Hyperfine Structure of the Spectra of Nd and Gd†

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Study of the hyperfine structure of the spectrum of Nd II with separated isotopes yielded the result that the nuclear magnetic moments of the odd isotopes are $\mu(\text{Nd}^{143}) = -1.1\pm0.1$ nm and $\mu(\text{Nd}^{145}) = -0.69\pm0.10$ nm. The isotope shift in Nd I λ4924 was measured and the ratio of the distances of the neighboring isotopes was found to be $\Delta\nu(142-143):\Delta\nu(143-144):\Delta\nu(144-145):\Delta\nu(145-146):\Delta\nu(146-148):\Delta\nu(148-150) = 0.4_0:0.5_7:0.2_0:0.7_3:1:1.5$. From the hyperfine structure of the spectrum of Gd I the nuclear spins of the odd isotopes Gd^{155} and Gd^{157} were found to be equal to or larger than 5/2, the magnetic moments being negative. Under the assumption that the spins are equal to the values predicted by the nuclear shell structure theory, namely 7/2, the magnetic moments were calculated to be $\mu(\text{Gd}^{155}) = -0.19\pm0.05$ nm and $\mu(\text{Gd}^{157}) = -0.33\pm0.06$ nm. An anomalous isotope shift of the even isotopes of Gd was found: $\Delta\nu(160-158):\Delta\nu(158-156):\Delta\nu(156-154):\Delta\nu(154-152) = 1:0.97\pm0.02:1.21\pm0.05:2.2\pm0.1$.

I. HYPERFINE STRUCTURE OF THE SPECTRUM OF Nd

USING a liquid-air cooled or water cooled hollow cathode discharge tube, a Febry-Pérot etalon and enriched isotopes of Nd^{143} and Nd^{145} whose isotopic constitutions are shown in Table I, Ross and the author studied the hyperfine structure (hfs) of the spectrum of Nd. A short summary of the result of the investigation was published,[1] it being found that the spins of Nd^{143} and Nd^{145} are both 7/2 and the ratio of the magnetic moments is

$$\mu(\text{Nd}^{143})/\mu(\text{Nd}^{145}) = 1.60\pm0.06. \qquad (1)$$

In a somewhat earlier work, Bleaney and Scovil[2] deduced the same nuclear spins, using paramagnetic resonance technique, and got a more precise value of the ratio of the magnetic moments (1.6083 ± 0.0012).

One of the purposes of the present paper is to deduce the magnetic moments of Nd^{143} and Nd^{145} by a calculation that is more rigorous than that given in reference 1.

In the previous work[1] the line Nd II λ4303 ($4f^36s\ ^6I_{7/2} - {}^6K_{9/2}{}^0$)[3] was studied in detail and it was found that Nd^{143} has the total splitting of 0.260 cm^{-1}, the component of the lowest frequency side being the strongest. The line Nd II λ4401 ($4f^46s\ ^6I_{9/2} - {}^6K_{9/2}{}^0$) was found to be single in both the 143 and 145 samples, as far as the diagonal components are concerned. We may assume that the interval factors of the initial and the final levels of λ4401 are equal within the accuracy of 5 percent. These findings about the hfs of λ4303 and λ4401 lead, therefore, to the relation

$$28A^{143}(4f^46s\ ^6I_{7/2}) - 35A^{143}(4f^46s\ ^6I_{9/2}') = 0.260\ \text{cm}^{-1}. \quad (2)$$

The complex $4f^4$ will be assumed to form an ion ^5I via an LS coupling, the $6s$-electron being combined to ^5I by an intermediate coupling. The wave function that depends upon the coupling will be suffixed by ′. In order to solve Eq. (1) we have to determine the coefficients of coupling by solving the energy matrix[4] which is given in Table II. The levels $^{4,6}I$ from the configuration $4f^46s$ with higher J values are near to levels from the configuration $4f^36s5d$ and are likely to be perturbed, so in solving the energy matrix levels with lower J values were given larger weights. In this way we get $\zeta_{4f} = 870$ and $G = 216$. Substituting these values in the energy matrix and using the usual procedure, we get the wave function for $4f^4(^5I)6s$ in intermediate coupling in terms of wave functions in LS coupling:

$$^6I_{9/2}' = K_1\ ^6I_{9/2} + K_2\ ^4I_{9/2}, \quad K_1 = 0.9885, \quad K_2 = 0.1511. \quad (3)$$

Then the interval factor is calculated to be

$$\begin{aligned} A(^6I_{9/2}') = {} & K_1^2[-(17/495)a(s) + (18632/16335)a_f] \\ & + K_2^2[(3/55)a(s) + (749/605)a_f] \\ & + 2K_1K_2[\{4(14)^{\frac{1}{2}}/165\}a(s) \\ & \qquad - \{13(14)^{\frac{1}{2}}/10890\}a_f], \end{aligned} \quad (4)$$

in which we have neglected the relativity correction for the f electrons and have put

$$a_f = \frac{\zeta_f}{Z_f{}^*1836}\frac{\mu}{I}. \quad (5)$$

TABLE I. Isotopic constitution (percent) of the samples of neodymium.

Sample label	Isotope 142	143	144	145	146	148	150
143	4.04	83.93	8.83	1.78	1.16	0.15	0.11
145	1.18	0.84	4.83	78.60	13.71	0.63	0.21
Natural	27.13	12.20	23.87	8.30	17.18	5.72	5.60

† The work with enriched isotopes was performed at the University of Wisconsin in 1950–1951 and was supported by the U. S. Office of Naval Research. The work with natural samples was performed in Tokyo. The enriched isotopes were produced by the Y-12 plant, Carbide and Chemical Division, Oak Ridge, and were obtained by allocation from the U. S. Atomic Energy Commission.

[1] K. Murakawa and J. S. Ross, Phys. Rev. **82**, 967 (1951).
[2] B. Bleaney and H. E. D. Scovil, Proc. Phys. Soc. (London) **63**, 1369 (1950).
[3] The Nd II spectrum was classified by Albertson, Harrison and McNally, Phys. Rev. **61**, 167 (1942).
[4] The notations are the same as those in the book of E. U. Condon and G. H. Shortley, *The Theory of Atomic Spectra* (Cambridge University Press, London, 1935).

TABLE II. Energy matrix of $4f^4(^5I)6s$ in LS coupling.

	$J=17/2$			$J=15/2$	
^6I	$3\zeta-(7/2)G$		^6I	$(13/10)\zeta-(7/2)G$	$-(1/10)51^{\frac{1}{2}}\zeta$
			^4I	$-(1/10)51^{\frac{1}{2}}\zeta$	$(27/10)\zeta+(3/2)G$
	$J=13/2$			$J=11/2$	
^6I	$-(1/5)\zeta-(7/2)G$	$-(1/10)66^{\frac{1}{2}}\zeta$	^6I	$-(3/2)\zeta-(7/2)G$	$-(3/4)\zeta$
^4I	$-(1/10)66^{\frac{1}{2}}\zeta$	$(9/20)\zeta+(3/2)G$	^4I	$-(3/4)\zeta$	$-(3/2)\zeta+(3/2)G$
	$J=9/2$			$J=7/2$	
^6I	$-(13/5)\zeta-(7/2)G$	$-(3/20)14^{\frac{1}{2}}\zeta$	^6I	$-(7/2)\zeta-(7/2)G$	
^4I	$-(3/20)14^{\frac{1}{2}}\zeta$	$-(63/20)\zeta+(3/2)G$			

The level $^6I_{7/2}$ is independent of coupling, and we have

$$A(^6I_{7/2})=-(1/9)a(s)+(448/297)a_f. \qquad (6)$$

Substituting the Goudsmit-Fermi-Segrè formula with the finite nuclear volume correction and Eq. (5) in Eqs. (3), (4), and (6), we get

$$A(^6I_{9/2}')=0.005298\mu, \quad A(^6I_{7/2})=-0.001508\mu. \qquad (7)$$

Putting Eq. (7) in Eq. (2), we get finally

$$\mu(\mathrm{Nd}^{143})=-1.1\pm0.1 \text{ nm},$$

and by the aid of the relation (1), we get

$$\mu(\mathrm{Nd}^{145})=-0.69\pm0.10 \text{ nm}.$$

In a recent work Bleaney *et al.*[5] quote the calculation of Elliott and Stevens: $|\mu(\mathrm{Nd}^{143})|=1.0\pm25$ percent, $|\mu(\mathrm{Nd}^{145})|=0.62\pm25$ percent.

The shift of the even isotopes in the spectrum of Nd I was studied by Klinkenberg,[6] and he found an anomalously large isotope shift of $\mathrm{Nd}^{148}-\mathrm{Nd}^{150}$. In the present work, using natural neodymium also, Klinkenberg's lines were measured with an accuracy better than that of Klinkenberg. None of them was found among the classified lines of Schuurmans[7] and Klinkenberg.[8] Of the classified lines, the line λ4924 ($4f^46s^2\,^5I_4-20_5$) [the level 20300.86 ($J=5$) found by Klinkenberg[8] is here denoted as 20_5 for brevity] was found to have the largest shift. The measured hfs for this line is given in Fig. 1. The hfs of the same line for the enriched isotopes was not resolved, but from the measured intensity distribution the centers of gravity of Nd^{143} and Nd^{145} could be determined, and are given at the bottom of Fig. 1. The shift in the even isotopes will be discussed in Sec. III.

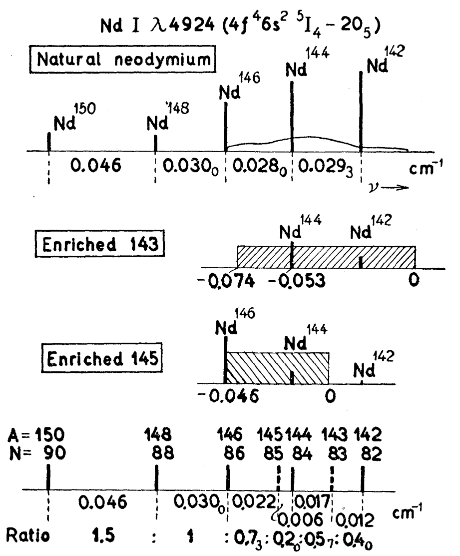

FIG. 1. Hfs of Nd I λ4924 and the isotope shift. A and N are atomic number and neutron number respectively.

II. HYPERFINE STRUCTURE OF THE SPECTRUM OF Gd

The shift of even isotopes in the spectrum of Gd was first measured by Klinkenberg.[8] Suwa[9] and Brix and Engler[10] measured more accurately the hfs of the Gd lines, but none of them detected components due to Gd^{154} and Gd^{152}.

Natural gadolinium is known to consist of seven isotopes:[11] 160 (21.79), 158 (24.78), 157 (15.71), 156 (20.59), 155 (14.78), 154 (2.15), and 152 (0.20). A number in parentheses represents the abundance in percent. The purpose of the present work is to detect the components due to the rarer isotopes Gd^{154} and Gd^{152} and to measure the magnetic moments of the odd isotopes. A liquid-air cooled hollow-cathode discharge tube which has been improved since 1951 was used.[12] In lines in which the splittings of Gd^{155} and Gd^{157} respectively are negligibly small, the component due to Gd^{154} could be clearly observed. In several lines the component due to Gd^{152} could be recognized on heavily exposed plates. A typical example is the line λ4092.7,[13] and the result of measurement is shown in Fig. 2. Some of the interference patterns of λ4092.7 are repro-

[5] Bleaney, Scovil, and Trenam, Proc. Roy. Soc. (London) **223**, 1 (1954).

[6] P. F. A. Klinkenberg, Physica **11**, 327 (1945). Quite recently G. Nöldeke and A. Steudel [Z. Physik **137**, 632 (1954)] have studied the same subject as Klinkenberg more accurately.

[7] P. Schuurmans, Physica **11**, 419 (1946).

[8] P. F. A. Klinkenberg, Physica **12**, 33 (1946).

[9] S. Suwa, Phys. Rev. **86**, 247 (1952); J. Phys. Soc. (Japan) **8**, 377 (1953).

[10] P. Brix and H. D. Engler, Z. Physik **133**, 362 (1952).

[11] D. C. Hess, Phys. Rev. **74**, 773 (1948).

[12] K. Murakawa, J. Phys. Soc. Japan **9**, 391 (1954).

[13] The Ge I and Gd II spectra were classified extensively by H. N. Russell, J. Opt. Soc. Am. **40**, 550 (1950).

duced in Fig. 3. The position of the Gd^{152} component relative to the Gd^{160} component is somewhat difficult to measure, and an error of ± 0.006 cm^{-1} must be admitted.

In the unclassified line λ4436.1 (see Fig. 2) the positions of the components due to Gd^{160}, Gd^{158}, and Gd^{156} are least disturbed by the odd isotopes. From the hfs of λ4436.1 and λ4092.7 the ratio of the distances of the neighboring isotopes is observed to be $\Delta\nu$ (160−158) : $\Delta\nu$ (158−156) : $\Delta\nu$ (156−154) : $\Delta\nu$ (154−152) = 1 : 0.97 $\pm$0.02 : 1.21$\pm$0.05 : 2.2$\pm$0.1. With respect to the odd isotopes we get $\Delta\nu$ (160−158) : $\Delta\nu$ (158−157) : $\Delta\nu$ (156−155) = 1 : 0.92$\pm$0.02 : 0.74$\pm$0.02. Suwa[9] obtained $\Delta\nu$ (160−158) : $\Delta\nu$ (158−156) = 1 : 0.97$\pm$0.06. Brix and Engler[10] found 160−158−156 to be equidistant within the accuracy of their measurements.

The line λ4436 has large splittings of the odd isotopes (see Fig. 2). The intensity distribution shows that the

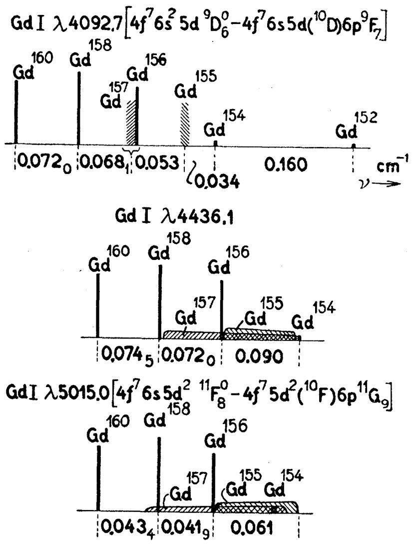

FIG. 2. Hfs of Gd I λ4092.7, λ4436.1 and λ5015.0.

spins of Gd^{157} and Gd^{155} are at least 2, so that they must be 5/2, 7/2, or 9/2. With respect to the signs of the magnetic moments of the odd isotopes, the hfs of λ5015.0 (see Fig. 2) is more favorable for negative than for positive moments, although a slight amount of room is left for positive moments. The single-particle model of the nuclear shell structure predicts the state $f_{7/2}$ for Gd^{157} and Gd^{155}, so we shall assume that the spins of Gd^{157} and Gd^{155} are 7/2 and calculate the magnetic moments.

The total width of the Gd^{155} structure in λ5015 (see Fig. 2) was found to be approximately 0.061 cm^{-1}. Thus we have

$$(119/2)A(4f^7 6s5d^2\ ^{11}F_8{}^0) - (133/2)A(4f^7 5d^2 6p\ ^{11}G_9) = -0.061\ \mathrm{cm}^{-1}.$$

Assuming that the complex $4f^7\ ^8S_{7/2}$ is of LS coupling and therefore contributes nothing to the hfs splitting,

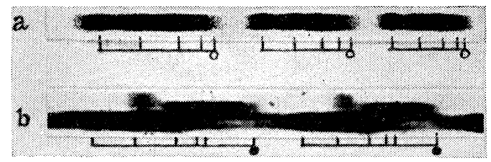

FIG. 3. Enlargement of interference patterns of Gd I λ4092.7. (a) 15-mm etalon. (b) 8-mm etalon. The components due to Gd^{154} and Gd^{152} are marked with ○ and ● respectively.

and using the relations

$$A(4f^7 5d^2 6s\ ^{11}F_8) = (37/80)a_d' + (3/80)a_d'' + (1/5)a_d''' + (1/16)a(s),$$

$$A(4f^7 5d^2 6p\ ^{11}G_9) = (37/90)a_d' + (1/30)a_d'' + (8/45)a_d''' + (1/6)a_p',$$

we get

$$\mu(Gd^{155}) = -0.19 \pm 0.05\ \mathrm{nm},$$

where we have assumed that $\zeta_{5d} = 864$ cm^{-1} and $\zeta_{6p} \approx 1250$ cm^{-1} for Gd I. Similarly we get

$$\mu(Gd^{157}) = -0.33 \pm 0.06\ \mathrm{nm}.$$

Suwa[9] deduced $|\mu(Gd^{155})| = 0.2_5 \pm 0.15$ nm and $|\mu(Gd^{157})| = 0.3 \pm 0.2$ nm from the measurement of the same spectrum.

III. DISCUSSION OF THE ISOTOPE SHIFT

The large isotope shifts between the neutron numbers (N) 90 and 88 in Nd and Sm are now well known. A third example has been found in Gd in the present work. The ratio of the distances between neighboring isotopes in these elements is summarized in Table III, and it is seen that the trend is the same in all of the three elements listed.

The subshell of $h_{9/2}$ neutrons is completed at $N = 92$. The above-mentioned anomaly can be phenomenologically described by the apparent attraction of the $N90(=92-2)$ component to the $N92$ component and the resultant large shift between the $N90$ and $N88$ components. Exactly the same phenomenon is observed at the completion of the subshell of $h_{11/2}$ neutrons which occurs at $N = 76$: Ross and the author[14] detected in the spectrum of Te II an anomalously large shift between the $N74(=76-2)$ and $N72$ components and a relatively small shift between the $N76$ and $N74$ components. The observed facts seem to show that the

TABLE III. Isotope shift in Gd, Nd, and Sm.

Element	N 96	94	92	90	88	86	84	82
Gd		0.83 :	0.80 :	1 :	1.9			
Nd					2 :	1.30 :	1.22 :	1.27
Sm[a]				1 :	1.85 :	1.26 :		1.105×2

[a] Data were taken from P. Brix and H. Kopfermann, Z. Physik 126, 344 (1949). Sm isotope for N = 84 does not exist, so the shift for 86 −82 is given.

[14] J. S. Ross and K. Murakawa, Phys. Rev. 85, 559 (1952).

conspicuous isotope shift anomaly is connected with the completion of the subshell of neutrons with high l value.

Ford [15] and Wilets *et al.*[16] correlated the isotope shift in atomic spectra of heavy elements with the nuclear intrinsic deformation parameter (β) that was introduced by Bohr,[17] in such a way that the actual shift minus the shift predicted by the finite volume theory determines $d\beta/dN$ but not β itself. The above-mentioned shift anomaly would enter as anomalous slope in the β *versus* N curve in even-even nuclei (Fig. 4 of the article of Ford[15]). In this connection it may be remarked that in even-even nuclei the β *versus* N curve is relatively simple, β vanishing only at magic numbers, and the completion of subshell has a minor effect on the general trend of β, according to Ford's Fig. 4. In contrast to this, in even-odd nuclei the completion of subshell has often a marked influence on the form of the β *versus* Z curve (Z=proton number).[18] These facts will probably be connected with the more pronounced importance of the rôle which the alpha-particle model plays in even-even nuclei.

I should like to thank Dr. Ross for his kind cooperation in the work with enriched isotopes. The very pure sample of gadolinium which was used in the present experiment was kindly given to me by Professor Mack to whom my thanks are due.

[15] K. W. Ford, Phys. Rev. **90**, 29 (1953).
[16] Wilets, Hill, and Ford, Phys. Rev. **91**, 1488 (1953).
[17] A. Bohr, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **26**, No. 14 (1952).
[18] See, for example, K. Murakawa and T. Kamei, Phys. Rev. **92**, 325 (1935).

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Stern-Gerlach Experiment on Polarized Neutrons

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A neutron beam was polarized by total reflection from a magnetized iron mirror. The beam was then analyzed by passing it through an inhomogeneous magnetic field. From the deflection pattern obtained, it is inferred that the resultant neutron spin in the polarized beam was parallel to the magnetic field applied to the mirror. Thus the nuclear and magnetic scattering amplitudes for iron are of the same sign when the neutron spin and electronic spin are oppositely directed, and conversely.

INTRODUCTION

When a beam of neutrons passes through a material body the process may be considered analogous to the passage of light through a refractive medium. Thus we may define an index of refraction $n=[(E-V)/E]^{\frac{1}{2}}$, where E is the energy of a neutron, and V is the average potential energy of the neutron while in the body. If θ is the glancing angle of incidence, it follows that total reflection will occur for $\theta=(V/E)^{\frac{1}{2}}$. In other words, all wavelengths greater than the critical wavelength,

$$\lambda_c=\theta(h^2/2mV)^{\frac{1}{2}},$$

will be totally reflected.

In the case of a magnetized iron mirror, the potential energy is $V=V_n+V_m$, where V_n is that due to the nuclei and $V_m=-\boldsymbol{\mu}\cdot\mathbf{B}$ is the potential energy of the neutron's magnetic moment $\boldsymbol{\mu}$ in the average net field $\mathbf{B}$. Thus, if λ_c^+ and λ_c^- are the critical wavelengths for $\boldsymbol{\mu}$ parallel to $\mathbf{B}$ and $\boldsymbol{\mu}$ antiparallel to $\mathbf{B}$, respectively, we have

$$\lambda_c^+>\lambda_c^- \quad (B\neq 0).$$

The spectrum of reflected neutrons will therefore have the general appearance shown in Fig. 1 (if the spectrum of the incident beam is assumed to be Maxwellian). Thus the spin state with $\boldsymbol{\mu}$ antiparallel to $\mathbf{B}$ is more abundant. Since the gyromagnetic ratio of the neutron is negative,[1] we have the result that the resultant spin of the neutrons in the totally reflected

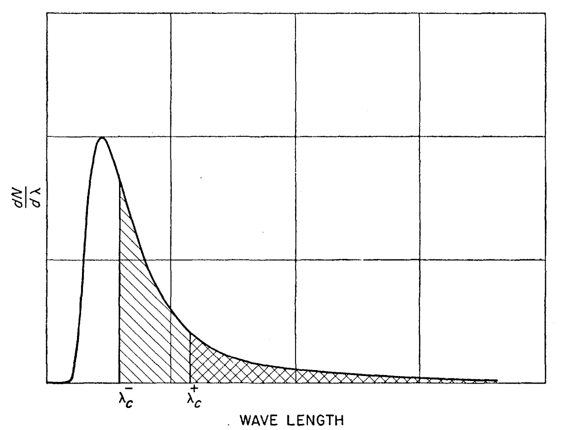

FIG. 1. Intensity distribution of neutrons reflected from magnetized iron mirror. For $\boldsymbol{\mu}$ parallel to $\mathbf{B}$, $\lambda>\lambda_c^+$. For $\boldsymbol{\mu}$ antiparallel to $\mathbf{B}$, $\lambda>\lambda_c^-$.

[1] P. N. Powers, Phys. Rev. **54**, 827 (1938).

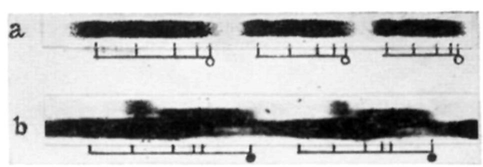

FIG. 3. Enlargement of interference patterns of Gd I λ4092.7. (a) 15-mm etalon. (b) 8-mm etalon. The components due to Gd^{154} and Gd^{152} are marked with ○ and ● respectively.