TABLE IV. A comparison of the oxygen results to those of other authors.

Author reference	Energy of yield "break" (Mev) Yield threshold	Lower "break"	Upper "break"[b]
7	15.61±0.03	16.03±0.03	(17.13 and 17.18)±0.03
3	···	16.03±0.04	(17.11 and ···)±0.04
14	15.59±0.05	16.14±0.05	(17.15 and 17.21)±0.05
4	15.79±0.03	16.26±?	17.33±?
5	15.73±0.07	16.18±0.05	···
This work[a]	15.77±0.05	16.19±0.04	17.25±0.04

[a] The yield threshold was obtained as described in the text.
[b] The upper "break" is actually a close-spaced doublet which was not resolved in this work, nor, presumably, in the work of reference 4.

at 16.21 Mev (with 24-kev width) and 17.29 Mev (with 90-kev width).[20]

The energy assignments made by Penfold and Spicer (Table III) were to a large extent determined by the value of 15.61±0.01 Mev which they adopted for the $O^{16}(\gamma,n)O^{15}$ threshold, and by their assumption that the threshold for observable yield corresponded to this energy. The most recent value for the oxygen threshold is 50 kev higher,[5] but this is not a big enough change to bring their data into agreement with the present work. When the results of the present work are combined with the data shown in Fig. 3 of reference 7, one gets a value of 15.77±0.05 Mev for the observed $O^{16}(\gamma,n)O^{15}$ threshold. This value does not correspond to the kinematic threshold, but probably indicates an excited state in O^{16} located at about 15.79 Mev.[21] A state at 15.79 Mev has in fact been observed in the $N^{15}(p,\alpha\gamma)C^{12}$ reaction.[20,22] A comparison of the oxygen results obtained in this work with the results of other laboratories is shown in Table IV. As already discussed, the energy assignments made by the authors of reference 7 were largely based on the assumption that the observed yield threshold corresponds with the kinematic threshold. The same criterion was presumably used by the author of reference 3.

On the other hand, the energy assignments made in references 4 and 5 were based on thresholds other than oxygen and it was noted that the yield threshold for oxygen does not correspond to the kinematic threshold. The present experimental results support this contention, and are also in good over-all agreement with references 4 and 5.

The energy assignments made in reference 14 were also based on thresholds other than oxygen and the results for the two break energies are in reasonable agreement with this work and with the work of references 4 and 5. The energy of the observed yield threshold is not in such good agreement, however, and it is even lower than that expected from masses (15.66 Mev).

CONCLUDING REMARKS

It should be emphasized again that the copper threshold which is obtained need not correspond to the kinematic threshold, although it is not expected to be more than about 20 kev from it.

[20] The error on these energy assignments is estimated to be between 10 and 15 kev.
[21] A similar conclusion was drawn by the authors of reference 4.
[22] Lidofsky, Jones, Brent, Weil, Kruse, Barton, and Havens, Jr., Bull. Am. Phys. Soc. Ser. II, 1, 212 (1956).

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Ratios of Relative Abundance, Magnetic Moments, and Capture Cross Sections of Gadolinium Isotopes from Paramagnetic Resonance Spectrum*

W. LOW, *Department of Physics, The Hebrew University, Jerusalem, Israel,*
AND
D. SHALTIEL, *Department of Physics, Israel Institute of Technology, Haifa, Israel*
(Received November 20, 1958)

Precision measurements on the hyperfine structure of the transitions corresponding to $\Delta M=4$ of the cubic-field paramagnetic resonance spectrum of gadolinium in single crystals of thorium oxide yield the following values: isotopic abundance: even isotopes, 69.45%; Gd^{155}, 15.05±0.2%; Gd^{157}, 15.5±0.2%. Ratio of magnetic moments: $\mu^{155}/\mu^{157}=0.7495\pm0.0045$.

Irradiation of crystals at the Harwell pile with a thermal neutron flux of 1.2×10^{12} n/cm^2 sec, and a fast flux of 2.3×10^{11} n/cm^2 sec, yields the ratio of nuclear capture cross sections $\sigma^{157}/\sigma^{155}=2.82$, using the above abundance values.

No *F*-center spectrum was detected in crystals irradiated with a total neutron flux of about 10^{18} n/cm^2. The paramagnetic resonance spectrum of gadolinium in irradiated crystals is unchanged, indicating negligible radiation damage in the neighborhood of the paramagnetic ions.

I. INTRODUCTION

RECENT investigations of the paramagnetic resonance spectrum of trivalent gadolinium in single crystals of thorium oxide revealed a number of interesting features.[1] It was found that the gadolinium takes the place of the thorium ion in the lattice and is surrounded by the cubic crystal field of eight oxygen ions. The over-

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[1] W. Low and D. Shaltiel, J. Phys. Chem. Solids 6, 315 (1958).

all splitting of the $^8S_{7/2}$ ground state is about 0.1755 cm^{-1}, and is sufficiently large so that low-field transitions corresponding to $\Delta M=\pm 3$, ± 4, and ± 5 can be observed with fairly large intensity. The line width of these low-field transitions was found to be less than one gauss along the [100] crystal direction. The unusually narrow lines permitted the detection and recording of the complete hyperfine structure spectrum of the odd gadolinium isotopes 155 and 157 of spin $\frac{3}{2}$.

In the course of these investigations it became clear that these narrow lines would permit to measure a number of nuclear and solid state properties. This paper will report in detail a determination of the isotopic abundance of the ratio of the magnetic moments, and of the ratio of the neutron capture cross sections of the two odd-mass gadolinium isotopes. In fact we have used our paramagnetic resonance spectrometer as a mass spectrometer. To our knowledge this is the time first that a paramagnetic resonance spectrometer has been used in this way.

II. EXPERIMENTAL DETAILS

A standard paramagnetic resonance reflection-type spectrometer has been used. Measurements were made at 3 cm at room temperature.

The difficulties in the precision measurements to be reported below lie in the facts that the abundance of each odd isotope is about 15%. The hyperfine lines are, therefore, only about one twentieth of the intensity of the center lines of the even isotopes. In addition the total extent of the hyperfine splittings is about 17 gauss. Into this range there are crowded 8 hyperfine lines (each isotope of spin $\frac{3}{2}$) and the strong center line. Indeed, had the lines been only slightly wider than one gauss, these precision measurements could not have been possible. It is precisely because of this that the $\Delta M=\pm 4$ transitions were chosen for these measurements rather than the more intense $\Delta M=\pm 1$ transitions. The $\Delta M=\pm 4$ transitions fall in the region of about 800 gauss at 3-cm wavelength. At such low fields our electromagnets are sufficiently homogeneous. Moreover, the magnetic field is fairly stable at these low currents. Therefore, small variations in the current and the remaining inhomogeneity are not contributing factors to the linewidth. Another advantage is that the magnetic field can be varied linearly in this low-field region.

The magnetic field was modulated at 450 cps, the signal amplified, phase-detected, and recorded. The modulation was held to a fraction of the linewidth so as not to cause modulation broadening. The spectrum was scanned for several days under various conditions of amplification, and time constants of the phase-detecting circuit. This was of importance in order to check possible errors in the line shape because of time-constant effects in the detecting circuit. We also checked whether the relative amplitude of the signal depends on the inertia of the recording pen.

TABLE I. Isotopic abundance of gadolinium isotopes in crystals irradiated with neutrons.

No. of measurements	Amplification in mv	Isotopic abundance in % Even isotopes	Gd^{155}	Gd^{157}
3	6	77.3	12.4	10.3
3	3	77.3	12.8	10.8
4	6	77.2	12.5	10.2
5	2	77.2	12.6	10.05

A large number of recordings were made. Only those recordings were considered acceptable in which all derivatives of the absorption lines showed the same shape, and in which the scanning was linear. A set of representative data of the isotopic abundance of gadolinium isotopes in crystals irradiated with neutrons is given in Table I.

Three crystals were irradiated at the Harwell at the BEPO reactor with thermal neutrons of about 5×10^8 neutrons/cm^2 and a fast-neutron flux of 9×10^{17} neutrons/cm^2 over a sample volume of 0.3 cm^3. The sample was intensely radioactive, in part because of radioactive isotopes of gadolinium, and in part because of fission products caused by the fast-neutron flux. A special cavity was used into which the sample was placed. The cavity could be attached to the microwave system in a fraction of a second by means of a special mount. The whole magnet and microwave assembly was heavily shielded by means of lead bricks and the experiments were performed at safe distances. The sample remained strongly radioactive even after several months, and the microwave cavity was too hot to be used again after the experiments were completed.

III. EXPERIMENTAL RESULTS

III.1. Ratio of Magnetic Moments

Since gadolinium has a ground state of $^8S_{7/2}$, a measure of the ratio of the hyperfine structure splittings is of considerable interest. For S-state ions the ratio of the hyperfine structure splittings as measured in this experiment, is expected to differ somewhat from the ratio of the magnetic moments as determined by nuclear magnetic resonance. This difference is usually called the hyperfine anomaly and is associated, in part at least, with structure effects of the nucleus.[2] Our measurements yield the new value, $A^{155}/A^{157}=0.7495\pm 0.0045$, in good agreement with the earlier value of 0.744 ± 0.007 determined from the $\Delta M=\pm 1$ transitions.[1] It is planned to measure μ^{155}/μ^{157} by means of the double-resonance technique.[3]

III.2. Abundance of Odd Gadolinium Isotopes

By carefully measuring the amplitude and the integrated intensity of the two outer pairs of the hyperfine structure components we have been able to measure the abundance of the even and odd isotopes. These results as well as those of previous workers are contained

[2] A. Bohr and V. Weisskopf, Phys. Rev. 77, 94 (1950).
[3] G. Feher, Phys. Rev. 103, 500 (1956).

TABLE II. Isotopic constitution of gadolinium.

Mass	Atomic percentage Even(%)	155(%)	157(%)
Aston[a]	62	21	17
Dempster and Aston[b]	62	21	17
Wahl[c]	67.97	15.61	16.42
Hess[d]	69.51	14.78	15.71
Leland[e]	69.68	14.68±0.15	15.64±0.16
Collins *et al.*[f]	69.2	15.1 ±0.15	15.7 ±0.16
Low and Shaltiel	69.45	15.05±0.2	15.5 ±0.2

[a] F. W. Aston, Proc. Roy. Soc. (London) **A146**, 46 (1934).
[b] A. J. Dempster, Phys. Rev. **53**, 727 (1938).
[c] W. Wahl, Soc. Sci. Fennica, Commentationes Phys.-Math. **11**, 1 (1941).
[d] D. C. Hess, Jr., Phys. Rev. **74**, 773 (1948).
[e] W. T. Leland, Phys. Rev. **77**, 634 (1950).
[f] Collins, Rourke, and White, Phys. Rev. **105**, 196 (1957).

TABLE III. Ratio of cross section of gadolinium isotopes 157/155. Measured abundances for thermal neutron flux 1.2×10^{12} $n/cm^2/sec$ and fast flux 2.3×10^{11} $n/cm^2/sec$: Gd^{157} 10.4%±0.3 and Gd^{155} 12.7%±0.25.

Using normal isotope abundance	Ratio of cross section $\sigma^{157}/\sigma^{155}$
(a) Leland[a]	2.82:1±0.2
(b) Collins *et al.*[b]	2.39:1±0.2
(c) Our value	2.37:1±0.2

[a] See reference 7.
[b] See reference 8.

in Table II. Inspection of the table shows that our results are in fair agreement with those obtained by various workers during the last few years. It has to be pointed out, however, that since ThO_2 crystals are formed at very high temperatures (~3000°C) the isotopic abundance may slightly differ from that found in natural samples.

III.3. Neutron Capture Cross Section

It has been pointed out by Lapp *et al.*[4] that the large cross section of gadolinium is due to the odd gadolinium isotopes. We have measured the abundance of the gadolinium isotopes after 4 weeks' bombardment in the Harwell pile. The new abundances are Gd^{155}:12.7 ±0.25%, and Gd^{157}:10.4±0.3%. The quoted error is the average derivation from the mean value. The ratio of the neutron capture cross sections can be computed with the following assumptions: (a) The neutron cross section of the even isotopes is negligible. (b) The fission products contribute a negligible amount to the gadolinium present in the sample. (c) The neutron bombardment does not remove preferentially various gadolinium isotopes from the cubic lattice sites. (d) The radiation (α,β,γ) of the sample causes no transmutations of the gadolinium isotopes.

It is easily seen that the assumptions (b), (c), and (d) are plausible. Assumption (a) may, however, contribute a slight error to our results. Seren *et al.*[5] have measured the activation energy of Gd^{153} and found it to be 125 barns. Presumably the other even isotopes will have similar cross sections. The total cross section of the even isotopes is not expected to be larger than about 1000 barns compared with 46 000±2000 barns of normal gadolinium.[6] The isotopic abundance of Gd^{157} may be slightly too high because of a small contribution from Gd^{156} (20.5%). The error, however is probably small.

[4] Lapp, Van Horn, and Dempster, Phys. Rev. **71**, 745 (1947).
[5] Seren, Friedlander, and Turkel, Phys. Rev. **72**, 888 (1947).

TABLE IV. Absolute cross section of gadolinium isotopes 155 and 157.[a]

Isotope abundance	σ^{155} (barns)	σ^{157} (barns)
(a) Leland[b]	78 000	220 000
(b) Collins *et al.*[c]	87 200	209 000
(c) Our value	88 700	210 000
Value listed by Hughes[d]	70 000±20 000	160 000±60 000

[a] The values are computed using $\sigma_{Gad}=46\,000\pm2000$ barns. Error of cross-section values is estimated to be about 10%.
[b] See reference 7.
[c] See reference 8.
[d] See reference 6.

We have computed the ratio of the cross sections using the relative abundance values measured by Leland,[7] by Collins *et al.*,[8] and by us. The error quoted is that of our measurements and does not include the error in the abundance measurements of normal unbombarded gadolinium. These values are given in Table III. In Table IV we computed the absolute cross section assuming a cross section of 46 000±2000 barns for normal gadolinium.

The method of measuring thermal-neutron capture cross sections by using paramagnetic resonance is limited to a few nuclei only. It can only be used (a) on paramagnetic nuclei, (b) on those which have a large cross section, and (c) on those which have a fairly large, but not too large, hyperfine structure splitting. These requirements restrict this method to the rare earth nuclei and in particular to Sm, Eu, and Gd.

IV. SIGNIFICANCE TO SOLID-STATE PHYSICS

One of the purposes of the experiment with irradiated ThO_2 was the hope that a sufficient density of F centers would be obtained to be detectable with electron spin resonance techniques. Since both thorium and oxygen have no nuclear spin this F-center resonance should be exceedingly narrow and would thus enable us to check a number of theories regarding line width of F centers. However, no such resonance was found. If F centers are present they must be less than 10^{14} cm^{-3} in the sample. Possibly these centers are annealed by the intense radioactivity of the sample.

We also hoped that we might get a sufficient density of dislocations from neutron bombardment so as to see a gadolinium spectrum in a noncubic field. We have found no essential change in the gadolinium spectrum. There is only a very slight change in the linewidth. The radiation damage in thorium oxide crystals is very small, or the damage is annealed very rapidly.

[6] D. J. Hughes and J. A. Harvey, *American Institute of Physics Handbook* (McGraw Hill Book Company, New York, 1957).
[7] W. T. Leland, Phys. Rev. **77**, 634 (1950).
[8] Collins, Rourke, and White, Phys. Rev. **105**, 196 (1957).