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Spin and Magnetic Moment of Eu^{152} and Eu^{154} by Paramagnetic Resonance*

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The microwave paramagnetic resonance hyperfine structures of 13-year Eu^{152} and 15-year Eu^{154} have been observed in a sample of Eu^{2+} which was incorporated in a KCl powder. From the number of hfs lines the spins were found to be $I(Eu^{152})=3$ and $I(Eu^{154})=3$. Neglecting hfs anomalies, from the ratios of the total hfs splitting these magnetic moment ratios were obtained: $|\mu(Eu^{154})|/|\mu(Eu^{153})|=1.308\pm0.004$, $|\mu(Eu^{152})|/|\mu(Eu^{151})|=0.5574\pm0.006$, $|\mu(Eu^{151})|/|\mu(Eu^{153})|=2.264\pm0.006$. Using $\mu(Eu^{153})\cong1.6$ nm, we find $|\mu(Eu^{154})|\cong2.1$ nm and $|\mu(Eu^{152})|\cong2.0$ nm. These results are discussed in relation to the nuclear shell model.

I. INTRODUCTION

THIS paper describes the measurement of the spins and magnetic moments of pile-produced Eu^{152} and Eu^{154} by the method of microwave paramagnetic resonance absorption[1]; some of these results were briefly announced earlier.[2] For details on techniques and experimental apparatus we refer to previous papers.[3]

II. THEORY

The ground state of Eu^{++} is a half-filled $4f$ shell giving a configuration of $4f^7$, $^8S_{7/2}$. Owing to the zero orbital angular momentum the ions behave almost like free spins in a cubic crystalline field. The spectroscopic splitting factor g is isotropic and very nearly 2.0, allowing the use of a powder instead of a single crystal. Paramagnetic resonance in Eu was first observed by Bleaney and Low[4] in powdered cubic SrS containing a trace of Eu. An isotropic hyperfine structure of the two stable isotopes, Eu^{151} (47.77%) and Eu^{153} (52.23%), was clearly resolved; $I=5/2$ for each. The magnitude of the hfs splitting suggests the promotion of an s electron to the $6s$ state by configuration coupling.

In a powdered cubic crystal the spectra may be described by a spin Hamiltonian of the form

$$\mathcal{H}=g\beta(\mathbf{H}\cdot\mathbf{S}+A\mathbf{I}\cdot\mathbf{S}), \qquad (1)$$

where β is the Bohr magneton and the first term represents the electronic Zeeman interaction in the external magnetic field $\mathbf{H}$; the second term is the magnetic hyperfine interaction of the electronic spin $\mathbf{S}$ with the nuclear spin $\mathbf{I}$. We have omitted all crystalline-field splitting terms because in a powder only the transitions $S_z=M=\frac{1}{2}\rightarrow-\frac{1}{2}$ are independent of crystalline orientation to first order, so that normally only these transitions will be observed as sharp lines. There are $2I+1$ of these magnetic dipole transitions, whose positions for a constant applied microwave frequency $\nu=g\beta H_0/h$ are, to second order,

$$H(m)=H_0-Am-\left[\frac{A^2}{2(H_0-Am)}\right][I(I+1)-m^2], \qquad (2)$$

where $m=I_z$. Since the hyperfine coupling constant A is proportional to $g_I=\mu/I$ (if hyperfine anomalies are neglected), we see that the total hfs splitting $\Delta H=[H(m=-I)-H(m=I)]\propto\mu$, to high order.

In addition to the strongly allowed transitions described above, the spectra also show weak lines occurring in pairs between the main hfs lines. The positions of these lines, but not the intensities, may be explained by adding the term $g_I\beta\mathbf{H}\cdot\mathbf{I}$ to the Hamiltonian (1). The weak lines then correspond to the transitions $M=\frac{1}{2}\rightarrow-\frac{1}{2}$, $\Delta m=\pm1$. They will not be discussed further here, although if the above explanation proves to be correct the spacing between the lines will give a direct measurement of g_I.

III. EXPERIMENTAL PROCEDURES AND RESULTS

A few millicuries of 15-year Eu^{154} were prepared in the (n,γ) reaction by irradiating natural-abundance Eu to a total integrated thermal-neutron flux of $nvt=7.3\times10^{20}/cm^2$ in the Materials Testing Reactor at Arco, Idaho. Eu^{151} and Eu^{152} were burned out by this high flux leaving ~88% Eu^{153} and ~12% Eu^{154} in this sample, referred to as A. Another sample, B, was reclaimed from the fission products of Pu, containing 78.3% Eu^{153}, 15.2% Eu^{154}, and 6.5% Eu^{155} as determined by mass spectrographic analysis. The 13-year Eu^{152} was made by the (n,γ) reaction in enriched Eu^{151} (91%) by irradiating in the Materials Testing Reactor at Arco, Idaho to a total intergrated thermal-neutron flux of $nvt=0.63\times10^{20}/cm^2$. This sample C had the approximate abundance 85% Eu^{151}, 7% Eu^{152}, 8% Eu^{153}. The Eu was converted to the chloride and added to about 300 mg of KCl which served as a cubic host and also made the sample magnetically dilute, which is essential in order to prevent dipole-dipole broadening of the resonance lines. The sample was dried in the

* Supported in part by the U. S. Atomic Energy Commission.
1 B. Bleaney and K. W. H. Stevens, Repts. Progr. in Phys. 16, 108 (1953); K. D. Bowers and J. Owen, Repts. Progr. in Phys. 18, 304 (1955).
2 Kedzie, Abraham, and Jeffries, Bull. Am. Phys. Soc. Ser. II, 1, 390 (1956).
3 Dobrowolski, Jones, and Jeffries, Phys. Rev. 101, 1001 (1956); Kedzie, Abraham, and Jeffries, Phys. Rev. 108, 54 (1957), preceding paper.
4 B. Bleaney and W. Low, Proc. Phys. Soc. (London) A68, 65 (1955); W. Low, Phys. Rev. 98, 430 (1955).

presence of HCl gas to insure the formation of Eu^{2+}. The usual oxidation state Eu^{3+} has the nonmagnetic ground state $^7F_0$ and shows no paramagnetic resonance. The mixture was fused, ground to a powder, and placed in the microwave cavity of the paramagnetic resonance apparatus.[3]

The magnetic resonance was observed at 77°K and a frequency of 9300 Mc/sec. Both samples A and B showed the six hfs lines of stable Eu^{153}, upon which were superposed a weaker set of seven hfs lines with the same g factor. These we identify as due to Eu^{154} by comparison of the intensities with those calculated from the mass spectrographic measurements. We thus conclude that $I(Eu^{154})=3$. From the ratios of the total hfs splittings ΔH, we find

$$\Delta H(Eu^{154})/\Delta H(Eu^{153})=1.308\pm0.004 = |\mu(Eu^{154})|/|\mu(Eu^{153})|, \quad (3)$$

neglecting hfs anomalies. From optical hfs measurements, Schuler and Schmidt[5] and Brix[6] have found $\mu(Eu^{151})\cong3.6$ nm and $\mu(Eu^{153})\cong1.6$ nm. Using the latter value and our ratio (3) yields $|\mu(Eu^{154})|\cong2.1$ nm.

Sample B did not show any hfs lines that could be attributed to Eu^{155}, which was 6.5% abundant. The signal-to-noise ratio was such that they should have been observed unless they were closely superposed on the strong Eu^{153} lines.

The spectrum of sample C showed the six strong hfs lines of Eu^{151}, six weaker lines of Eu^{153} and, in addition, a set of seven partially obscured hfs lines with the same g factor. These we attribute to Eu^{152}, showing $I(Eu^{152})=3$. The measured ratios of total hfs splittings are

$$\Delta H(Eu^{152})/\Delta H(Eu^{151})=0.5574\pm0.006 = |\mu(Eu^{152})|/|\mu(Eu^{151})|, \quad (4)$$

and

$$\Delta H(Eu^{151})/\Delta H(Eu^{153})=2.264\pm0.006 = |\mu(Eu^{151})|/|\mu(Eu^{153})|. \quad (5)$$

Using $\mu(Eu^{151})=3.6$ nm[5,6] yields $|\mu(Eu^{152})|=2.0$ nm.

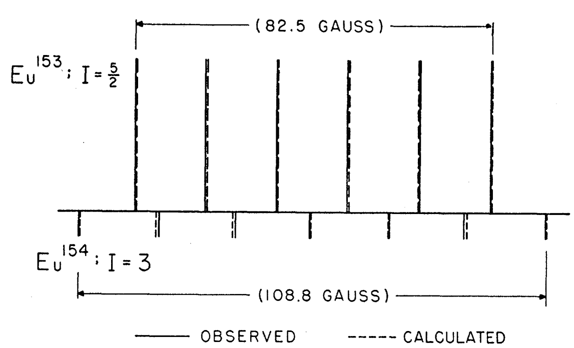

FIG. 1. Schematic representation of the observed and calculated paramagnetic hfs lines of Eu^{153} and 15-year Eu^{154} in powdered KCl.

[5] H. Schuler and T. Schmidt, Z. Physik **94**, 457 (1935); T. Schmidt, Z. Physik **108**, 408 (1938).

[6] P. Brix, Z. Physik **132**, 579 (1952); P. F. A. Klinkenberg, Revs. Modern Phys. **24**, 63 (1952).

From the ratios (3), (4), and (5) one obtains

$$\Delta H(Eu^{152})/\Delta H(Eu^{154})=0.965\pm0.010 = |\mu(Eu^{152})|/|\mu(Eu^{154})|. \quad (6)$$

The above assignments of spin and magnetic moments were further checked in each case by comparing all the observed lines with those calculated by using the second-order expression (2). Excellent agreement was obtained. Figure 1 is a schematic representation of the observed and calculated spectra for Eu^{153} and Eu^{154}.

The absolute magnitude of the hfs coupling constant A was measured, with the results

$$A(Eu^{151})=0.003256\pm0.000006\ \text{cm}^{-1},$$
$$A(Eu^{152})=0.001512\pm0.000015\ \text{cm}^{-1},$$
$$A(Eu^{153})=0.001438\pm0.000003\ \text{cm}^{-1},$$
$$A(Eu^{154})=0.001567\pm0.000006\ \text{cm}^{-1}.$$

IV. DISCUSSION

The measured spin $I=3$ of both $_{63}Eu_{89}{}^{152}$ and $_{63}Eu_{91}{}^{154}$ is consistent with their respective decay schemes.[7] The experimental fact that the nuclear magnetic moments are very nearly the same is consistent with the simple assumption that the configuration of each is an odd proton jj coupled to an odd neutron with the nuclear g factors very similar in each case. Although the quite different magnetic moments of Eu^{151} and Eu^{153} can only be understood in terms of the collective model,[8] we nevertheless assume a $d_{5/2}$ proton state for Eu^{152} and Eu^{154}. The measured spin $I=3/2$ for $_{64}Gd_{91}{}^{155}$ is strong empirical evidence for a $p_{3/2}$ neutron state for Eu^{152} and Eu^{154}. For the $(d_{5/2},p_{3/2})$ configuration in simple jj coupling to $I=3$, we calculate $\mu=2.96$ nm, if we take the Schmidt-limit values for g_p and g_n. On the other hand, if we take the empirically determined values $g_n=-0.21$ (from Gd^{155} measurements)[9] and $g_p\cong+1.04$ (the average value for Eu^{151} and Eu^{153}),[5,6] we obtain $\mu=2.03$ nm in surprisingly good agreement with our measured values above. This is perhaps somewhat fortuitous, since if one uses instead of the average g_p for Eu^{151} and Eu^{153}, the individual values, one obtains $\mu=2.93$ nm and $\mu=1.17$ nm, respectively.

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[7] M. Goldhaber (private communication).

[8] B. R. Mottelson and S. G. Nilsson, Phys. Rev. **99**, 1615 (1955); K. Gottfried, Phys. Rev. **103**, 1017 (1956); K. Gottfried, thesis, Massachusetts Institute of Technology, June, 1955 (unpublished).

[9] D. R. Speck, Phys. Rev. **101**, 1725 (1956); W. Low, Phys. Rev. **103**, 1309 (1956).