from the \( A \) value is \( \mu_1 = 0.504 \) nm.

The value of the \( \text{Er}^{169} \) moment can be compared to a predicted value of 0.7 based on the Bohr-Mottelson Nuclear Model. We have assumed that the state of the 101st neutron is characterized by \( [521 1/2] \) and a deformation parameter \( \delta \approx 0.3 \).
REFERENCES

Table I. Comparison of the observed frequencies with the frequencies predicted from the Hamiltonian (in Kc.) $725.46 \mathbf{I} \cdot \mathbf{J} + 1.1638 \mu_0 \mathbf{J} \cdot \mathbf{H} - 5.55 \times 10^{-4} \mu_0 \mathbf{I} \cdot \mathbf{H}$.

<table>
<thead>
<tr>
<th>Field (gauss)</th>
<th>Predicted frequency (Kc.)</th>
<th>Observed frequency (Kc.)</th>
<th>Residual (Kc.)</th>
<th>Transition</th>
</tr>
</thead>
<tbody>
<tr>
<td>467.595</td>
<td>704496.5</td>
<td>704499.0(4.0)</td>
<td>2.5</td>
<td>F = 13/2</td>
</tr>
<tr>
<td>504.329</td>
<td>760020.2</td>
<td>760020.0(4.0)</td>
<td>-0.2</td>
<td>F = 13/2</td>
</tr>
<tr>
<td>540.903</td>
<td>815336.0</td>
<td>815332.0(4.0)</td>
<td>-4.0</td>
<td>F = 13/2</td>
</tr>
<tr>
<td>540.903</td>
<td>815336.0</td>
<td>815336.0(2.4)</td>
<td>0.0</td>
<td>F = 13/2</td>
</tr>
<tr>
<td>613.698</td>
<td>925539.9</td>
<td>925533.0(4.0)</td>
<td>-6.9</td>
<td>F = 13/2</td>
</tr>
<tr>
<td>649.962</td>
<td>980996.1</td>
<td>980995.0(2.4)</td>
<td>-1.1</td>
<td>F = 13/2</td>
</tr>
<tr>
<td>686.158</td>
<td>1035387.7</td>
<td>1035388.0(2.4)</td>
<td>0.3</td>
<td>F = 13/2</td>
</tr>
<tr>
<td>722.298</td>
<td>1090232.4</td>
<td>1090237.0(2.4)</td>
<td>4.6</td>
<td>F = 13/2</td>
</tr>
<tr>
<td>722.298</td>
<td>1090232.4</td>
<td>1090235.0(4.0)</td>
<td>2.6</td>
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<tr>
<td>794.442</td>
<td>1199836.1</td>
<td>1199835.0(2.4)</td>
<td>-1.1</td>
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<tr>
<td>196.279</td>
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<tr>
<td>317.719</td>
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<td>557480.0(5.6)</td>
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<tr>
<td>467.595</td>
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<td>820290.0(4.0)</td>
<td>-3.9</td>
<td>F = 11/2</td>
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<tr>
<td>467.595</td>
<td>820293.9</td>
<td>820292.0(8.0)</td>
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<tr>
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<tr>
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</tr>
<tr>
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<td>0.1</td>
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</tr>
<tr>
<td>794.442</td>
<td>1392177.4</td>
<td>1392180.0(4.0)</td>
<td>2.6</td>
<td>F = 11/2</td>
</tr>
</tbody>
</table>
FIGURE LEGENDS

1. Hyperfine structure of the system $I = 1/2, J = 6$, showing the observable transitions.

2. Sample resonance in the $F = 13/2$ level.
Fig. 1
Er\textsuperscript{169}

\[ H = 540.903 \text{ gauss} \]

\[ I = \frac{1}{2} \]

\[ J = 6 \]

\[ F = \frac{13}{2} \]

Fig. 2
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