

Spin States of Neutron Resonances in Gadolinium and Europium

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The spin states associated with several neutron resonances in gadolinium and europium have been assigned by observing the transmission of polarized neutrons through polarized targets. For the levels in Gd^{155} at 0.0268, 2.01, 2.57, and 6.30 eV, the spin states were found to be $J=2, 1, 2,$ and $2,$ respectively, and for the 0.0314-eV level in Gd^{157} , $J=2$. For the resonances in Eu^{151} at 0.32, 0.46, 1.06, and 3.37 eV, the spin states were found to be $J=3, 3, 3,$ and $2,$ respectively, and for the negative level, $J=3$. For the 2.46-eV resonance in Eu^{153} , $J=3$. There appears to be no correlation between these measured spin states and other resonance parameters. The magnitude and direction of the observed effects are discussed in terms of the effective magnetic field at the nucleus and the magnetic properties of the metals at low temperature.

INTRODUCTION

THE slow neutron resonances in gadolinium^{1,2} and europium³⁻⁵ have been extensively investigated and many of the resonance parameters have been measured with accuracy, but the spin states have remained unknown. We have therefore undertaken an investigation of these spin states primarily to determine if any correlation exists between the spins and the other resonance parameters, as had been observed⁶ in the case of In^{115} . Spin assignments were made by observing the interaction of polarized neutrons with polarized targets. Since both of these metals are magnetic materials, we must know their magnetic behavior at low temperature, and also information on the hyperfine magnetic field acting at the nucleus. In the case of gadolinium, a value for the hyperfine coupling constant can be obtained from the data. This was not possible in the case of europium, where the magnetic behavior at low temperature is much more complicated. The relevant nuclear and magnetic properties are listed in Table I.

EXPERIMENT

The transmission of polarized, monochromatic neutrons through polarized targets was observed as a function of the orientation of the neutron spin with respect to the target polarization. For each resonance, $J=I+\frac{1}{2}$ or $J=I-\frac{1}{2}$, since only s -wave neutrons are involved. The target polarization was produced by placing the sample in an 18.0-kOe field and reducing its temperature to a few hundredths of a degree Kelvin via thermal contact with a demagnetized paramagnetic salt. The cooling salts were grown around flattened silver or gold wires and the samples were attached to the other end of the wires. Both chromium potassium alum and iron ammonium alum salts were used. The (111) planes of a magnetized cobalt-iron crystal were used to select

neutrons from the NRL reactor spectrum. Experimental details about the cooling procedure and the polarized-neutron crystal spectrometer have been given previously.⁶ Metallic samples⁷ were used so that the nuclear spin-lattice relaxation times were short. Since europium metal deteriorates rapidly in air, foils were pressed under kerosene. In some of the experiments, the foils were coated with Apiezon-N grease and squeezed to the silver wires with Teflon clamps. In other experiments, the foils were coated with copper or silver, and the wires were then soldered to it. Both methods provided satisfactory thermal contact. The salt-sample assemblies were suspended by thin nylon fibers for thermal isolation. The temperature of the cooling salt was monitored by taking magnetic susceptibility measurements; the average salt temperature for a run lasting several hours was typically about 0.06°K.

Whenever possible, filters were used to minimize the effects of second-order reflections. This proved to be particularly important when the cross section for first-order neutrons was large. Some of our preliminary spin assignments, based upon data in which filters were not used, were incorrect.⁸ The second-order contaminant was measured at several neutron energies (using filters),

TABLE I. Nuclear and magnetic properties of gadolinium and europium. The nuclear spins are in units of \hbar and the magnetic moments are in nuclear magnetons.

Isotope	I	μ	Magnetic behavior at low temperature
Gd^{155}	$\frac{3}{2}^a$	-0.32 ^a	Ferromagnetic ^d
Gd^{157}	$\frac{3}{2}^a$	-0.40 ^a	Ferromagnetic ^d
Eu^{151}	$\frac{5}{2}^b$	+3.42 ^c	Antiferromagnetic (helical) ^e
Eu^{153}	$\frac{5}{2}^b$	+1.51 ^c	Antiferromagnetic (helical) ^e

^a N. I. Kaliteevskii, M. P. Chaika, I. Kh. Pacheva, and É. E. Fradkin, *Zh. Eksperim. i Teor. Fiz.* **37**, 882 (1959) [English transl.: *Soviet Phys.-JETP* **10**, 629 (1960)]; D. R. Speck, *Phys. Rev.* **101**, 1725 (1956).

^b H. Schuler and T. Schmidt, *Z. Physik* **94**, 457 (1935).

^c F. M. Pichanick, P. G. H. Sandars, and G. K. Woodgate, *Proc. Roy. Soc. (London)* **A257**, 277 (1960).

^d See Ref. 21.

^e See Ref. 35.

⁷ We wish to thank F. H. Spedding of the Ames Laboratory, Iowa, and T. T. Campbell of the U. S. Bureau of Mines Experimental Station, Albany, Oregon, for kindly supplying us with europium metal in the early stages of this work.

⁸ A. Stolovy, *Bull. Am. Phys. Soc.* **5**, 294 (1960); **6**, 275 (1961).

¹ E. T. Florance, see H. B. Møller, F. J. Shore, and V. L. Sailor, *Nucl. Sci. Eng.* **8**, 183 (1960).

² F. B. Simpson and R. G. Fluharty, *Bull. Am. Phys. Soc.* **2**, 42 (1957).

³ H. H. Landon and V. L. Sailor, *Phys. Rev.* **98**, 1267 (1955).

⁴ F. Dominac and E. T. Patronis, *Phys. Rev.* **114**, 1577 (1959).

⁵ S. Tassan, A. Hellsten, and V. L. Sailor, *Nucl. Sci. Eng.* **10**, 169 (1961).

⁶ A. Stolovy, *Phys. Rev.* **118**, 211 (1960).

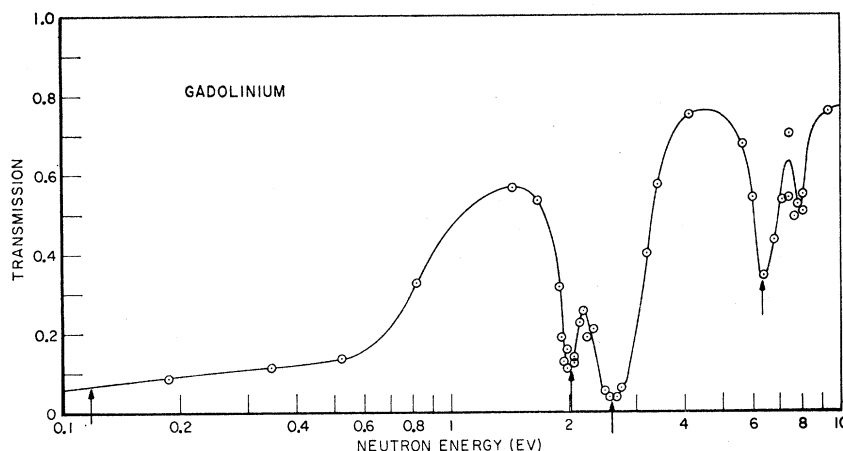


FIG. 1. Transmission of 0.63-cm-thick gadolinium metal sample versus neutron energy. Data on resonance spin states were taken at the positions shown by arrows.

and was found to vary between 5 and 8% of the open-beam intensity. All the experimental results were corrected for the presence of second order. Higher orders are assumed to be negligible. The results were also corrected for the effects of spectrometer resolution in the manner outlined by the Brookhaven group.⁹ The integrals involved were evaluated numerically. When necessary, corrections were made for the presence of other resonances, and for potential scattering.

RESULTS AND DISCUSSION

A. Gadolinium

A transmission plot of resonances in gadolinium taken with the polarized-neutron spectrometer is shown in Fig. 1. Data on resonance spins were taken at the positions indicated by arrows. The observed percent changes in the transmitted intensity $\Delta t/t$ upon reversing the neutron-beam polarization direction are given in Table II. We call the sign of this effect positive if it is in the direction we would expect if the nuclei are polarized parallel to the applied external field and $J=I+\frac{1}{2}$, i.e., the transmission is smaller when the neutron spins are parallel to the applied field. The effect observed at 0.118 eV is due to a superposition of the high-energy tails of the resonances at 0.0268 eV in Gd¹⁵⁵ and at 0.0314 eV in Gd¹⁵⁷, with the latter predominating.¹ This energy was chosen so that an Er filter could be used to remove almost all of the second-order contamination. Similarly, a Sm filter at 2.01 eV and a Ta filter at 2.57 eV effectively remove second-order reflections. A cadmium filter was used for all resonances above 1 eV to reduce the thermal-neutron background.

The magnitude of the observed transmission changes is quite large in view of the small magnetic moments of the gadolinium isotopes. This indicates that the effective field at the nucleus is more than an order of magni-

tude larger than the externally applied field. All available information indicates that this effective field is *negative* (i.e., opposite to the applied field). Calculations by Watson and Freeman¹⁰ indicate a negative field in the vicinity of the nucleus of the Gd³⁺ ion, and measurements by Caspari *et al.*¹¹ have shown that the hyperfine field is negative in Gd. Since the magnetic moment is also negative, the nuclear spins are polarized *parallel* to the applied field.

We now examine the origin of the hyperfine field. The total magnetic field at the nucleus can be considered as a sum of several contributions^{12,13}: (1) a local field which consists of the external, Lorentz, and demagnetizing fields; (2) a contact field from partially polarized 6s conduction electrons; (3) a hyperfine field due to the interaction of the nucleus with its orbital 4*f* electrons; and (4) a hyperfine field due to the exchange interaction between the inner core *s* electrons and the 4*f* electrons. Jaccarino *et al.*¹⁴ have demonstrated that the second contribution is negative for both Gd and Eu from

TABLE II. Observed percent transmission changes ($\Delta t/t$) upon reversing the neutron-beam polarization for resonances in gadolinium. A positive change means that the transmission was smaller when the neutron spins were parallel to the applied field. Gadolinium metal samples were used.

Neutron energy (eV)	Filters	Sample thickness (cm)	Observed transmission	($\Delta t/t$) in %
0.118	Er	0.0114	0.176	+4.72±0.07
2.01	Sm, Cd	0.190	0.424	-2.88±0.11
2.57	Ta, Cd	0.0635	0.298	+2.58±0.16
6.30	Cd	0.635	0.360	+0.86±0.13

¹⁰ R. E. Watson and A. J. Freeman, Phys. Rev. Letters **6**, 277 (1961); **6**, 388 (1961).

¹¹ M. E. Caspari, S. Frankel, D. Ray, and G. T. Wood, Phys. Rev. Letters **6**, 345 (1961); and private communication.

¹² W. Marshall, Phys. Rev. **110**, 1280 (1958).

¹³ J. Kondo, J. Phys. Soc. Japan **16**, 1690 (1961).

¹⁴ V. Jaccarino, B. T. Matthias, M. Peter, H. Suhl, and J. H. Wernick, Phys. Rev. Letters **5**, 251 (1960); and private communication).

⁹ H. Postma, H. Marshak, V. L. Sailor, F. J. Shore, and C. A. Reynolds, Phys. Rev. **126**, 979 (1962); H. Marshak, H. Postma, V. L. Sailor, F. J. Shore, and C. A. Reynolds, *ibid.* **128**, 1287 (1962).

Knight-shift measurements. The third contribution is usually by far the most important in the rare-earth metals.¹³ However, gadolinium metal is trivalent,¹⁵ so that the electronic configuration is $4f^7 \ ^8S_{7/2}$, which is a half-filled shell. Thus, the third contribution is zero. The fourth contribution appears to be the dominant one in this case. It arises because the exchange interaction of an inner s electron with the magnetic f electrons depends upon whether the s electron is parallel or antiparallel to the total f electron spin.¹⁶ Since the probability density of s electrons at the nucleus is large, the result is a large negative effective field. This is the same mechanism which has been proposed^{17,18} to explain the large negative effective field in iron.¹⁹

We may write a nuclear spin Hamiltonian (containing only terms involving the nuclear spin) as

$$\mathcal{H}_I = A \mathbf{J}_e \cdot \mathbf{I} - g_N \beta_N (\mathbf{H}_{10c} + \mathbf{H}_c) \cdot \mathbf{I}, \quad (1)$$

where A is the hyperfine structure constant, \mathbf{J}_e is the electronic angular momentum, \mathbf{I} is the nuclear spin, $g_N \equiv \mu_N / I \beta_N$ is the nuclear g factor, β_N is the nuclear magneton, \mathbf{H}_{10c} is the local magnetic field (primarily the applied field), and \mathbf{H}_c is the effective field produced by the contact interaction with polarized $6s$ conduction electrons. Thus, the total effective field at the nucleus can be written as

$$\mathbf{H}_{\text{eff}} = \mathbf{H}_{10c} + \mathbf{H}_c - (A \mathbf{J}_e / g_N \beta_N). \quad (2)$$

For $\mu_N H_{\text{eff}} \ll kT$, the nuclear polarization is then given by²⁰

$$f_N = -\frac{1}{3} \frac{I + 1}{I} \frac{\mu_N}{kT} \left[H_{10c} + \left(H_c - \frac{A J_e}{g_N \beta_N} \right) f_e \right], \quad (3)$$

where f_e is the fraction of magnetic saturation of the electron spins.

Gadolinium metal is known to be ferromagnetic at low temperature,²¹ and under the conditions of this experiment, the magnetic domains are completely aligned²² so that $f_e = 1$ and $J_e = -\frac{7}{2}$. We can then obtain f_N from the size of the observed effects after taking into account the effects of instrumental resolution as in Ref. 9, and correcting for second-order contamination. Because the sample was magnetically saturated, we consider depolarization of the beam while passing through the sample to be negligible, even for thermal neutrons. We have also considered the effect of magnetic

TABLE III. Spin assignments for resonances in gadolinium. Also listed are the total radiation widths.

Resonance energy (eV)	Isotope	J	Γ_γ (eV)
0.0268	155	2	0.108 ± 0.001^a
0.0314	157	2	0.106 ± 0.001^a
2.01	155	1	0.110 ± 0.001^a
2.57	155	2	0.111 ± 0.001^a
6.30	155	2	0.106 ± 0.020^b

^a See Ref. 1.

^b See Ref. 2 and Hughes and Schwartz, Brookhaven National Laboratory Report BNL-325, 1958 (unpublished).

scattering.²³ An experiment was performed at 0.118 eV with a 0.019-cm-thick sample at liquid-nitrogen temperature, which is well below the Curie point. Since the nuclear polarization is negligible here, the observed effect $\Delta t/t = +[0.41 \pm 0.12]\%$ is due to the magnetic electrons. It is only a small part of the observed effect at 0.085°K. We consider this correction to be negligible for the resonances above 2 eV. Finally, we must correct the data taken at 2.01 eV for the influence of the 2.57-eV resonance which has the opposite spin.

Spin assignments for five resonances in gadolinium are given in Table III. Our assignments for the two thermal energy resonances are in agreement with those of Bartholomew and co-workers.²⁴ The 0.0314-eV resonance in Gd^{157} predominates at 0.118 eV, for which clearly $J=2$. The spin of the 0.0268-eV resonance in Gd^{155} is obtained by comparing our observed effect with the effects we would expect for the two possible cases. In making these calculations, we use the cross sections given by Møller *et al.*,¹ and the hyperfine structure constants given by Low²⁵ to compute the nuclear polarization from Eq. (3). The sample temperature was taken to be 0.085°K to within 10%. This was obtained from an independent experiment with a Re-Fe alloy in which the polarization effect was observed as a function of the temperature. Using an applied field of 18.0 kOe and a saturation magnetization²⁶ of 1990 Oe/cm³, the local field¹² polarization was calculated to be only 0.17% for Gd^{155} and 0.20% for Gd^{157} . The contact field was taken to be $H_c = -50$ kOe.¹⁴ The results of these calculations are as follows: if the spin of the 0.0268-eV resonance is $J=1$, we expect to see a $+2.88\%$ effect, and if it is $J=2$, the expected effect is $+4.56\%$, with uncertainties of about 20%. The observed effect is $+ [4.72 \pm 0.07]\%$, of which about $+0.25\%$ is due to magnetic electron scattering. Thus, $J=2$ is the correct assignment for the 0.0268-eV resonance.

Our spin assignment for the 2.57-eV level is *not* in agreement with resonance scattering measurements of

²³ R. I. Schermer, Phys. Rev. **130**, 1907 (1963).

²⁴ G. A. Bartholomew, in *Proceedings of the International Conference on Nuclear Structure, Kingston, Canada* (The University of Toronto Press, Toronto, 1960), p. 573.

²⁵ W. Low, Phys. Rev. **103**, 1309 (1956).

²⁶ R. M. Bozorth, *Ferromagnetism* (D. Van Nostrand, Inc., New York, 1951), p. 342.

¹⁵ A. F. Kip, C. Kittel, A. M. Portis, R. Barton, and F. H. Spedding, Phys. Rev. **89**, 518, (1953).

¹⁶ V. Heine, Phys. Rev. **107**, 1002 (1957).

¹⁷ D. A. Goodings and V. Heine, Phys. Rev. Letters **5**, 370 (1960).

¹⁸ A. J. Freeman and R. E. Watson, Phys. Rev. Letters **5**, 498 (1960).

¹⁹ S. S. Hanna, J. Heberle, G. J. Perlow, R. S. Preston, and D. H. Vincent, Phys. Rev. Letters **4**, 513 (1960).

²⁰ M. E. Rose, Phys. Rev. **75**, 213 (1949).

²¹ J. P. Elliott, S. Legvold, and F. H. Spedding, Phys. Rev. **91**, 28 (1953).

²² W. E. Henry, J. Appl. Phys. **29**, 524 (1958).

Ceulemans.²⁷ Such experiments are difficult because highly accurate measurements are required to make an unambiguous spin assignment, while the counting rates are usually low. We can demonstrate that $J=2$ is the correct assignment by using the very elegant method of Postma, Shore, and Reynolds,²⁸ which requires no assumptions about the direction of the effective field at the nucleus or the sign of the magnetic moment. It depends simply on the ratio of the factor ρ (which occurs in the formulas for the observed effect⁹ $\Delta t/t$) for two resonances with different spin, since $\rho = I/I+1$ for a $J = I + \frac{1}{2}$ resonance and $\rho = -1$ for a $J = I - \frac{1}{2}$ resonance. In our case, this is closely approximated by

$$\frac{\rho_{2.01}}{\rho_{2.57}} = \frac{(\Delta t/t)_{2.01} \langle N\sigma t \rangle_{2.57}}{(\Delta t/t)_{2.57} \langle N\sigma t \rangle_{2.01}}, \quad (4)$$

where the average values $\langle N\sigma t \rangle$ are resolution corrected values given closely by

$$\langle N\sigma t \rangle = \frac{\int_0^\infty R(E-E') e^{-N\sigma t} N\sigma t dE'}{\int_0^\infty R(E-E') e^{-N\sigma t} dE'}, \quad (5)$$

where $R(E-E')$ is the resolution function. The integrals were evaluated numerically using the resolution parameters given by Møller *et al.*¹ to obtain $\langle N\sigma t \rangle_{2.01} = 0.710$ and $\langle N\sigma t \rangle_{2.57} = 0.950$ for the samples given in Table II. The observed effect for the 2.01-eV resonance must be corrected for the influence of the 2.57-eV resonance to yield a true effect $(\Delta t/t)_{2.01} = 3.36\%$. Thus we obtain $\rho_{2.01}/\rho_{2.57} = -1.74 \pm 0.25$, which is in good agreement with the theoretical value $-5/3$ for the spin assignments we have made, and is incompatible with the reverse assignment of spins which would yield $-\frac{3}{5}$. This constitutes proof that the effective field at the nucleus is negative.

The analyzed data for the 2.01- and 2.57-eV resonances yield for the nuclear polarization $f_N = [2.80 \pm 0.20]\%$ at an average temperature of 0.075°K . We can now use this to compute the hyperfine structure constant A from Eq. (3). The result for Gd^{155} is, in temperature units, $A_{155}/k = [6.6 \pm 1.5] \times 10^{-4}\text{K}$, where the error is due primarily to uncertainties in H_c and the average sample temperature. Since we have removed the effects of polarized conduction electrons and the magnetization of the sample, we can compare this result directly to the electron spin resonance data of Low²⁵ obtained in non-metallic host crystals. His result, in temperature units, is $A_{155}/k = [5.4 \pm 0.4] \times 10^{-4}\text{K}$. The agreement is quite good in view of the rather small value of A . We can also obtain A for Gd^{157} from the data taken at 0.118 eV by

subtracting the effect of the 0.027-eV level. We thus obtain $A_{157}/k = [7.0 \pm 1.6] \times 10^{-4}\text{K}$. Low's results are $A_{157}/k = [7.15 \pm 0.23] \times 10^{-4}\text{K}$ or $[7.6 \pm 0.4] \times 10^{-4}\text{K}$ depending upon the host crystal used. Again the agreement with our result is good. The total hyperfine field in the metal corresponding to these results (i.e., including H_c , but excluding H_{loc}) is $H_{hf} = [-324 \pm 60]\text{kOe}$, where the error includes the spread in measurements of the magnetic moment. Freeman and Watson²⁸ suggest an exchange-interaction contribution to the hyperfine field of the form $-90(g_J - 1)J$ kOe for trivalent rare-earth ions. For Gd^{3+} , $g_J = 2$ and $J = \frac{7}{2}$ so this becomes -315 kOe (not including H_c), which agrees reasonably well with our result.

The total radiation widths listed in Table III do not appear to be spin-dependent. In particular, the 2.01- and 2.57-eV resonances have the same radiation width although the spins are different. However, recent resonance capture gamma-ray work by Vogt³⁰ indicates that these two resonances have somewhat different decay schemes; the 2.01-eV resonance shows a transition to the zero-spin ground state, while for the 2.57-eV resonance the transition appears to go instead to the first excited state at 89 keV with spin 2. Assuming dipole transitions, this is consistent with our spin assignments.

B. Europium

A transmission plot for resonances in europium is shown in Fig. 2. Data on resonance spins were taken at the eight positions indicated by arrows. At many of these positions, more than one resonance contributes to the observed effect. The results are given in Table IV. Europium metal is divalent,³¹ so it has the same electronic configuration as Gd^{2+} , and we would expect the hyperfine field to be of the same sign and order of magnitude as in gadolinium metal. An electron-nuclear double resonance experiment³² has shown that the hyperfine interaction is negative for Eu^{2+} ions. Since the field produced by the polarized conduction electrons is also negative,¹⁴ the hyperfine field in Eu metal must be negative. The magnetic moments of the Eu isotopes are positive, so the nuclear spins are polarized *opposite* to the applied field.

Although the magnetic moment of Eu^{151} is an order of magnitude greater than that for Gd^{155} , the observed effects are rather small. This indicates that the magnetic behavior of europium metal at low temperature is very different from that of gadolinium metal. Indeed, magnetic susceptibility measurements at low temperature^{33,34} show that europium is not ferromagnetic,

²⁷ A. J. Freeman and R. E. Watson, Phys. Rev. **127**, 2058 (1962).

²⁸ R. H. Vogt (private communication).

²⁹ M. Peter and B. T. Matthias, Phys. Rev. Letters **4**, 449 (1960).

³⁰ W. E. Blumberg and J. Eisinger, Bull. Am. Phys. Soc. **6**, 141 (1961).

³¹ R. M. Bozorth and J. H. Van Vleck, Phys. Rev. **118**, 1493 (1960).

³² R. L. Zanowick and W. E. Wallace, Phys. Rev. **126**, 537 (1962).

²⁷ H. Ceulemans (private communication).

²⁸ H. Postma, F. J. Shore, and C. A. Reynolds, Physica (to be published).

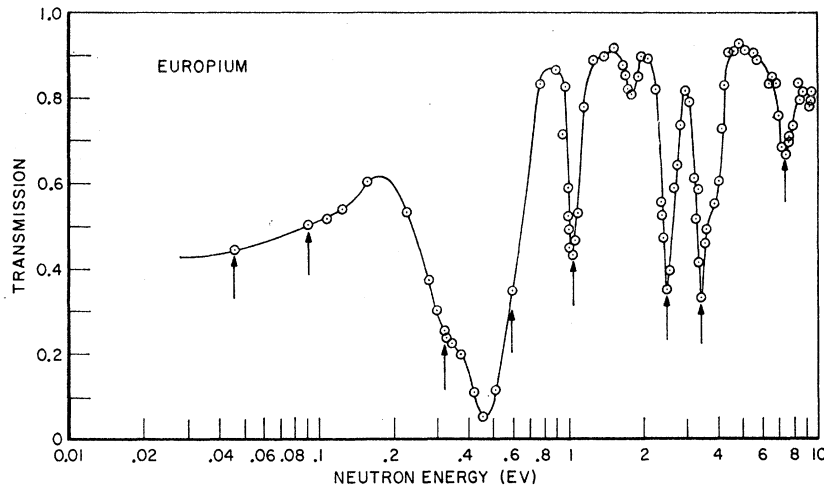


FIG. 2. Transmission of Eu_2O_3 powder sample with 6.80×10^{20} Eu atoms/cm² versus neutron energy. Data on resonance spin states were taken at the positions shown by arrows.

but instead has a high paramagnetic susceptibility. In addition, neutron diffraction investigations³⁵ have shown that europium metal is antiferromagnetic at low temperature with a helical spin structure. The paramagnetism is large enough, however, to produce a measurable nuclear polarization opposite to the external field. With this in mind, we have made the spin assignments listed in Table V. Unfortunately, we cannot use the method of Postma *et al.*²⁸ to make an independent determination of the absolute spins, since the 3.37-eV resonance was the only one found with $J = I - \frac{1}{2}$, and there is too much interference here from resonances of unknown spin. The spin assignment for the negative level is based upon the data taken at 0.046 eV, where a thin Cd filter was used to reduce the second-order contamination. Using the resonance parameters given by Tassan *et al.*,⁵ the contributions of the 0.32- and 0.46-eV resonances are computed to be 230 and 885 b, respectively, so that the negative level contribution is

TABLE IV. Observed percent transmission changes ($\Delta t/t$) upon reversing the neutron-beam polarization for neutron resonances in europium. A positive change means that the transmission was smaller when the neutron spins were parallel to the applied field. Europium metal samples were used.

Neutron energy (eV)	Filter	Sample thickness (cm)	Observed transmission	($\Delta t/t$) in %
0.046	Cd	0.071 ^a	0.028	-2.25 ± 0.25
0.092	None	0.064	0.36	-0.69 ± 0.06
0.32	None	0.089	0.089	-0.66 ± 0.16
0.32	Rh	0.018 ^a	0.33	-2.14 ± 0.10
0.60	None	0.089	0.166	-2.27 ± 0.18
0.60	Hf	0.018 ^a	0.66	-0.65 ± 0.14
1.06	Cd	0.089	0.174	-2.53 ± 0.14
2.46	Cd	0.089	0.170	-0.73 ± 0.13
3.37	Cd	0.089	0.171	$+0.92 \pm 0.17$
7.30	Cd	0.240	0.36	-0.19 ± 0.22

^a Copper coated.

³⁵ C. E. Olsen, N. G. Nereson, and G. P. Arnold, *J. Appl. Phys. Suppl.* **33**, 1135 (1962).

about 1280 b, or a little over half of the total elemental cross section. The negative level spin must therefore be the same as for the first two positive levels; otherwise the observed effect would have been more than an order of magnitude smaller and in the reverse direction. Similarly, the spins of the 0.32- and 0.46-eV levels, which overlap, can be seen to be the same since the observed effects at 0.32 and 0.60 eV (which are equal energy intervals from the large resonance at 0.46 eV) go in the same direction. Note from Table IV how the use of filters improves the data. It is worth examining the data taken with the 0.018-cm-thick sample in detail. Since this is a rather thin sample, the difference between cross sections computed using Eq. (5) and those obtained from the observed transmissions is not important in this energy range. We first correct the observed transmissions for the presence of the copper coating on this sample, which has a transmission of 0.92. We thus obtain 2780 and 910 b for the observed cross sections at 0.32 and 0.60 eV, respectively. The ratio of these cross sections is 3.06, which is quite close to the ratio of the observed effects, 3.30, as expected for equal spins. The 1.06- and 2.46-eV resonances are well separated from other resonances, so these present no difficulties. At

TABLE V. Spin assignments for resonances in europium. Also listed are the total radiation widths and the relative isomeric activation ratios.

Resonance energy (eV)	Isotope	J	Γ_γ (eV)	R
Negative	151	3	0.067 ^a	0.24 ± 0.03^d
0.32	151	3	0.0795 ± 0.002^b	0.19 ± 0.02^d
0.46	151	3	0.087 ± 0.002^b	0.11 ± 0.03^d
1.06	151	3	0.085 ± 0.003^c	0.08 ± 0.03^d
2.46	153	3	0.092 ± 0.002^c	
3.37	151	2	0.092 ± 0.003^c	$\sim 0.06^d$

^a N. Holt, *Phys. Rev.* **98**, 1162(A) (1955).

^b See Ref. 5.

^c See Ref. 4.

^d See Ref. 40.

3.37 eV there are several contributing resonances, but the observed effect is due primarily to the 3.37-eV resonance (the effect of the 3.29-eV resonance in Eu¹⁵³ is small because of the smaller magnetic moment of this isotope). It seems likely that the absence of a real effect at 7.30 eV indicates that the unresolved levels at 7.24 and 7.47 eV have different spins. Using the unresolved resonance parameters given by Sailor *et al.*³⁶ to make the necessary resolution and Doppler calculations, we find that we would expect to see a -0.74% effect if both spins were $I+\frac{1}{2}$, or $+1.04\%$ if both spins are $I-\frac{1}{2}$.

From the data on the 0.32-, 0.46-, and 1.06-eV resonances which were analyzed to account for the effects of resolution and second order, we obtain an average nuclear polarization $f_N = [1.6 \pm 0.4]\%$ at an average temperature of 0.085°K. We cannot compute the hyperfine structure constant because of our lack of knowledge of the degree of magnetic saturation of the electron spins f_e . However, we can obtain an approximate value of f_e from Eq. (3) by assuming $H_e = -50$ kOe⁴⁴ and using the double resonance determination $A = -100$ Mc/sec.³¹ We thus obtain $f_e = 10.7\%$ in an external field of 18 kOe. Recently, the hyperfine field at the nucleus has been measured by a Mössbauer technique³⁷ to yield $H_{hf} = 264$ kOe. If we use this instead, we obtain $f_e = 13.1\%$. The uncertainty in these values is large ($\sim 50\%$), but in any case, f_e appears to be rather small.

In Table V we have listed, along with our spin assignments, measurements of total radiation widths and relative isomeric activation ratios (the ratio of the absorption cross section for the 9.3-h isomeric state to the total absorption cross section). Since both Γ_γ and R seem to fall roughly into two groups, it seems reasonable that these would be associated with different resonance spin states.³ However, our spin measurements show that no such correlation exists, which is somewhat surprising. Huizenga and Vandenbosch³⁸ have computed values of the isomeric activation ratio with a step-by-step calculation, assuming that after each cascade γ emission, there are many spin states available for the next dipole transition. Using the level-density distribution of the free-gas model³⁹ and assuming reasonable values of the parameters involved, they obtain values of R which are strongly dependent on the spin of the initial compound-nuclear state. A comparison of their computations for Eu¹⁵¹ with Wood's experimental data⁴⁰ seems to favor $J=2$ for the negative and 0.32-eV resonances, and $J=3$ for the 0.46- and 1.06-eV resonances. Keisch⁴¹ has measured the isomeric activation ratio in the thermal and epi-cadmium regions and has used the model of Huizenga and Vandenbosch to indi-

cate that one or more of the positive energy levels in the 0.3–1.1-eV range has $J=I-\frac{1}{2}$. These results are not compatible with our spin assignments. Measurements have also been made on the low-energy capture γ -ray intensities⁴² and multiplicities⁴³ associated with the 0.46- and 1.06-eV resonances; no significant differences were found.

CONCLUSIONS

There appears to be no correlation between the compound-nuclear spin states and either the isomeric activation ratios or the total radiation widths. Although such a correlation was found⁶ in the case of In¹¹⁵, any spin assignments^{38,41,44} made on the basis of the isomeric activation ratio alone are questionable. The situation seems to be more complicated than the simple model of Huizenga and Vandenbosch indicates. The spin of a compound-nuclear level undoubtedly influences how it will decay and populate an isomeric level. However, its influence is obscured by the large fluctuations in transition probabilities (partial radiation widths) which have been observed⁴⁵ between resonances, even if their spins are the same, corresponding to a small number of degrees of freedom ($\nu \approx 1$) in a Porter-Thomas distribution.

As to the distribution of the spin states, there are many more $I+\frac{1}{2}$ levels than $I-\frac{1}{2}$ levels, in agreement with Sailor's observation.⁴⁶ We have found 4 out of 5 resonances in Gd, and 5 out of 6 resonances in Eu with $J=I+\frac{1}{2}$. Although the statistical sampling is poor, these results are not inconsistent with a $2J+1$ dependence of the level density.³⁹

With respect to the magnetic properties of these two metals, our results are consistent with the assumption that Gd is magnetically saturated under the conditions of this experiment, so that we obtain good agreement with the electron-spin-resonance results for the hyperfine structure constants. The effective field at the nucleus was shown to be negative. The results for Eu indicate that it is antiferromagnetic down to temperatures a little below 0.1°K; if it became ferromagnetic,³⁷ we would have observed much larger effects than we did. It would be interesting to examine the magnetic properties of some of the newly discovered ferromagnetic europium compounds⁴⁷ with this technique.

{*Note added in proof.* Recent determinations of resonance spins in gadolinium by the groups at Brookhaven [F. J. Shore, V. L. Sailor, G. Brunhart, and C. A. Reynolds, *Bull. Am. Phys. Soc.* **9**, 21 (1964)], and at Mol [F. Poortmans and H. Ceulemans, *Bull. Am. Phys. Soc.* **9**, 178 (1964)] are in complete agreement with ours.}

³⁶ V. L. Sailor, H. H. Landon, and H. L. Foote, *Phys. Rev.* **93**, 1292 (1954).

³⁷ P. H. Barrett and D. A. Shirley, *Phys. Rev.* **131**, 123 (1963).

³⁸ J. R. Huizenga and R. Vandenbosch, *Phys. Rev.* **120**, 1305 (1960).

³⁹ H. A. Bethe, *Rev. Mod. Phys.* **9**, 69 (1939).

⁴⁰ R. E. Wood, *Phys. Rev.* **95**, 453 (1954).

⁴¹ B. Keisch, *Phys. Rev.* **129**, 769 (1963).

⁴² J. E. Draper and A. A. Fleischer, *Nucl. Phys.* **13**, 53 (1959).

⁴³ J. E. Draper and T. E. Springer, *Nucl. Phys.* **16**, 27 (1960).

⁴⁴ J. Rondio and Z. Wilhelm, *Acta Phys. Polon.* **23**, 221 (1963).

⁴⁵ L. M. Bollinger, J. P. Coté, and T. J. Kennett, *Phys. Rev. Letters* **3**, 376 (1959); L. M. Bollinger, R. E. Coté, and J. P. Marion, *Bull. Am. Phys. Soc.* **6**, 274 (1961).

⁴⁶ V. L. Sailor, *Phys. Rev.* **104**, 736 (1956).

⁴⁷ M. W. Shafer, T. R. McGuire, and J. C. Suits, *Phys. Rev. Letters* **11**, 251 (1963), and references given here.

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Dissociation of $\text{Li}^{6\dagger}$

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Interactions of Li^6 ions with carbon and nickel targets have been investigated at incident Li^6 energies of 36 and 63 MeV, utilizing a $(dE/dx) \times E$ product identification system capable of separating individual product isotopes. Contrary to expectations based on previous studies in this laboratory on the $\text{Li}^6(\text{Li}^6, d)\text{B}^{10}$ reaction at 6 MeV which demonstrated a direct reaction mechanism involving the transfer of an alpha particle, no evidence was obtained in the experiments reported herein for deuteron groups corresponding to population of isolated residual states. Each deuteron energy spectrum exhibited a single broad peak, centered at an energy corresponding to the beam velocity, indicating that a direct dissociation mechanism dominates lithium interactions at these higher energies, thus precluding use of Li^6 ions at high energies as nuclear spectroscopic probes. The total dissociation cross section for 63-MeV Li^6 ions on carbon, for example, was found to be 24% of the total geometric cross section. In order to establish whether the Li^6 dissociation proceeds sequentially through well-defined excited states, the elastic and inelastic scattering both of a C^{12} beam from a Li^6 target, and of a Li^6 beam from a carbon target, were studied. These data demonstrate that processes wherein binary dissociation follows inelastic excitation of unbound Li^6 states can account for less than 5% of the observed events. It is concluded that the dissociation mechanism is a direct one, reflecting strong alpha-particle plus deuteron-cluster amplitudes in the Li^6 wave function. Analysis of the dissociation product angular distributions suggests that the dominant interaction involved in these studies is nuclear scattering of the center of mass of the Li^6 ion from the target. Preliminary studies on the dissociation of Li^7 and of B^{10} and B^{11} have also been carried out; in each case, an alpha particle is again a dominant dissociation product.

I. INTRODUCTION

EXPERIMENTS¹⁻⁶ carried out using Li^6 ion beams at low incident energies have provided convincing evidence that Li^6 has a well-developed alpha-particle plus deuteron-cluster structure. It has been generally assumed that this would have important consequences concerning the possible utility of lithium ion induced reactions in the investigation of nuclear structure.

Stripping reactions, induced by these ions, in which the alpha particle or the deuteron is transferred to the target nucleus, offer a valuable and perhaps unique probe for the experimental determination of alpha- or deuteron-reduced widths of bound nuclear states, which cannot be obtained directly from resonant scattering measurements.

One of the initial objectives of the experimental studies reported herein was the measurement of the alpha-particle reduced width for the 7.12-MeV, 1^- state in O^{16} . This state is bound, lying 42 keV below the binding energy of an alpha particle in O^{16} , and can be formed by p -wave capture of an alpha particle by C^{12} . The alpha-particle width of this state is of vital importance in nucleogenesis studies,⁷ since it is the single important parameter that remains unknown in the calculation of the helium burning process in stellar interiors. On the basis of earlier studies² in this laboratory and elsewhere¹ on the $\text{Li}^6(\text{Li}^6, d)\text{B}^{10}$ and $\text{Li}^7(\text{Li}^6, t)\text{B}^{10}$ reactions, which were shown to proceed through direct transfer of an alpha particle, it was hoped that study of the $\text{C}^{12}(\text{Li}^6, d)\text{O}^{16}$ reaction would provide a direct determination of this and other reduced widths in O^{16} . It was further anticipated that use of higher energy Li^6 ions

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¹ G. C. Morrison, Phys. Rev. Letters **5**, 565 (1960); see also *Direct Interactions and Nuclear Reaction Mechanisms*, edited by E. Clementel and C. Villi (Gordon and Breach Publishers, New York, 1963).

² D. A. Bromley, K. Nagatani, L. C. Northcliffe, R. Ollerhead, and A. R. Quinton, *Proceedings of the Rutherford Jubilee International Conference*, edited by J. B. Birks (Heywood and Company, Ltd., 1962), p. 597.

³ M. N. Huberman, M. Kamegai, and G. C. Morrison, Phys. Rev. **129**, 791, (1963).

⁴ Pham-Dinh-Lien and L. Marquez, Nucl. Phys. **33**, 202 (1962).

⁵ R. K. Hobbie and F. F. Forbes, Phys. Rev. **126**, 2137 (1962).

⁶ J. M. Blair and R. K. Hobbie, Phys. Rev. **128**, 2282 (1962).

⁷ A. G. W. Cameron (private communication).