

Spins of Neutron Resonances and the Hyperfine Coupling Constant in Gadolinium Metal*

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The spins of several resonances in the neutron cross section of gadolinium have been determined from measurements with polarized neutrons and polarized nuclei. Use was made of the difference in the statistical weighting factors to make the spin determinations absolute, i.e., independent of the sign of the nuclear hyperfine interaction or of any assumed directions.

Isotope	Neutron energy (eV)	J
155	0.0268	2
157	0.0314	2
155	2.008	1
155	2.568	2
157	2.825	2

Assuming $\mu^{155}/\mu^{157} = +0.750$, the magnitude of the nuclear magnetic hyperfine interaction is determined to be $A/k = (8.0 \pm 0.8) \times 10^{-4} \text{K}$ for Gd^{157} and $(6.0 \pm 0.6) \times 10^{-4} \text{K}$ for Gd^{155} . If we assume the nuclear magnetic moment of Gd^{157} to be -0.33 nm we obtain a hyperfine magnetic field at the nucleus, $H_{\text{hf}} = -(3.48 \pm 0.34) \times 10^6 \text{ Oe}$.

I. USE OF STATISTICAL WEIGHTING FACTOR FOR ABSOLUTE DETERMINATION OF THE SPIN OF THE NUCLEAR COMPOUND STATE

A RESONANCE in the slow-neutron cross section corresponds to formation of the compound nucleus in an excited state. Such states have a definite total angular momentum J . At low neutron energies the interactions are of the s -wave type; hence the spin of the excited state is limited to $J = I \pm \frac{1}{2}$ where I is the spin of the target nucleus. In recent years many resonances have been studied by measuring the transmission of polarized monochromatic neutrons through polarized targets.¹⁻³ Such measurements yield, in a rather direct way, the spin of the compound state *provided that the sign of the nuclear polarization is known*. Often, however, the sign is subject to some uncertainty, particularly when the nuclear polarization is the consequence of nuclear hyperfine interaction. The doubtful cases can sometimes be of special interest since the uncertainty stems from peculiarities in the electronic wave functions. Such an example is the Gd^{3+} ion which has an $^8S_{7/2}$ ground-state configuration. For this state, the magnetic hyperfine interaction arises from distortions of the atomic core as proposed by Sternheimer.⁴ Electrostatic interactions with the outer electrons distort the wave functions of the core electrons. If the spin density of the outer electrons is not paired there results a cor-

responding unpaired spin density at the nucleus which couples with the nuclear magnetic moment. Since Sternheimer's original contribution, this idea has been found to have interesting applications to ferromagnetic substances.^{5,6} Although some calculations have been made⁷ for Gd^{3+} , the details of the wave functions are not known with sufficient accuracy to yield reliable theoretical values of either the sign or the magnitude of the magnetic hyperfine interaction.

In a recent paper, Postma *et al.*⁸ have shown that spin determinations with polarized nuclei and neutrons can be absolute *provided that the cross section contains at least two observable resonances of opposite spin states*. This method takes advantage of the fact that in the equations for the transmission effect, a statistical weighting factor⁹ appears which has two different magnitudes depending on whether $J = I + \frac{1}{2}$ or $I - \frac{1}{2}$. Recent data of Stolovy¹⁰ and the data to be presented here¹¹ show that the gadolinium resonances satisfy this condition. In fact, Stolovy made use of Postma's absolute method in making part of his spin assignments.

In this paper we shall present an extension of the weighting-factor method to the case of overlapping resonances, and derive a set of equations suitable for general application. In contrast to Stolovy's work, which is partly based on estimated magnetic fields, all our assign-

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¹ L. D. Roberts, S. Bernstein, J. W. T. Dabbs, and C. P. Stanford, Phys. Rev. **95**, 105 (1954); J. W. T. Dabbs, L. D. Roberts, and S. Bernstein, *ibid.* **98**, 1512 (1955).

² A. Stolovy, Phys. Rev. **118**, 211 (1960).

³ Hans Postma, H. Marshak, V. L. Sailor, F. J. Shore, and C. A. Reynolds, Phys. Rev. **126**, 979 (1962).

⁴ R. M. Sternheimer, Phys. Rev. **86**, 316 (1952); **95**, 436 (1954); **105**, 158 (1957).

⁵ W. Marshall, Phys. Rev. **110**, 1280 (1958).

⁶ R. E. Watson and A. J. Freeman, Phys. Rev. **123**, 2027 (1961).

⁷ R. E. Watson, and A. J. Freeman, Phys. Rev. Letters **6**, 277 (1961); **6**, 388 (E) (1961).

⁸ H. Postma, F. J. Shore, and C. A. Reynolds, Physica **30**, 713 (1964).

⁹ Note that this is not the same statistical weighting factor that appears in the Breit-Wigner formula.

¹⁰ A. Stolovy, Phys. Rev. **134**, B68 (1964).

¹¹ Preliminary results were presented previously; see F. J. Shore, V. L. Sailor, G. Brunhart, C. A. Reynolds, Bull. Am. Phys. Soc. **9**, 21 (1964).

ments are based on the internal consistency of our experimental data.

The neutron cross section of gadolinium contains many closely spaced resonances occurring mostly in Gd¹⁵⁵ and Gd¹⁵⁷. Many of these have been carefully analyzed and isotopic identification made.^{12,13} We have used the resonance parameters of Møller *et al.*¹³ Our calculations differ materially from those of Stolovy since he did not include the effect of the resonance at 2.825 eV.

II. THE TRANSMISSION OF POLARIZED NEUTRONS THROUGH POLARIZED TARGETS

A. Definition of Transmission Effect

In measurements involving polarized neutrons and polarized nuclei, a convenient quantity, the transmission effect \mathcal{E} , may be defined³

$$\mathcal{E} \equiv \frac{\mathcal{T}_P - \mathcal{T}_A}{\mathcal{T}_P + \mathcal{T}_A} = \frac{C_P - C_A}{C_P + C_A - 2B}, \quad (1)$$

where \mathcal{T} is the transmission of the sample, the subscripts P and A refer to the situations in which the neutron and nuclei are, respectively, parallel or antiparallel, C is the counting rate of the transmitted beam, and B is the background counting rate.

Let us first consider the simple situation which meets the following conditions: (1) the target is monoisotopic; (2) the cross section is entirely due to one level in the compound nucleus; (3) the neutron beam is perfectly monoenergetic; and (4) the nuclear polarization of the target is small. In this situation the transmission effect is related to the polarization of the neutron beam f_n^0 and the polarization of the target nuclei f as follows:

$$\mathcal{E} \approx -f_n^0 \tanh(N\sigma\rho ft). \quad (2)$$

In the equation, N is the density of the target nuclei, t the target thickness, and σ the total cross section. The statistical weighting factor ρ arises from the Clebsch-Gordan coefficients involved in combining the angular momenta of the target nucleus and the incident neutron. For s -wave interactions, ρ has the two possible values $\rho = I/(I+1)$ if $J = I + \frac{1}{2}$, or $\rho = -1$ if $J = I - \frac{1}{2}$. In certain cases it may be possible to distinguish J as a result of an experimental determination of \mathcal{E} because of the substantial difference in the magnitude of ρ for the two possibilities. Thus, for the simple case meeting the four conditions listed above, the sign of the product $f\rho$ is immediately determined from the sign of the measured \mathcal{E} . If, *in addition*, the absolute sign of f is known, the sign of \mathcal{E} determines whether the cross section is due to a $J = I + \frac{1}{2}$ or a $J = I - \frac{1}{2}$ interaction. However, the work on terbium⁸ involving well-separated resonances showed that the choice of $I + \frac{1}{2}$ or $I - \frac{1}{2}$ requires no knowledge of

¹² F. B. Simpson and R. G. Fluharty, *Bull. Am. Phys. Soc.* **2**, 42 (1957).

¹³ H. Bjerrum-Møller, F. J. Shore, and V. L. Sailor, *Nucl. Sci. Eng.* **8**, 183 (1960).

the sign of μ nor of H if there are two resonances present which are of opposite spin in the same isotope.

B. Several Resonances and Several Isotopes with Monoenergetic Neutrons

Often more than one isotope is present in the target, and also two or more resonances may overlap so that the observed cross section is a mixture of two spin states of the compound nuclei. In this case the net \mathcal{E} is obtained from an algebraic summation over the contributing resonances and isotopes.

To analyze the problem, assume we have isotopes $1, \dots, \alpha, \dots$ present in the sample, and that each isotope can have $1, \dots, i, \dots$ resonances which contribute materially to the measured effect. Let the beam of monoenergetic neutrons contain, respectively, w_+ and w_- neutrons oriented with their spins parallel and antiparallel to an applied magnetic field. The beam will be depleted in passing through the sample thickness dt according to the following simultaneous differential equations:

$$\begin{aligned} dw_+ &= [-w_+ \sum_{i,\alpha} F_\alpha N \sigma_{i,\alpha} (1 + \rho_{i,\alpha} f_\alpha) - D(w_+ - w_-)] dt, \\ dw_- &= [-w_- \sum_{i,\alpha} F_\alpha N \sigma_{i,\alpha} (1 - \rho_{i,\alpha} f_\alpha) + D(w_+ - w_-)] dt. \end{aligned} \quad (3)$$

These and the following equations are straightforward extensions of the equations for a single resonance.⁸ The sample contains N target nuclei per cm³, and of these a fraction F_α consists of isotope α so that $N_\alpha = F_\alpha N$. The nuclear cross section $\sigma_{i,\alpha}$ is the appropriate total cross section for isotope α due to resonance i ; $\rho_{i,\alpha}$ is the weighting factor for the resonance; f_α is the polarization of nuclei of type α and D^{-1} is the mean free path for spin reversal of the neutrons within the sample of non-nuclear origin.

It is convenient to refer the nuclear polarization for all the isotopes of one target element to that for one of the isotopes f_β , since the effective magnetic field at the nuclei of the various isotopes of the element will be practically identical. Accordingly let $f_\alpha = r_\alpha f_\beta$. For a saturated ferromagnetic sample the factor r_α is the ratio of the Brillouin functions for the nuclei α and β . In the case of very small nuclear polarizations in a non-saturated ferromagnetic sample,

$$f_\alpha = \frac{M}{M_\infty} B_I \approx \frac{M}{M_\infty} \frac{I_\alpha + 1}{I_\alpha} \frac{\mu_\alpha H_e}{3kT}, \quad (4)$$

where M is the magnetization of the sample, M_∞ the saturation magnetization, B_I is the Brillouin function, μ_α is the nuclear magnetic moment of isotope α , H_e is the effective magnetic field at the nucleus, k is Boltzmann's constant, and T is the absolute temperature. In the present case all isotopes of the same element are assumed to have the same H_e and T , and M is uniform

throughout. Therefore,

$$r_\alpha \equiv \frac{f_\alpha}{f_\beta} = \frac{\mu_\alpha I_\alpha + 1}{\mu_\beta I_\beta + 1} \frac{I_\beta}{I_\alpha}. \quad (5)$$

The solution of Eq. (3) when substituted into Eq. (1) gives, for the transmission effect,

$$\mathcal{E} = \frac{-\frac{1}{2}(1+\varphi)f_n^0\tau \sinh\kappa t}{\cosh\kappa\tau - (\frac{1}{2}(1-\varphi)\tau f_n^0 - \nu) \sinh\kappa t}, \quad (6)$$

where f_n^0 is the polarization of the incident neutron beam,

$$\kappa = [(Nf_\beta \sum_{i,\alpha} F_\alpha r_{\alpha\rho i,\alpha} \sigma_{i,\alpha})^2 + D^2]^{1/2},$$

$$\tau = (Nf_\beta \sum_{i,\alpha} F_\alpha r_{\alpha\rho i,\alpha})/\kappa, \quad \nu = D/\kappa,$$

and φ is the efficiency for reversing the sense of the neutron spin.

C. Other Experimental Factors

Before comparison is made with measured values, the transmission effect \mathcal{E} , as derived in Eq. (6), must be modified to take account of several experimental factors. Among these are (1) the neutron energy spread in the spectrometer beam, (2) corrections for second-order contamination of the neutron beam, (3) Doppler broadening of resonances, (4) depolarization of the neutron beam, (3) Doppler broadening of resonances, (4) depolarization of the neutron beam by non-nuclear processes, and (5) changes in the temperature of the sample during a counting period. Some of these effects have been discussed previously,^{3,8} and need only a few special comments here.

The observed transmission effect $\langle \mathcal{E} \rangle$ is a consequence of convoluting the spectrometer resolution function, $R(E-E')$, with the solutions of Eq. (3) which lead to Eq. (6). This operation gives the following result:

$$\langle \mathcal{E} \rangle = \frac{-\frac{1}{2}(1+\varphi)f_n^0 \int_0^\infty R(E-E')\tau \exp(-\delta t) \sinh\kappa t dE'}{\int_0^\infty R(E-E') \exp(-\delta t) \{ \cosh\kappa t - [\frac{1}{2}(1-\varphi)\tau f_n^0 - \nu] \sinh\kappa t \} dE'}, \quad (7)$$

where the symbol $\langle \mathcal{E} \rangle$ is to be identified with a *measured* value of the transmission effect, and $\delta = N \sum_{i,\alpha} F_\alpha \sigma_{i,\alpha}$.

The correction for second-order contamination does not appear explicitly, but is well understood and is made routinely on the raw data.⁸

Wherever σ appears in Eq. (7) it is understood to be an energy-dependent cross section given by an appropriate Doppler-broadened, single-level formula.¹⁴ The treatment of Doppler broadening at low temperatures requires the use of an effective rather than actual temperature to correct for the effects of crystalline binding. The effective temperature, $T_{\text{eff}} \approx \frac{3}{8}\Theta_D$, where Θ_D is the characteristic Debye temperature.¹⁵ More complicated corrections for crystal binding do not appear necessary in this application.¹⁶

A polarized neutron beam passing through a magnetic medium will be partially depolarized. This depolarization results primarily from misoriented magnetic domains in the target, and disappears as the sample becomes magnetically saturated. Equations to account for this type of depolarization have been presented pre-

viously,¹⁷ and it is accounted for in Eq. (3) by the terms including D . In order to make this correction, the polarization of the transmitted beam must be measured as a function of neutron energy.

In the case of the gadolinium samples, a new type of beam depolarization has been noted which is energy-independent. We attribute this to magnetic field gradients at the front and back surfaces of the target. The target is in the form of a slab held between the pole pieces of the target polarizing magnet. Since the target is supported inside the cryostat which fits through the magnet gap, several mm of air gap exist in the magnetic circuit, producing indeterminate gradients in the vicinity of the sample. If the sample is symmetrically positioned, half of the depolarization caused by external field gradients occurs before the neutrons enter the target and half after they emerge. Only the first half affects the measured transmission effect by reducing the polarization of the neutron beam incident on the target. Thus the polarization of the *incident beam* is $(f_n'/f_n^0)^{1/2}f_n^0$ where f_n' is the measured beam polarization after passage through the target, while f_n^0 is the beam polarization with no target present.

In order to show how $\langle \mathcal{E} \rangle$ depends upon the temperature of the sample, we consider the case where, in Eq. (7), $f_\beta \ll 1$, for which the approximation $\sinh\kappa t \approx \kappa t$ is

¹⁴ A. Bernabei, L. B. Borst, and V. L. Sailor, Nucl. Sci. Eng. 12, 63 (1962).

¹⁵ J. E. Lynn, and E. R. Rae, J. Nucl. Energy 4, 418 (1957).

¹⁶ A. Bernabei, Brookhaven National Laboratory No. BAL860 (T344), 1964 (unpublished).

¹⁷ See Ref. 3, Sec. IVB.

valid, and assuming $D=0$. Then,

$$\langle \mathcal{E} \rangle \simeq -\frac{1}{2}(1+\varphi)f_n^0 f_\beta Nt \sum_{i,\alpha} \frac{\int_0^\infty R(E-E') F_{\alpha} r_{\alpha} \rho_{i,\alpha} \sigma_{i,\alpha} \exp(-\delta t) dE'}{\int_0^\infty R(E-E') \exp(-\delta t) \{ \cosh \kappa t - [\frac{1}{2}(1-\varphi)\tau f_n^0 - \nu] \sinh \kappa t \} dE'}. \quad (8)$$

Calling

$$\langle \sigma_{i,\alpha} \rangle \equiv \frac{\int_0^\infty R(E-E') \sigma_{i,\alpha} \exp(-\delta t) dE'}{\int_0^\infty R(E-E') \exp(-\delta t) \{ \cosh \kappa t - [\frac{1}{2}(1-\varphi)\tau f_n^0 - 1] \sinh \kappa t \} dE'}, \quad (9)$$

then

$$\langle \mathcal{E} \rangle \simeq -\frac{1}{2}(1+\varphi)f_n^0 f_\beta Nt \sum_{i,\alpha} F_{\alpha} r_{\alpha} \rho_{i,\alpha} \langle \sigma_{i,\alpha} \rangle. \quad (10)$$

From (4) and (10),

$$\langle \mathcal{E} \rangle \simeq -\frac{1}{2}(1+\varphi)f_n^0 \frac{M}{M_\infty} \frac{I_\beta + 1}{I_\beta} \frac{\mu_\beta H_e}{3kT} \times Nt \sum_{i,\alpha} F_{\alpha} r_{\alpha} \rho_{i,\alpha} \langle \sigma_{i,\alpha} \rangle. \quad (11)$$

When the external magnetic field at the sample is held constant, at a given setting of the spectrometer energy, then it is seen that $\langle \mathcal{E} \rangle T \simeq \text{constant}$. This fact is utilized in treating the raw data, since data obtained with different, but known, temperatures can be grouped together to enhance the over-all statistical accuracy.

D. Computation of $\langle \mathcal{E} \rangle$ versus Neutron Energy

No practical method exists for unfolding the Doppler broadening and the resolution smearing. The only reasonable method for comparing experiment with theory is to insert the various parameters into the convoluted formulas and to generate families of computed curves which can then be compared with observed data. A computer program has been written to cover the general case of several isotopes, several overlapping Doppler-broadened resonances, correction for domain depolarization, and convolution of the spectrometer resolution function. The program computes $\langle \mathcal{E} \rangle$ as a function of neutron energy using Eq. (8). The input data include sample thickness, isotopic abundances, parameters of resonances, beam polarization, spectrometer resolution function, nuclear polarization f_α and the $\rho_{i,\alpha}$. In the usual situation all quantities are accurately known except for f_α and the $\rho_{i,\alpha}$. These latter are treated as variables, and families of curves are generated for comparison with the experimental curves. If $f_\alpha \ll 1$, the computed $\langle \mathcal{E} \rangle$ is linear with f_α . Since each resonance has two possible values of $\rho_{i,\alpha}$, there will be 2^K combinations of the $\rho_{i,\alpha}$ where K is the number of resonances involved. Actually the number of cases which must be computed

is considerably less, because at any energy the result is sensitive only to nearby resonances. If a unique set of $\rho_{i,\alpha}$ can be selected from comparison of the computed and measured curves, the spin assignment of each resonance is unique and absolute, as is also the assignment of the absolute sign of f_α .

When the proper set of $\rho_{i,\alpha}$ has been selected, the normalization of the computed curve to the measured curve gives the absolute magnitude of the nuclear polarization f_α . This in turn yields the magnitude of the magnetic hyperfine interaction. From the magnitude and sign of the hyperfine interaction and the nuclear magnetic moment, the magnitude and sign of the hyperfine field may be obtained.

III. EXPERIMENTAL RESULTS

A. Transmission

In addition to measuring and calculating the transmission effect $\langle \mathcal{E} \rangle$, it is also useful⁸ to measure and compute the transmission $\langle \mathcal{T} \rangle$. This serves as a sensitive check to demonstrate that proper values have been used for the resonance parameters, resolution function, etc. The calculated transmission is included in the computer program described above.

The experimental setup has been described earlier.³ The total cross section of gadolinium from 1.5 to 3.0 eV is shown in Fig. 1. Shown in the upper curve is the computed transmission \mathcal{T} , using the Co(111) reflection with 15-min angular resolution, and also the measured transmission. The small deviations in the region ~ 1.7 to 2.4 eV have not been accounted for, but they introduce minor uncertainties in the computed transmission effects (below). The high point at 1.6 eV was caused by a trace of indium in the solder used in the sample assembly. It is high because of a slight excess of solder at the "open beam" position of the target unit. Calculated and measured transmissions at lower energies are in good agreement. In addition to the data of Fig. 1, transmission measurements were made between 0.3 and 1.5 eV. Several thinner samples of various isotopic enrichments were also measured at 0.0778 eV.

B. Observed Transmission Effects

The transmission effect $\langle \mathcal{E} \rangle$ was measured at many neutron energies as a function of temperature over the range from approximately 0.05 to 0.20°K. Because of the small magnitude of $\langle \mathcal{E} \rangle$ (0 to ~2%) it was necessary to take several data runs at each energy. The individual points for each run were averaged and the standard deviation of the neutron count was used as an indication of the statistical uncertainty of the average value.

Data were also taken at 0.95 and 4.2°K to determine the magnitude of the effect of the spin-dependent magnetic scattering (neutron scattering from the magnetic moments of the atoms). Such scattering is temperature-independent¹⁸ and at such high temperatures the effect due to nuclear polarization was negligibly small. Magnetic scattering is appreciable only at the lowest energies (<0.2 eV).

Data obtained between 1.5 and 3.0 eV are shown in Fig. 2. This region involves the three resonances shown in the cross-section plot of Fig. 1.

C. Comparison with Computed Curves

Families of curves were generated giving $\langle \mathcal{E} \rangle T$ versus E using various combinations of the $\rho_{i,\alpha}$ for the three resonances in the 1.5- to 3.3-eV region. Input data in the computer program included the resonance parameters, spectrometer resolution, Debye temperature,

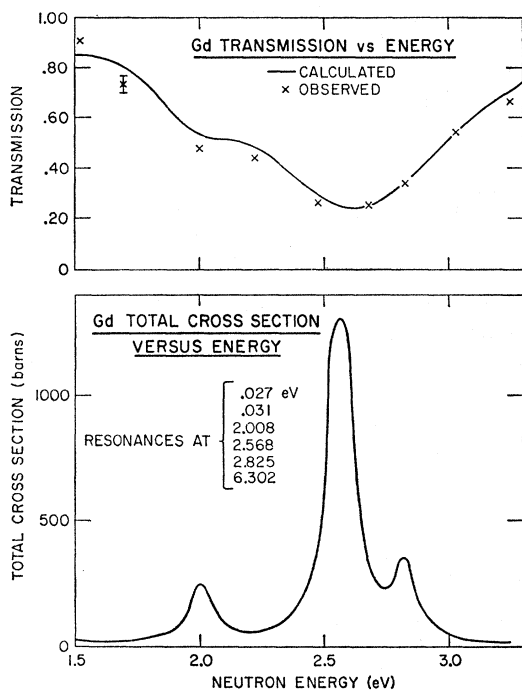


FIG. 1. Neutron cross section of gadolinium between 1.5 and 3.0 eV. The lower curve is calculated with parameters taken from Ref. 13. The upper curve shows the computed and observed transmission curves for the same energy region using the polarization spectrometer.

¹⁸ R. I. Schermer, Phys. Rev. 130, 1909 (1963).

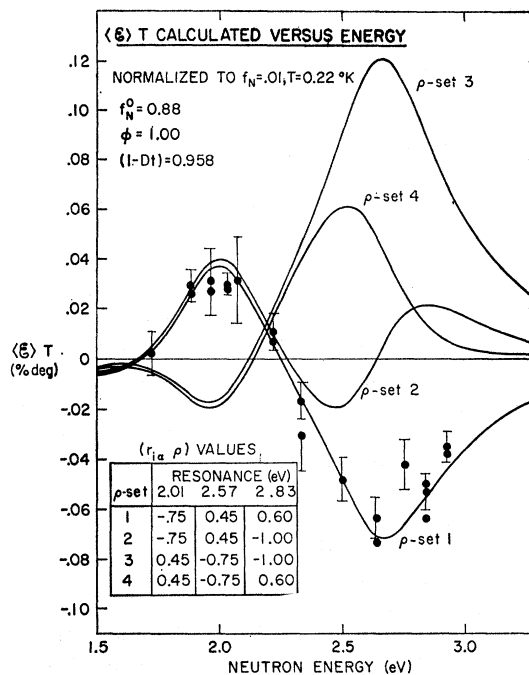


FIG. 2. Experimental and calculated values of $\langle \mathcal{E} \rangle T$ as a function of neutron energy. Each curve corresponds to one possible set of $\rho_{i,\alpha}$. The absolute sign of these curves is arbitrary; i.e., a reflection of the curves about the abscissa would be equally possible depending on the absolute sign of f_{α} . The amplitude is the only other adjustable constant. The points are the experimental values and the error bars reflect the uncertainty due to counting statistics. The curves as shown have already been normalized to " ρ -set 1" which can be seen to be the best choice. In making the normalization only one parameter, the amplitude, can be adjusted. This parameter yields $f_{\alpha} T$. A weighted average of all experimental points was used in making the normalization.

etc. Because of the difference between the magnetic moments of Gd^{155} and Gd^{157} it was necessary to use an appropriate value of r_{α} in the contributing term of each resonance. Examination of Table I, which lists values from the literature for the nuclear magnetic moments

TABLE I. Summary of nuclear magnetic moments of Gd^{155} and Gd^{157} . Values are in nuclear magnetons.

μ^{155}	μ^{157}	μ^{155}/μ^{157}	Method	Reference
-0.19 ± 0.05	-0.33 ± 0.06	0.58^a	Opt. spect.	e
-0.24^b	-0.32^b	0.75 ± 0.07	Par. res.	d
-0.30 ± 0.04	-0.37 ± 0.04	0.80 ± 0.02	Opt. spect.	e
		0.744 ± 0.007	Par. res.	f
-0.25	-0.34	0.73 ± 0.03	Par. res.	g
		0.7495 ± 0.0045	Par. res.	h
-0.32 ± 0.04	-0.40 ± 0.04	0.79 ± 0.02	Opt. spect.	i
		0.763 ± 0.006	Par. res.	j

^a Moments incorrectly calculated on basis of $I=7/2$ instead of $I=3/2$.

^b Depends on μ for Eu^{151} being 3.6 nm.

^c K. Murakawa, Phys. Rev. 96, 1543 (1954).

^d W. Low, Phys. Rev. 103, 1309 (1956).

^e R. D. Speck, Phys. Rev. 101, 1725 (1956).

^f W. Low and D. Shaltiel, J. Phys. Chem. Solids 6, 315 (1958).

^g Mansenkov and Prokhorov, Zh. Eksperim. i Teor. Fiz. 33, 1116 (1957) [English transl.: Soviet Phys.—JETP 6, 860 (1958)].

^h W. Low and D. Shaltiel, Phys. Rev. 115, 424 (1959).

ⁱ N. I. Kaliteevskii, M. P. Chaika, I. Pacheva, and E. E. Fradkin, Zh. Eksperim. i Teor. Fiz. 37, 882 (1959) [English transl.: Soviet Phys.—JETP 10, 629 (1960)].

^j C. F. Hempstead and K. D. Bowers, Phys. Rev. 118, 131 (1960).

TABLE II. Spin assignments of gadolinium resonances.

Resonance energy (eV)	Isotope	$J = I \pm \frac{1}{2}$
0.0268	155	2
0.0314	157	2
2.008	155	1
2.568	155	2
2.825	157	2

shows consistently higher values for the ratio μ^{155}/μ^{157} obtained by optical spectroscopy which measures the free-atom value compared to paramagnetic resonance methods which employ bound atoms. We choose 0.750 for the ratio since we use a metal. This is also the value of r_α , selecting Gd¹⁵⁷ as the standard isotope, since the spins of both Gd¹⁵⁵ and Gd¹⁵⁷ are $\frac{3}{2}$. Account was also taken of the isotopic abundances F_α as required in Eq. (3). Implicit is the assumption that the magnetic field is the same at both nuclei, and that $M = M_\infty$ at the applied field of 16 kOe.¹⁹

The computed curves are also shown in Fig. 2. Since the absolute sign and magnitude of H_e is initially uncertain, the curves as computed, could be reflected about the abscissa in Fig. 2 and the normalization varied. Comparison with the experimental data (plotted points in Fig. 2) permits a unique choice of the proper set of $\rho_{i,\alpha}$ and hence uniquely determines the sign. The amplitude is obtained by normalizing the curve to the experimental points. As can be seen from Fig. 2 the fitted curve is quite satisfactory. It can be seen that the other $\rho_{i,\alpha}$ sets are all definitely excluded since they do not have the proper shape. The other four possible $\rho_{i,\alpha}$ combinations (not shown in Fig. 2), which would place the 2.01- and 2.57-eV resonances in the same spin state, are in much poorer agreement with the data.

D. Spin Assignments

The choice of the proper $\rho_{i,\alpha}$ set in Fig. 2 immediately gives the spin assignments for the 2.008-, 2.568-, and 2.825-eV resonances (Table II). This choice of $\rho_{i,\alpha}$ establishes the absolute sign of f_α ; i.e., the ρ -set 1 shown in Fig. 2 is the correct curve rather than its reflection about the abscissa.

The spin assignment for the two thermal resonances²⁰ at 0.0268 eV (Gd¹⁵⁵) and 0.0314 eV (Gd¹⁵⁷) is less straightforward because of their almost complete overlap. In effect, it is necessary to use the absolute results obtained in the 1.5–3.0-eV region for normalization of data on the thermal resonances. Several factors conspire to reduce the accuracy of the normalization: (1) Account must be taken of the variation with neutron

energy of spectrometer characteristics such as second-order contamination, neutron polarization, beam depolarization, and resolution. Although these factors are known with adequate precision for most purposes, the spectacular variation of the low-energy gadolinium cross section enters in such a way as to amplify the uncertainties. (2) Normalization in the 1.5–3.0-eV region requires a moderately thick sample which in turn makes it necessary to study the thermal resonances on their high-energy tail in the region between 0.2 and 0.4 eV. This taxes the precision of the resonance parameters,¹³ which are called upon to reproduce faithfully the cross-section variation in the tail. (3) In addition, the spin-dependent magnetic scattering is strongly energy-dependent¹⁸ and must be removed from the observed $\langle \mathcal{E} \rangle T$.

The two thermal resonances differ markedly in strength, the Gd¹⁵⁷ resonance being some four times the stronger. As a result the cross section in the region from ~ 0.1 to 0.5 eV is dominated ($\sim 80\%$) by the 0.0314-eV resonance, as is also the transmission effect. It follows that a measurement of the sign of $\langle \mathcal{E} \rangle$ at any energy in this region is sufficient to determine the spin of the stronger resonance. Measurements at 0.22 and 0.357 eV gave negative $\langle \mathcal{E} \rangle$ which requires that $J = I + \frac{1}{2}$ for the 0.0314-eV resonance. This is an unambiguous result.

To determine the spin of the weaker 0.0268-eV Gd¹⁵⁵ resonance, the *magnitude* of $\langle \mathcal{E} \rangle$ must be compared with calculated values based on the two alternative choices, i.e., $J = I + \frac{1}{2}$, or $J = I - \frac{1}{2}$. For this purpose, measurements were made at 0.357 eV, an energy at which the second-order contamination can be removed by means of an indium filter. The calculated values require as input the resonance parameters,¹³ as well as the various experimental constants discussed in preceding sections. Also the calculations are effectively normalized to the 2.0–3.0-eV data, by making use of the absolute value of the nuclear polarization f_α yielded by those data. Results are shown in Table III. As can be seen, the experimental value falls between the two choices, slightly favoring the choice $J = I + \frac{1}{2}$. Although this conclusion agrees with that of Stolovy,¹⁰ the agreement is probably fortuitous since his calculation suffered from the same limitation listed above, and, in addition, he was forced to use independent measurements of the hyperfine constant and make estimates of the internal field.

The indecisiveness of the spin assignment of the

TABLE III. Attempt to determine J for the Gd¹⁵⁵ resonance at 0.0268 eV. The $\langle \mathcal{E} \rangle T$ values are for a neutron energy of 0.357 eV. Because of the much stronger resonance in Gd¹⁵⁷ at 0.0314 eV, the two choices of J yield only small difference in $\langle \mathcal{E} \rangle T$. The $J = I + \frac{1}{2}$ is slightly favored.

$\langle \mathcal{E} \rangle T$ Measured percent	$\langle \mathcal{E} \rangle T$ Calculated $J = I + \frac{1}{2}$	$\langle \mathcal{E} \rangle T$ Calculated $J = I - \frac{1}{2}$
-2.90 ± 0.58	-3.44	-2.08

¹⁹ W. E. Henry, J. Appl. Phys. 29, 524 (1958).

²⁰ It should be noted that a definite, unambiguous assignment for these two resonances is of considerable practical importance to nuclear-engineering calculations and for analysis of capture gamma-ray spectra. Therefore, we have expended much effort in obtaining unambiguous results.

0.0268-eV resonance led us to make a series of special measurements with separated isotopes.²¹ For this purpose three Gd₂O₃ samples were studied, the first composed of the normal isotopic abundances, the second almost pure Gd¹⁵⁵ and the third almost pure Gd¹⁵⁷. Because of unknown magnetic ordering of the Gd₂O₃ at very low temperatures, the mechanism producing the nuclear polarization was not completely understood²²; however, it was assumed that all three samples exhibited the same magnetic behavior. Internal consistency of the data on the three samples strongly support this assumption. Measurements at an energy of 0.0778 eV showed that the transmission effect $\langle \mathcal{E} \rangle$ had the same sign for all three samples. We conclude, therefore, that the 0.0268- and 0.0314-eV resonances have the same J value which is in fact $J = I + \frac{1}{2}$ based on our prior unambiguous assignment of the 0.0314-eV resonance. Thus, the J value of the 0.0268-eV resonance is unambiguously $J = I + \frac{1}{2}$.

E. The Hyperfine Constant for Gadolinium Metal

The magnitude of the transmission effect $\langle \mathcal{E} \rangle$ can be used to determine the nuclear magnetic hyperfine interaction. This is possible because the magnitude of $\langle \mathcal{E} \rangle$ depends on the nuclear polarization which in turn depends on the hyperfine interaction. The normalizing factor between the measured and computed curves (see Fig. 2) yields $f_\beta T$ where f_β is the nuclear polarization of isotope β at temperature T . Although the individual data points show considerable scatter, the value of $f_\beta T$ has good precision since it is the only free parameter in the normalization, and hence all data points contribute to a weighted average. The experimental value of $f_\beta T$ can be inserted in Eq. (4) to obtain $\mu_\beta H_e$. Note that this quantity is not exactly the hyperfine interaction because the effective field at the nucleus H_e includes the effect of the externally applied field.

We must now correct for the effects of the external field. From the J -value assignments in Table II and from the observed negative sign of $\langle \mathcal{E} \rangle$, we conclude that the product $\mu_\beta H_e$ is positive. Since μ_β is negative,²³ H_e must be negative, i.e., opposite to the applied field. Let us assume that the effective field at the nucleus, $H_e = H_l + H_{\text{hf}}$, where H_l is the local field resulting from the external field and H_{hf} is the true hyperfine field re-

TABLE IV. Hyperfine coupling constant for Gd¹⁵⁷.

$A/k(10^{-4} \text{ }^\circ\text{K})$	Sample	Reference
8.0±0.8	Metal	Present work
7.0±1.6	Metal	a
7.64	ThO ₂	b
7.19	BiMg ₃ (NO ₃) ₁₂ ·24H ₂ O	c
7.67	LaCl ₃ ·7D ₂ O	c
7.86	CaWO ₄	d

^a A. Stolovy, Phys. Rev. **134**, B68 (1964).

^b W. Low and D. Shaltiel, J. Phys. Chem. Solids **6**, 315 (1958).

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

^d C. F. Hempstead and K. D. Bowers, Phys. Rev. **118**, 131 (1960).

sulting from the coupling of the conduction and atomic electrons with the nucleus. To a good approximation the local field is given by

$$H_l = H_{\text{ext}} - \mathfrak{D}M + \frac{4}{3}\pi M, \quad (12)$$

where H_{ext} is the external field of 16 kOe, \mathfrak{D} is the demagnetization factor, and M is the saturation magnetization which is taken to be 1990 G. From Osborne's graphs²⁴ \mathfrak{D} is taken to be 1.167 for our 1.10×0.68-in. sample dimension. This gives $H_l = 22 \times 10^3$ Oe.

Our data yield the following results: from normalization for an external field $H_{\text{ext}} = 16$ kOe, $f_\beta T = (2.19 \pm 0.20) \times 10^{-30}$ K, where β refers to Gd¹⁵⁷ which was selected as the "standard" isotope for computing purposes. Using Eq. (4) and the value $\mu^{157} = -0.33$ nm, we obtain $H_e = -(3.26 \pm 0.33) \times 10^5$ Oe. Thus, the hyperfine field $H_{\text{hf}} = -(3.48 \pm 0.34) \times 10^5$ Oe. This can be expressed in terms of the hyperfine coupling constant which is conventionally taken as $A/k = (\mu H_{\text{hf}})/(J I k)$ where $J = \frac{7}{2}$ for the Gd³⁺ ion. Substitution yields $A/k = (8.02 \pm 0.80) \times 10^{-40}$ K.

It is interesting to compare the present value for a metal sample with those obtained by other workers, chiefly with salts. This is done in Table IV in which the coupling constant for Gd¹⁵⁷ is tabulated. We see that the present value agrees within a few percent of those given for ions in different salts. From this it would appear that if the main difference between metallic and nonmetallic samples is due to conduction electrons, their contribution must be a few percent at most.

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²⁴ J. A. Osborne, Phys. Rev. **67**, 351 (1945).

²¹ The enriched isotopes were obtained on loan from the Nuclear Cross Sections Stable Isotopes Pool of the U. S. Atomic Energy Commission.

²² The nuclear polarization in Gd₂O₃ is now undergoing further investigation and results will be described in a future paper.

²³ See references listed in Table I.