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## Optical Double Resonance in Radioactive Atoms: Spin and Nuclear Moments of the Ground State of Cd<sup>109</sup>†

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The optical double-resonance technique has been employed for the study of the Zeeman effect and hyperfine structure of the  $(5s5p) {}^3P_1$  state of 470-day Cd<sup>109</sup>. The nuclear spin,  $I$ , magnetic hyperfine interaction constant,  $A$ , and quadrupole interaction constant,  $B$ , are:  $I = \frac{5}{2}$ ,  $A = -1, 148.6 \pm 2.0$  Mc/sec, and  $B = -167.3 \pm 2.0$  Mc/sec. If nuclear structure and quadrupole antishielding corrections are neglected, the corresponding nuclear moments are  $\mu = -0.8286(15)\mu_N$  and  $Q = +0.78(10)$  b. These values are discussed in terms of the configuration mixing model of Arima and Horie. A number of the problems encountered in the application of the double resonance technique to radioactive atoms are discussed. The sensitivity of the method is limited by the shot noise in the instrumentally scattered light. In the case of the present apparatus, Zeeman resonances are detectable with  $2 \times 10^6$  cadmium atoms in the vapor phase.

## I. INTRODUCTION

THE optical double-resonance technique as suggested by Brossel and Kastler<sup>1</sup> and Brossel and Bitter<sup>2</sup> is well suited for the study of the Zeeman effect and hyperfine structure of the optically excited states of radioactive atoms. Such studies are useful for the determination of nuclear moments that do not produce an interaction in the normal state of the atom. For example, the double-resonance technique has found considerable application in the determination of the quadrupole moments of the alkali atoms.<sup>3-12</sup> Here we

report in detail<sup>13</sup> on the application of this method to 470-day Cd<sup>109</sup>. This isotope and the other Group II elements are characterized by diamagnetic  $(ns)^2 {}^1S_0$  electronic ground states that exhibit no hfs. The determination of the nuclear moments of these elements by the well-known radioactive atomic-beam method would be difficult at best. With the double-resonance technique, one can excite and study these atoms in the paramagnetic  $(nsnp) {}^3P_1$  state. This state exhibits both a magnetic-dipole and an electric-quadrupole interaction. The method is also clearly applicable to many other elements. This is the first report on a series of double-resonance measurements that we have in progress on the cadmium and zinc isotopes. For this reason, many details and general relations are given that will be of interest to the entire series of measurements.

With important exceptions, many of the atomic-beam studies that have been made of nuclear moments have been restricted to the case of odd-proton nuclei simply because such atoms exhibit paramagnetic ground states. The application of the double-resonance method to even-proton elements such as the alkaline earths and the rare gases will provide complimentary data on odd-neutron nuclei. It is particularly useful to obtain the magnetic and quadrupole moments for a series of isotopes of a given element so that the effect of adding

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<sup>1</sup> J. Brossel and A. Kastler, *Compt. Rend.* **229**, 1213 (1949).

<sup>2</sup> J. Brossel and F. Bitter, *Phys. Rev.* **86**, 308 (1952).

<sup>3</sup> P. L. Sagalyn, *Phys. Rev.* **94**, 885 (1954).

<sup>4</sup> H. Kruger and K. Scheffler, *J. Phys. Radium* **19**, 854 (1958).

<sup>5</sup> J. N. Dodd and R. W. N. Kinnear, *Proc. Phys. Soc. (London)* **75**, 51 (1960).

<sup>6</sup> G. J. Ritter and G. W. Series, *Proc. Roy. Soc. (London)* **A238**, 473 (1957).

<sup>7</sup> H. Bucka, H. Kopfermann, and J. Ney, *Z. Physik* **167**, 375 (1962).

<sup>8</sup> U. Meyer Berkhout, *Z. Physik* **141**, 185 (1955).

<sup>9</sup> H. Bucka, H. Kopfermann, and A. Minor, *Z. Physik* **161**, 123 (1961).

<sup>10</sup> K. Althoff, *Z. Physik* **141**, 33 (1955).

<sup>11</sup> H. Bucka, H. Kopfermann, E. W. Otten, *Ann. Physik* **4**, 39 (1959).

<sup>12</sup> H. Bucka and G. von Oppen, *Ann. Physik* **10**, 119 (1962).

<sup>13</sup> Preliminary results are reported by M. McDermott and R. Novick, *Bull. Am. Phys. Soc.* **6**, 142 (1961).

successive neutrons can be studied in detail. The alkaline earths and heavy rare gases are well suited for such studies.

One of the features that makes the double-resonance method particularly attractive for the present purpose is its high sensitivity. The sensitivity of the method derives from two facts: (1) that although resonances are effected by low-energy rf photons ( $h\nu_{rf} \ll kT$ ), they are detected optically ( $h\nu_{opt} \gg kT$ ), so that thermal noise is of no concern, and (2) that the effective-resonance scattering cross section is many times greater than the geometric cross section. The effective-resonance cross section is given by<sup>14</sup>

$$\sigma = 2 \left( \frac{\pi \ln 2}{\Delta\nu_A^2 + \Delta\nu_S^2} \right)^{1/2} \pi r_0^2 c f, \quad (1)$$

where  $\Delta\nu_A$  is the Doppler width for the absorbing atoms,  $\Delta\nu_S$  is the source width (assumed to have a Gaussian shape),  $f$  is the oscillator strength for the transition,  $r_0$  is the classical radius of the electron, and  $c$  is the velocity of light. If we consider the excitation of the  $^3P_1$ - $^1S_0$ ,  $\lambda=3261$  Å, transition in cadmium ( $f=0.002$ ) by a source of width  $0.14$  cm<sup>-1</sup> (see below) in a sample with a Doppler broadened absorption line of width  $0.046$  cm<sup>-1</sup> (200°C), the cross section is approximately  $3 \times 10^{-14}$  cm<sup>2</sup>, almost  $10^2$  times greater than the geometric cross section. In the case of a fully allowed transition the cross section would be larger by a factor of about 500. The optical system and light sources described below produce a photon flux of about  $3 \times 10^{15}$  photons per cm<sup>2</sup>/sec at the scattering cell. Thus, each atom scatters about 90 photons/sec. In practice, the detector noise is due almost entirely to the shot noise in the photo current which arises from instrumentally scattered light; this light comes from optical imperfections in the scattering cell and multiple reflections in the oven. We have found that we can observe Zeeman resonances in stable cadmium at temperatures at least as low as 13°C where the vapor pressure is about  $7 \times 10^{-12}$  Torr. This corresponds to a total of about  $2 \times 10^6$  atoms in the vapor phase. There is some indication that a substantial fraction of the atoms are to be found on the walls at any one time, in which case a total of  $2 \times 10^6$  atoms in a cell would give much lower signals than the same number in the vapor phase in equilibrium with an excess of the solid. In the case of another isotope<sup>15</sup> we have observed resonances with a total sample of  $10^{11}$  atoms.

The resolution of the method is determined by the lifetime of the excited state. The  $^3P_1$  state of the alkaline earths is metastable; in the case of cadmium the lifetime of this state is about 2.4 μsec and the

corresponding resonance width is about 130 kc/sec. Since a typical hyperfine splitting is in the order of 1000 Mc/sec, the resolution is at least 10 000. For shorter lived states, the resolution is, of course, correspondingly smaller.

An important requirement of the present technique is that the sample be isotopically enriched. This requirement arises from the fact that the total number of atoms in the vapor phase must be less than the number required for heavy trapping of the resonance radiation, while the number of atoms of the isotope under study must be sufficient to give a useful signal (in the case of a high-spin cadmium isotope we may need as much as  $2 \times 10^9$  atoms to produce an observable signal for individual Zeeman transitions). The number of atoms required for heavy trapping cannot be exactly specified, but our experience indicates that trapping effects become serious for the  $^3P_1$  states of Cd when the atom density is about  $10^{13}$  per cc. This implies that the concentration of the isotope under investigation must be greater than 20 ppm (assuming a 10-cc cell). This requirement precludes the use of ( $n, \gamma$ ) production reactions in all but a few favorable cases with large thermal neutron cross sections, unless, of course, some means is employed to enrich the sample.

The chemical identity of the sample is assured by the resonance fluorescence. The isotopic identification is somewhat less certain. Confusion between signals arising from stable and radioactive species can be avoided by comparing the observed spectrum with that obtained with a sample of stable material. Signals arising from short-lived isotopes can be eliminated by waiting several mean lives between production and observation. The possibility that the observed resonances may arise from an isotope having a lifetime comparable to the one under study cannot be completely eliminated; however, knowledge of the production and history of the sample, together with studies of the radioactivity, serve to reduce this hazard (see below). In Sec. II we give a general discussion of the problems encountered in the application of the double-resonance method to small samples of radioactive atoms. A number of general intensity and transition frequency formulas are presented. In Secs. III and IV we evaluate and discuss the Cd<sup>109</sup> moments.

## II. THE DOUBLE-RESONANCE METHOD: SPIN AND HYPERFINE INTERVALS IN Cd<sup>109</sup>

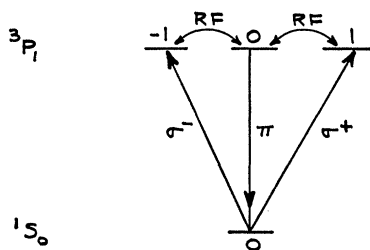
The double-resonance method has been described in a review article by Series.<sup>16</sup> Briefly, the method consists of optically exciting an atom with nonisotropic and/or polarized resonance radiation. Such excitation results in unequal populations in the various magnetic sub-levels of the optically excited state. When the excited atom decays to its ground state, it will radiate with a characteristic directional pattern and polarization. If

<sup>14</sup> This result is derived easily from material given in A. C. G. Mitchell and M. W. Zemansky, *Resonance Radiation and Excited Atoms* (Cambridge University Press, New York, 1961), Chap. 3.

<sup>15</sup> The isotope is Cd<sup>115m</sup>. B. Perry, M. McDermott, and R. Novick, *Bull. Am. Phys. Soc.* **7**, 533 (1962).

<sup>16</sup> G. W. Series, *Rept. Progr. Phys.* **22**, 280 (1959).

FIG. 1. Transitions involved in optical double resonance with unpolarized  $\sigma$  excitation and  $\pi$  detection for nuclear spin  $I=0$ .



radio-frequency transitions are induced among the magnetic substates of the excited atom, the polarization and directionality of the fluorescence will be modified. Thus, excited-state transitions may be detected by observing the intensity of the fluorescence in a given direction with a suitable analyzer. In the present experiment, the atoms are excited with unpolarized light directed along the static field. This leads to the selection rule  $\Delta m = \pm 1$  in absorption. In the case of a spin-zero ( $I=0$ ) isotope, this results in equal population of  $m = \pm 1$  substates and no population of the  $m=0$  substate of the  $^3P_1$  level. The detector is oriented at right angles to the field and contains an analyzer that accepts only  $\pi$  ( $\Delta m=0$ ) light. In the case of the spin-zero isotope, the atoms radiate  $\pi$  light only when transitions are induced from the  $m = \pm 1$  states to the  $m=0$  state with a suitable radio-frequency field. Ideally, the detector receives light only when a Zeeman transition is effected. (See Fig. 1.) In the case of isotopes with higher spin,  $\pi$  light is radiated even in the absence of a transition, but in all cases except transitions between  $m = +\frac{1}{2}$  and  $m = -\frac{1}{2}$  states,<sup>17</sup> Zeeman resonances result in a change of the intensity of the  $\pi$  light.

### A. Resonance Apparatus

The apparatus is shown schematically in Fig. 2. A system of quartz lenses of aperture<sup>18</sup>  $f/1.5$  focuses the 3261-Å intercombination line from a cadmium discharge lamp onto a quartz vessel containing the cadmium sample. The resonance cell is placed within a double-walled oven and is heated by a gas flame. The magnetic field is parallel to the direction of illumination and is provided by a Helmholtz pair capable of producing a field of 150 G. Two additional orthogonal sets of coils cancel the undesired components of the laboratory field. A second lens system, also of aperture  $f/1.5$ , collects light scattered at right angles to the incident light. This second set of lenses contains a linear polarizer<sup>19</sup> oriented so that only light polarized parallel to the magnetic field ( $\pi$  light) is passed. The  $\pi$  light collected

by the lenses is focused into an aluminized light pipe with a photomultiplier fixed at the far end. A Schott UG11 ultraviolet filter in front of the photomultiplier serves to partially isolate the 3261-Å intercombination line and to eliminate visible light. Radio-frequency transitions are excited by a two-turn coil oriented to provide an rf magnetic field perpendicular to the static magnetic field. The rf field is 100% amplitude modulated by a 280 cps square wave and the 280 cps component of the scattered  $\pi$  light is detected by means of a narrow-band amplifier and phase-sensitive detector. With the arrangement described, any rf transition may be detected which results in a change in the intensity of the scattered  $\pi$  light.

### B. Light Sources

The success of the double-resonance method depends critically upon the light source. A suitable source must produce intense, stable resonance radiation without self-reversal and without appreciable broadening. Light produced outside of the absorption line of the sample atoms will contribute to the noise but not to the signals. The cadmium isotope shifts are sufficiently small so that it is possible to use a lamp containing normal cadmium to excite the radioactive species. In Fig. 3 we have plotted the lamp profile which would be obtained if each isotope contributed to the intensity according to its relative abundance in the lamp. The spectral width of the individual lines is chosen to give a combined width approximately equal to that observed. The positions of the isotopes were obtained from work by Kelly and Tomchuk<sup>20</sup> and Stroke.<sup>21</sup>

Considerable preliminary investigation was necessary before a suitable source of Cd 3261-Å radiation was developed. In particular, a number of electrodeless discharge lamps operated at microwave frequencies were tried in view of the success of other workers<sup>22,23</sup>

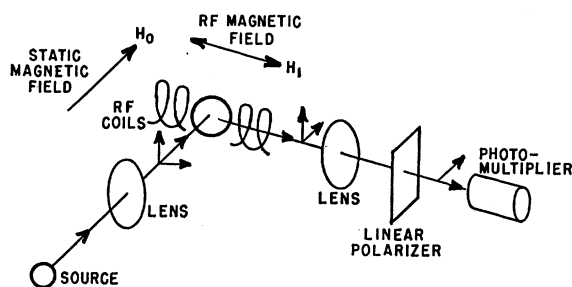


FIG. 2. Schematic diagram of the apparatus.

<sup>20</sup> F. M. Kelly and E. Tomchuk, Proc. Phys. Soc. (London) 74, 689 (1959).

<sup>21</sup> H. H. Stroke (private communication). Results for the Cd<sup>109</sup> isotope shift are quoted in H. H. Stroke, Quarterly Progress Report, Research Laboratory of Electronics, Massachusetts Institute of Technology (unpublished).

<sup>22</sup> M. Zelikoff, P. H. Wyckoff, L. M. Aschenbrand, and R. S. Loomis, J. Opt. Soc. Am. 42, 818 (1952).

<sup>23</sup> F. S. Tompkins and Mark Fred, J. Opt. Soc. Am. 47, 1087 (1957).

<sup>17</sup> This limitation only applies in the linear Zeeman region (see below). In Cd<sup>109</sup> we have observed these transitions in fields of 50 G and greater.

<sup>18</sup> The rather sharp convergence of the light introduces about 3% of  $\pi$  light. This slightly reduces the signal and increases the background.

<sup>19</sup> For a description of the polarizer, see M. N. McDermott and R. Novick, J. Opt. Soc. Am. 51, 1008 (1961).

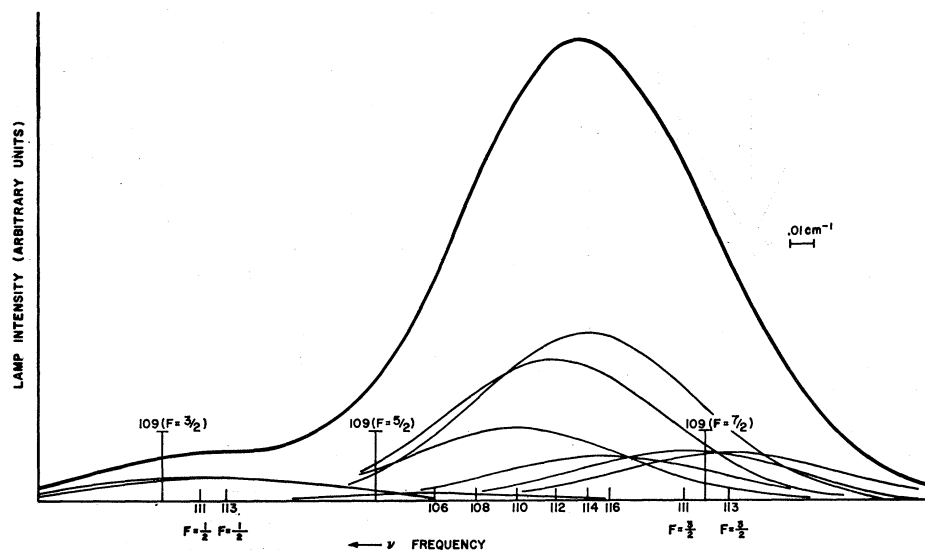


FIG. 3. Theoretical spectrum for a lamp containing natural cadmium.

with such lamps. The microwave lamps, however, proved to give no greater absorbable light flux (light within the Doppler profile of the absorbing atoms) at 3261 Å than did the simple design ultimately adopted. This consisted of a 1.25-in.-diam quartz sphere containing cadmium metal and argon gas at a pressure of about 1 Torr. This was excited by placing it directly in the tank coil of a 20 Mc/sec Hartley oscillator.

A rather careful filling procedure is required to produce lamps that have a useful life of at least 20 h; at the end of life the intensity drops appreciably and the lamps become unstable. The bulbs are evacuated to a pressure of about  $10^{-7}$  Torr and baked for 12 h at 1000°C. After bakeout, argon gas at a pressure of a few Torr is admitted and a vigorous rf discharge is maintained for several minutes to purge the walls of the bulb. The impurities liberated by this procedure change the color of the discharge and sometimes extinguish it. The contaminated argon is pumped from the bulb and the entire procedure repeated until the color of the discharge indicates that no further contaminants are being released. At the conclusion of this initial cleaning, the cell is again evacuated and a small amount of cadmium is distilled into it. The cadmium metal is cleaned with a discharge that is started with argon but maintained for a few minutes on the pure metal vapor. This is accomplished by pumping out the argon with the discharge running. The cadmium is driven from the bulb with this discharge and is then distilled back with external heating. The bulb is filled with argon at a pressure between 0.1 and 3.0 Torr and sealed off. The exact pressure is not critical.

The total output of these lamps in the 3261-Å line has been determined with both a calibrated Eppley thermopile and a photocell calibrated with a standard lamp. Under conditions that produce the largest double-resonance signals, the total power radiated in

the intercombination line is about 2 W or  $3 \times 10^{18}$  photons/sec. The 3261-Å output is a strongly peaked function of the exciter supply voltage.

The Hartley oscillator used to excite the lamp is operated from 0.01% regulated dc power supplies in both the filament and plate circuits. With these precautions, it is found that the noise arising from lamp fluctuations is less than the shot noise resulting from the instrumentally scattered light.

The spectral profile of the lamp was determined with a simple magnetic scanning technique; the effective width of the 3261-Å line was found to be about 0.14  $\text{cm}^{-1}$  or about three times the Doppler width at 200°C. The apparatus used for the profile studies is shown in Fig. 4. The light to be analyzed is linearly polarized at right angles to the scanning field. An absorption bulb containing cadmium metal is placed in the field and the light transmitted through the absorption cell is monitored with a photocell. The temperature of the scanning cell is adjusted to give a small, roughly 10%, absorption at zero field. The scanning curves obtained by varying the magnetic field on the absorption bulb

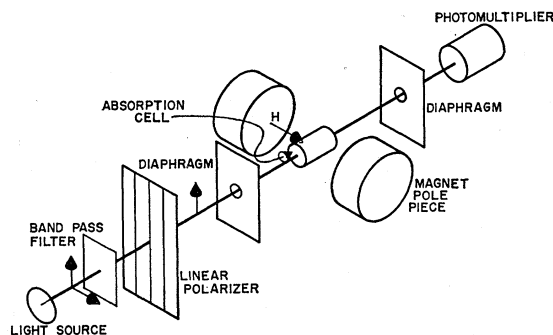


FIG. 4. Schematic diagram of the magnetic scanning apparatus.

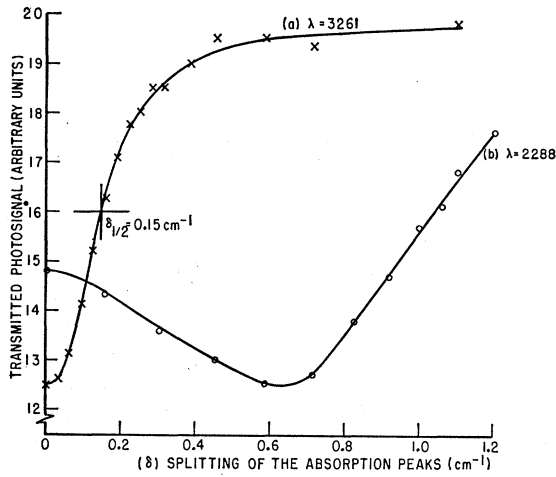


FIG. 5. Magnetic scanning curves for the Cd 3261 and Cd 2288 resonance lines.

are analyzed under the assumption that the absorption bulb contains only a single even isotope; i.e., the hyperfine structure of the odd isotopes and the isotope shifts are neglected. Under these assumptions, and with polarization of the incident light, only the  $^1S_0(m=0)$ – $^3P_1(m=\pm 1)$  transitions are effective in absorbing the light. The frequencies of these transitions vary linearly with the magnetic field; at zero field they coalesce into a single line and at higher fields they split into two lines symmetrically displaced from the zero-field position. The separation of the absorption peaks,  $\delta$ , is given by

$$\delta = 2g_J\mu_0 H/h, \quad (2)$$

where  $H$  is the magnetic field,  $\mu_0$  is the Bohr magneton,  $h$  is Planck's constant, and  $g_J$  is the Landé  $g$  factor for the  $^3P_1$  state of Cd. At sufficiently high fields, the line separation is much greater than the width of the lamp profile and there is no absorption; as the field is decreased the absorption lines move across the lamp profile with a consequent increase in absorption. This technique provides limited but adequate resolution for the present purposes.

For simplicity we assume that the source has a Gaussian profile and we simply seek to determine an effective width for the emission line. With this and the previous assumptions, the frequency separation  $\delta_{1/2}$  between the Zeeman-shifted absorption peaks at a field which restores half of the light absorbed at zero field is given by

$$\delta_{1/2} = [(\Delta\nu_S)^2 + (\Delta\nu_A)^2]^{1/2}, \quad (3)$$

where  $\Delta\nu_S$  and  $\Delta\nu_A$  are the full widths at half-maximum of the emission line and the absorption line, respectively. A typical magnetic scanning curve is shown in Fig. 5(a). Using the above analysis, we find that the effective width of the source is  $0.14 \text{ cm}^{-1}$ . Under less-favorable operating conditions, a lamp may exhibit considerable self-reversal. In Fig. 5(b) we show a scanning curve for

the 2288-Å resonance line produced by a lamp that was optimized for the intercombination line. The intensity near the center of the line is severely reduced, and most of the light is located in wings separated by nearly four Doppler widths.

### C. Resonance Cell and Isotope Preparation

The  $\text{Cd}^{109}$  used in this experiment was produced by a  $(p,n)$  reaction on silver. It was hoped that the sample obtained by this method would have a high isotopic purity. In spite of this precaution, the results indicated that the  $\text{Cd}^{109}$  was contaminated with a considerably larger amount of stable cadmium. This was only a problem at low values of the magnetic field where the Zeeman resonances were not well separated (see below). About 1 mCi of  $\text{Cd}^{109}$  was obtained<sup>24</sup> as a metallic cadmium plating on a small platinum wire.

The resonance cell consisted of a  $1\frac{3}{4}$ -in. diam, 2-in.-long cylindrical quartz bulb having two 6 mm tubular connections. One connection was used for pumping and the other for loading. The platinum wire was placed in a small quartz vessel attached to the loading sidearm. The  $\text{Cd}^{109}$  was evaporated from the wire by heating and condensed in the bulb by cooling it with liquid nitrogen. Geiger-counter readings indicated that substantially all of the activity was transferred from the wire into the bulb. The loading side arm was sealed off and the cell warmed up before final seal-off.

$\text{Cd}^{109}$  decays by  $K$  capture to the 88-keV level of  $\text{Ag}^{109}$ . The isomeric transition is almost entirely converted, and the principal radiation is the 25-keV silver x-ray line. This line and the much weaker 88-keV  $\gamma$  ray were detected with a proportional counter. There was no evidence for any other activity in the bulb.

### D. Low-Field Zeeman Transitions; Spin Determination

The nuclear spin is readily determined from the observation of low-field Zeeman transitions. In the case of a  $^3P_1$  atomic state, these resonances are characterized by the Landé  $g$  factors:

$$g_F(I+1) = g_J \frac{1}{I+1} + g_I \frac{I}{I+1}, \quad (4a)$$

$$g_F(I) = g_J \frac{1}{I(I+1)} + g_I \frac{I(I+1)-1}{I(I+1)}, \quad (4b)$$

$$g_F(I-1) = -g_J \frac{1}{I} + g_I \frac{I+1}{I}. \quad (4c)$$

Here  $g_F(F)$  is the  $g$  factor for the state of total angular momentum  $F$ ,  $I$  is the nuclear spin, and  $g_J$  and  $g_I$  are

<sup>24</sup> Nuclear Science and Engineering Corporation, Pittsburgh, Pennsylvania.

the electronic and nuclear  $g$  factors, respectively. The latter quantities are defined by

$$\mathbf{u}_J = -g_J\mu_0\mathbf{J} \quad \text{and} \quad \mathbf{u}_I = -g_I\mu_0\mathbf{I}, \quad (5)$$

where  $\mu_0$  is the Bohr magneton and  $\mathbf{u}_J$  and  $\mathbf{u}_I$  are the electronic and nuclear magnetic moments, respectively. The term in  $g_I$  is much smaller ( $10^{-3}$ ) than the leading term and can be safely neglected. In this approximation, the low-field Zeeman resonances are completely specified by the magnetic field and the nuclear spin. In a spin determination the magnetic field is calibrated with the spin-zero resonance in stable cadmium.

Unfortunately, there is a possible ambiguity in the determination of the spin from the observation of low-field Zeeman resonances. The state of maximum  $F$  of an atom with nuclear spin  $I$  has the same Zeeman frequency as the state of lowest  $F$  of an atom with nuclear spin  $I' = I + 1$ . Thus, the observation of a single low-field Zeeman resonance in one of these states does not uniquely determine the spin unless the state is further identified. Such identification can be obtained by increasing the magnetic field so that the Zeeman resonance splits into two  $F$  components as  $I$  and  $J$  become uncoupled. Alternatively, Zeeman resonances can be sought in two or more hyperfine states.

There is a unique relation between spin and  $g$  factor in the state  $F = I$ . Unfortunately, however, this state has a small magnetic moment and requires an intense rf field. In the case of a large nuclear spin, the required field may be great enough to induce spin-zero transitions in any stable cadmium that may contaminate the bulb, even though the spin-zero resonance condition is not fully satisfied. Under unfavorable conditions, the signal resulting from this power broadening of the spin-zero resonance may completely mask the desired  $F = I$  Zeeman signal.

In the case of  $\text{Cd}^{109}$  we have observed three low-field Zeeman signals at  $g_F/g_J = 0.28, 0.11$ , and  $0.40$ . These observations unambiguously show that the nuclear spin is  $\frac{5}{2}$ , in agreement with the earlier assignment based partially on the  $\log ft$  value of the nuclear decay.<sup>25</sup> We have further confirmed this value of the spin by observing the intermediate-field splitting of each of the Zeeman resonances (see below).

### E. Intermediate-Field Zeeman Transitions

The  $\text{Cd}^{109}$  hyperfine-structure intervals were determined from the observation of Zeeman transitions in the intermediate-field region where each low-field resonance is separated into two  $F$  components. The resonances were located by sweeping the magnetic field. This procedure avoided the problems associated with wide-band rf circuitry and permitted careful tuning of the rf loop at each frequency. The dipole and

quadrupole interaction constants,  $A$  and  $B$ , were determined by fitting the observed spectrum to the eigenvalues of the Hamiltonian

$$H = A\left(\frac{3}{2}k\right) + B \frac{\left(\frac{3}{4}\right)k(k+1) - I(I+1)J(J+1)}{2I(2I-1)J(2J-1)} + g_J\mu_0\mathbf{J} \cdot \mathbf{H} + g_I\mu_0\mathbf{I} \cdot \mathbf{H}, \quad (6)$$

where  $k = F(F+1) - I(I+1) - J(J+1)$ . The fitting was accomplished numerically with an IBM 7090 computer. The computer program was designed to vary  $A$  and  $B$  until the differences between the observed and theoretical spectra were minimized. In Eq. (6) we have neglected the hyperfine interactions with the neighboring  $^3P_2$  and  $^3P_0$  states. The corrections resulting from these interactions are of the order of 100 ppm and are not important in the present experiment. In analyzing the data we have taken<sup>26</sup>  $g_J = 1.499847(9)$ . The nuclear moments are readily derived from the interaction constants  $A$  and  $B$ . The zero-field hyperfine intervals,  $\Delta_1$  and  $\Delta_2$ , are given by

$$\Delta_1 = \frac{E_{I+1} - E_I}{h} = A(I+1) + \frac{3}{4}B \frac{I+1}{I}, \quad (7a)$$

$$\Delta_2 = \frac{E_I - E_{I-1}}{h} = AI - \left(\frac{3}{4}\right)B \frac{2I+3}{2I-1}, \quad (7b)$$

where  $E_F$  is the energy of the hyperfine level with total angular momentum  $F$ , and  $I$  is the nuclear spin.

Although the data were analyzed in terms of the exact eigenvalues of the Hamiltonian given in Eq. (6), it was convenient in the course of measurement to estimate the hyperfine intervals from the second-order Zeeman effect. Such estimates are useful in locating and identifying new resonances.

The *second-order Zeeman* effect splits each of the low-field resonances into two  $F$  equally spaced components. At a given value of the field the frequency separations,

$$\Delta(F) = [(E_{F,m+1} - E_{F,m}) - (E_{F,m} - E_{F,m-1})]/h$$

are given by

$$\Delta\nu(I+1) = \frac{-2I}{(2I+1)(I+1)^2} \left( \frac{g_J\mu_0 H}{h} \right)^2 \frac{1}{\Delta_1}, \quad (8a)$$

$$\Delta\nu(I) = +2 \frac{I}{(2I+1)(I+1)^2} \left( \frac{g_J\mu_0 H}{h} \right)^2 \frac{1}{\Delta_1} - 2 \frac{I+1}{(2I+1)(I)^2} \left( \frac{g_J\mu_0 H}{h} \right)^2 \frac{1}{\Delta_2}, \quad (8b)$$

$$\Delta\nu(I-1) = 2 \frac{I+1}{(2I+1)(I)^2} \left( \frac{g_J\mu_0 H}{h} \right)^2 \frac{1}{\Delta_2}, \quad (8c)$$

<sup>25</sup> *Nuclear Data Sheets*, compiled by K. Way *et al.* (Printing and Publishing Office, National Academy of Sciences-National Research Council, Washington 25, D. C., 1960).

<sup>26</sup> P. Thaddeus and R. Novick, *Phys. Rev.* **126**, 1774 (1962).

where  $\Delta_1$  and  $\Delta_2$  are the hyperfine intervals defined in Eq. (7); terms of the order of  $g_I/g_J$  have been neglected.

Since the actual experiments are performed with a fixed frequency,  $\nu$ , it is convenient to express the splittings in terms of the field increments,  $\Delta H(F)$ , between adjacent resonances. The signs are chosen so that a positive  $\Delta H$  indicates that the field must be increased to go from an  $F, m-1 \leftrightarrow F, m$  resonance to an  $F, m \leftrightarrow F, m+1$  resonance.

$$\Delta H(I+1) = 2 \frac{I(I+1)}{2I+1} \frac{h\nu^2}{g_J \mu_0 \Delta_1}, \quad (9a)$$

$$\Delta H(I) = 2 \frac{I(I+1)}{2I+1} \left\{ (I+1)^3 \frac{h\nu^2}{g_J \mu_0 \Delta_2} - I^3 \frac{h\nu^2}{g_J \mu_0 \Delta_1} \right\}, \quad (9b)$$

$$\Delta H(I-1) = 2 \frac{I(I+1)}{2I+1} \frac{h\nu^2}{g_J \mu_0 \Delta_2}. \quad (9c)$$

In applying Eqs. (8) and (9), it is essential to remember that they are only valid to second order. In practice, the magnetic fields used may be sufficiently high so that the splittings are unequal, in which case it is essential to use the exact eigenvalues of the Hamiltonian [Eq. (6)] for the determination of the nuclear moments.

The ordering of the hyperfine levels and, hence, the signs of the nuclear moments can be determined from the observation of individual Zeeman resonances with circularly polarized exciting light. A simple example will serve to illustrate this point. If the polarization of

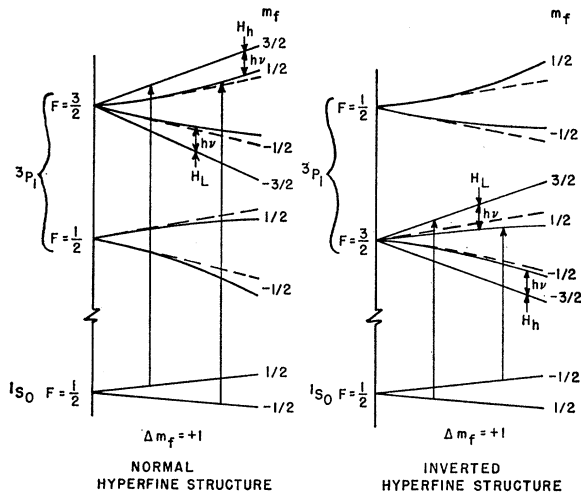


FIG. 6. Determination of the ordering of the hyperfine levels with  $\sigma^+$  circularly polarized exciting light. In the case illustrated ( $I = \frac{1}{2}$ ,  $F = \frac{3}{2}$ ), this illumination results in the elimination of the low-field resonance if the hfs is normal and elimination of the high-field resonance if the levels are inverted.

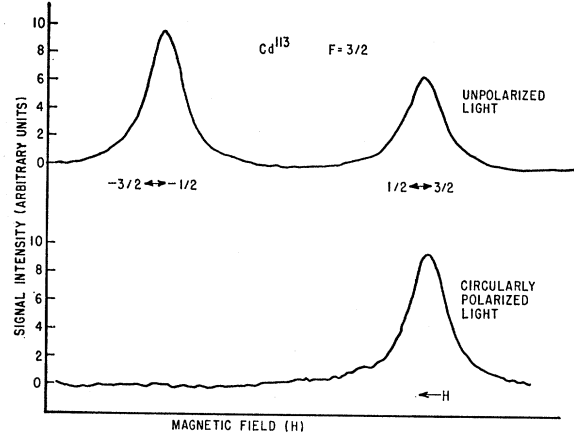


FIG. 7. Intermediate-field Zeeman resonances in the  $F = \frac{3}{2}$  state of  $\text{Cd}^{113}$  as observed with unpolarized and  $\sigma^+$  circularly polarized exciting light. Note that the field increases from right to left.

the exciting light is such that the selection rule  $\Delta m = +1$  is obeyed in excitation ( $\sigma^+$  light), then the states  $^3P_1(F, m_F) = ^3P_1(I+1, -1)$  and  $^3P_1(I+1, -1-1)$  will not be populated and the Zeeman transition between these states will have zero intensity, while the other transitions will have approximately normal intensities. By a similar argument, the  $^3P_1(I+1, I+1)$  and the  $^3P_1(I+1, I)$  Zeeman levels will not be populated with  $\sigma^-$  excitation ( $\Delta m_F = -1$ ). Thus, by using circularly polarized light, we can selectively eliminate and thereby identify the extreme Zeeman resonances in the state  $F = I+1$ .

If the ordering of the hyperfine intervals is normal, the state  $F$  lies above the state  $F-1$ , the  $^3P_1(I+1, -I-1) \leftrightarrow ^3P_1(I+1, -I)$  resonance will occur at a lower value of the magnetic field than the  $^3P_1(I+1, +I+1) \leftrightarrow ^3P_1(I+1, +I)$  resonance. If the levels are inverted, the ordering of the transitions will be interchanged. Since we can identify the resonances by means of polarized light, we can determine the level ordering by observing whether the resonance that disappears with a given polarization is on the high- or low-field side of the Zeeman pattern. For example, if the ordering is normal, the low-field resonance will disappear with  $\sigma^+$  illumination. These effects are illustrated in Figs. 6 and 7. These arguments are further supported by the intensity calculations given below.

## F. Intensities

Theoretical estimates of the intensities of the individual Zeeman transitions are useful in the identification of resonances. Here we will estimate these intensities for both unpolarized and circularly polarized exciting light. For simplicity we will assume that all of the Zeeman components are equally illuminated. Since natural cadmium is used in the lamp, this assumption is not completely valid; however, the relative intensities of neighboring resonances are not greatly modified by

the nonuniformities in the lamp profile. In any case, we lack the detailed knowledge of the lamp profile required for an exact calculation of this effect. We shall also assume that the mean time between the absorption of optical photons by a single atom is long compared to the lifetime of the excited state and to the nuclear spin-relaxation time in the ground state. In most optical double-resonance experiments this requirement is satisfied to a high degree of approximation.

Under the above assumptions, the rate of emission of  $\pi$  light from a given state equals the rate of excitation into the state times the fraction of the spontaneous decays from the state which result in the emission of  $\pi$  light. The signal recorded by the lock-in detector is proportional to the rf-induced change in the rate of  $\pi$  fluorescence. In the case of a transition from the state  $^3P_1(F, M)$  to the state  $^3P_1(F, M-1)$  the signal,  $S$ , is given by

$$S = -k[E(F, M) - E(F, M-1)] \times [\pi(F, M) - \pi(F, M-1)] \times \int_{-\infty}^t \frac{dt_0}{T} e^{-(t-t_0)/T} \frac{|b_{M, M-1}|^2}{(\omega_0 - \omega)^2 + |b_{M, M-1}|^2} \times \sin^2\left\{\frac{1}{2}[(\omega - \omega_0)^2 + |b_{M, M-1}|^2]^{1/2}(t - t_0)\right\}; \quad (10)$$

$k$  is a proportionality constant which depends on the spectral distribution of the lamp output, the apparatus geometry, the number of atoms in the scattering cell, etc. The integral occurring in Eq. (10) can be readily evaluated, yielding

$$S = -\frac{1}{2}k[E(F, M) - E(F, M-1)] \times [\pi(F, M) - \pi(F, M-1)] \times \frac{|b_{M, M-1}|^2}{(\omega - \omega_0)^2 + (1/T^2) + |b_{M, M-1}|^2}. \quad (11)$$

Here  $T$  is the mean lifetime or coherence time of the  $^3P_1$  state,  $\pi(F, M)$  is the square of the dipole matrix element for the emission of  $\pi$  light from the state  $^3P_1(F, M)$ ,  $E(F, M)$  is the relative probability for excitation to the same state,  $b_{MM'}$  is the matrix element of the rf perturbation between the state  $^3P_1(F, M)$  and  $^3P_1(F, M')$ ,  $\omega$  is the applied frequency,  $\omega_0$  is the resonant frequency for the transition  $^3P_1(F, M) \leftrightarrow ^3P_1(F, M-1)$ . We have assumed that the static field is sufficiently large so that the individual Zeeman resonances are separated by many linewidths. If this condition is not satisfied the spectrum is complicated by multiple quantum transitions. The relative  $\pi$  transition probability is given by

$$\pi(F, M) = |\langle I^1 S_0 I M | \mathbf{r} | I^3 P_1 F M \rangle|^2, \quad (12)$$

where  $\mathbf{r}$  is the electron coordinate. In the case of unpolarized exciting light (propagating along the field),

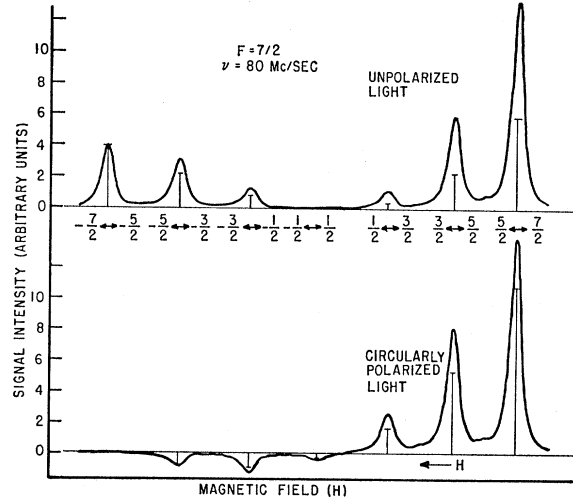


Fig. 8. Intermediate-field Zeeman resonances in the  $F = \frac{7}{2}$  state of  $\text{Cd}^{109}$  as observed with unpolarized and  $\sigma^+$  circularly polarized exciting light. The vertical bars indicate the theoretical intensities normalized to the high-field resonance (Table III). Note that the field increases from right to left.

the relative excitation probability is

$$E_u(F, M) = |\langle I^1 S_0 I M - 1 | \mathbf{r} | I^3 P_1 F M \rangle|^2 + |\langle I^1 S_0 I M + 1 | \mathbf{r} | I^3 P_1 F M \rangle|^2. \quad (13)$$

In the case of  $\sigma^+$  circularly polarized excitation,  $E(F, M)$  becomes

$$E_+(F, M) = |\langle I^1 S_0 I M - 1 | \mathbf{r} | I^3 P_1 F M \rangle|^2. \quad (14)$$

Since we are primarily concerned with transitions in the intermediate field region, it is essential that we allow for the field dependence of the matrix elements. An exact treatment of this effect would require the computation of the eigenvectors of the Hamiltonian [Eq. (6)] at each value of the magnetic field. Such an effort is not justified by the present problem. We can determine the trend of the matrix elements with first-order perturbation theory. In particular, we show with this perturbation treatment that the intensities of the  $^3P_1(F, -\frac{1}{2}) \leftrightarrow ^3P_1(F, +\frac{1}{2})$  transition obtained with circularly polarized light are proportional to the magnetic field. This is in agreement with our observations (see Fig. 8) and contrary to the prediction of zero intensity obtained with the zero-field matrix elements.

The dipole matrix elements including the first-order effect of the magnetic field are listed in Table I.

If we assume that the rf field intensity is great enough to saturate each of the resonances, ( $b_{M, M-1}^2 \gg 1/T^2$ ) then the relative intensities at resonance are given by the product of the first two terms in Eq. (11), since the third term reduces to unity. In the case of unpolarized excitation, we can take advantage of one of the well-known sum rules to show that

$$[E_u(F, M) - E_u(F, M-1)] = -[\pi(F, M) - \pi(F, M-1)]. \quad (15)$$



TABLE I. Electric dipole matrix elements including the first-order field-dependent corrections.<sup>a</sup>

$\langle I^1 S_0 I M \mp 1   \mathbf{P}   I^3 P_1 I + 1 M \rangle = (\mp) \frac{ P }{[(I+1)(2I+1)]^{1/2}} \left[ \frac{(I \pm M)(I+1 \pm M)}{2} \right]^{1/2} \left\{ \frac{I+1 \mp M}{I+1} \right\} \hat{e}_{\pm}$
$\langle I^1 S_0 I M   \mathbf{P}   I^3 P_1 I + 1 M \rangle = \frac{ P }{[(I+1)(2I+1)]^{1/2}} [(I+1+M)(I+1-M)]^{1/2} \left\{ 1 - x_1 \frac{M}{I+1} \right\} \hat{K}$
$\langle I^1 S_0 I M \mp 1   \mathbf{P}   I^3 P_1 I M \rangle = \frac{ P }{[(I)(I+1)]^{1/2}} \left[ \frac{(I+1 \mp M)(I \pm M)}{2} \right]^{1/2} \left\{ 1 \mp x_1 \frac{I}{(2I+1)(I+1)} (I+1 \pm M) \pm x_2 \frac{I+1}{(2I+1)(I)} (I \mp M) \right\} \hat{e}_{\pm}$
$\langle I^1 S_0 I M   \mathbf{P}   I^3 P_1 I M \rangle = \frac{ P }{[(I)(I+1)]^{1/2}} \left\{ M + x_1 \frac{I}{(2I+1)(I+1)} [(I+1)^2 - M^2] + x_2 \frac{I+1}{(I)(2I+1)} [I^2 - M^2] \right\} \hat{K}$
$\langle I^1 S_0 I M \mp 1   \mathbf{P}   I^3 P_1 I - 1 M \rangle = (\mp) \frac{ P }{[(I)(2I+1)]^{1/2}} \left[ \frac{(I+1 \mp M)(I \mp M)}{2} \right]^{1/2} \left\{ 1 \mp x_2 \frac{I \pm M}{I} \right\} \hat{e}_{\pm}$
$\langle I^1 S_0 I M   \mathbf{P}   I^3 P_1 I - 1 M \rangle = (-) \frac{ P }{[(I)(2I+1)]^{1/2}} [I^2 - M^2]^{1/2} \left\{ 1 - x_2 \frac{M}{I} \right\} \hat{K}$

<sup>a</sup>  $\hat{e}_{\pm} = (\hat{i} \pm i\hat{j})/\sqrt{2}$ ;  $\hat{e}_- = (\hat{i} - i\hat{j})/\sqrt{2}$ ;  $x_1 = (g_J - g_I)\mu_0 H / (E_{I+1} - E_I)$ ;  $x_2 = (g_J - g_I)\mu_0 H / (E_I - E_{I-1})$ ;  $|P|^2 = |\langle I^1 S_0 | \mathbf{P} | I^3 P_1 \rangle|^2 = 3\hbar\lambda^3 / [32\pi^3 r(I^3 P_1)]$ .

Thus, the saturated intensities  $S(\infty)$  obtained with unpolarized light,  $S_u(\infty)$ , are given by

$$S_u(\infty) = \frac{1}{2}k[\pi(F, M) - \pi(F, M-1)]^2 \quad (16)$$

or

$$S_u(\infty) = \frac{1}{2}k[|\langle I^1 S_0 I M | \mathbf{r} | I^3 P_1 F M \rangle|^2 - |\langle I^1 S_0 I M - 1 | \mathbf{r} | I^3 P_1 F M - 1 \rangle|^2]^2. \quad (17)$$

Similarly, the saturated intensities observed with  $\sigma^+$  circularly polarized light,  $S_c(\infty)$ , are

$$S_c(\infty) = -\frac{1}{2}k[|\langle I^1 S_0 I M - 1 | \mathbf{r} | I^3 P_1 F M \rangle|^2 - |\langle I^1 S_0 I M - 2 | \mathbf{r} | I^3 P_1 F M - 1 \rangle|^2] \times [|\langle I^1 S_0 I M | \mathbf{r} | I^3 P_1 F M \rangle|^2 - |\langle I^1 S_0 I M - 1 | \mathbf{r} | I^3 P_1 F M - 1 \rangle|^2]. \quad (18)$$

The saturated intensities are listed in Table II for each of the possible hyperfine states. These are based on the matrix elements given in Table I.

### G. $\text{Cd}^{109}$ Hyperfine Intervals

Intermediate-field Zeeman resonances in the  $^3P_1 F = \frac{7}{2}$ ,  $\frac{5}{2}$ , and  $\frac{3}{2}$  states of  $\text{Cd}^{109}$  are shown in Figs. 8, 9, and 10. These were obtained with both unpolarized light and circularly polarized light. The multiplicity of lines confirms the spin and level assignments. The sense of the circularly polarized light was determined from the known properties of the mica quarter-wave plate and confirmed by observing the intermediate-field Zeeman resonances in the  $F = \frac{3}{2}$  state of  $\text{Cd}^{113}$  (see Fig. 7). We determined that the light was polarized so as to produce  $\Delta m = +1$  transitions in absorption and, hence, that the  $\text{Cd}^{109}$  hyperfine levels were inverted. The theoretical intensities of the individual resonances are listed in Table III and compared with the experimental values

in Figs. 8, 9, and 10. The reversal of the sign of some of the signals and the appearance of the  $m = +\frac{1}{2} \leftrightarrow m = -\frac{1}{2}$  resonances are noteworthy and serve to confirm our interpretation of the spectra.

The general decrease in the intensities of the observed lines as the field is increased results from magnetic scanning. Stable cadmium was used in the light source; its spectrum does not coincide exactly with that of  $\text{Cd}^{109}$  and the various hyperfine levels receive different illumination. As the magnetic field is increased, certain of the  $\text{Cd}^{109}$  Zeeman levels move to regions of greater illumination and others to weaker illumination, resulting in the asymmetrical patterns shown in Figs. 8, 9, and 10.

The hyperfine interaction constants were derived

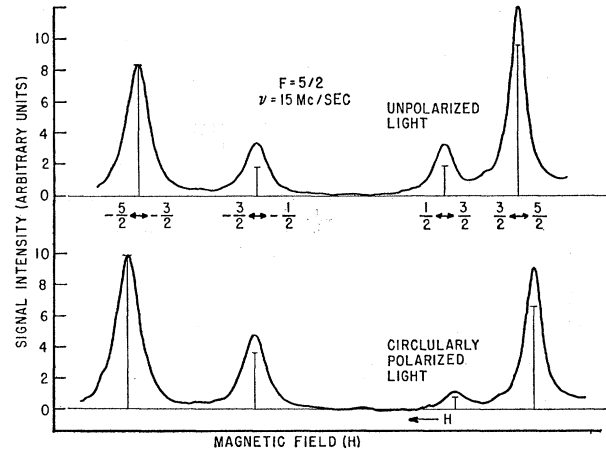


FIG. 9. Intermediate-field Zeeman resonances in the  $F = \frac{5}{2}$  state of  $\text{Cd}^{109}$  as observed with unpolarized and  $\sigma^+$  circularly polarized exciting light. The vertical bars indicate the theoretical intensities normalized to the high-field resonance (Table III). Note that the field increases from right to left.

TABLE II. Intensities of Zeeman transitions between the states ( $^3P_1FM$ ) and ( $^3P_1FM-1$ ) with first-order field corrections.<sup>a</sup>

<i>Unpolarized exciting light</i>	
$S_u(I+1) = \frac{1}{2} \frac{ P ^4}{(I+1)^2(2I+1)^2} \left\{ (2M-1)^2 + \frac{4x_1}{I+1} (2M-1)[I(I+2)-3M(M-1)] \right\}$	
$S_u(I) = \frac{1}{2} \frac{ P ^4}{(I)^2(I+1)^2} \left\{ (2M-1)^2 + 4x_1 \frac{I}{(I+1)(2I+1)} (2M-1)[I(I+2)-3M(M-1)] \right.$	$\left. + 4x_2 \frac{I+1}{(I)(2I+1)} (2M-1)[(I+1)(I-1)-3M(M-1)] \right\}$
$S_u(I-1) = \frac{1}{2} \frac{ P ^4}{I^2(2I+1)^2} \left\{ (2M-1)^2 + \frac{4x_2}{I} (2M-1)[(I+1)(I-1)-3M(M-1)] \right\}$	
<i>Circularly polarized exciting light</i>	
$S_c(I+1) = \frac{1}{2} \frac{ P ^4}{(I+1)^2(2I+1)^2} \left\{ (I+M)(2M-1) + x_1 \frac{I+M}{I+1} [(2M-1)(I+3-3M) + 2I(I+2)-6M(M-1)] \right\}$	
$S_c(I) = \frac{1}{2} \frac{ P ^4}{(I)^2(I+1)^2} \left\{ (M-1)(2M-1) + x_1 \frac{I}{(I+1)(2I+1)} [(2M-1)(I+M)(I+3-3M) + 2I(I+2)(M-1)-6M(M-1)^2] \right.$	$\left. + x_2 \frac{I+1}{I(2I+1)} [(2M-1)(I-2+3M)(I+1-M) + 2(I+1)(I-1)(M-1)-6M(M-1)^2] \right\}$
$S_c(I-1) = \frac{1}{2} \frac{ P ^4}{(I)^2(2I+1)^2} \left\{ (2M-1)(M-1-I) + x_2 \frac{M-1-I}{I} [(2-I-3M)(2M-1) + 2(I+1)(I-1)-6M(M-1)] \right\}$	

<sup>a</sup>  $S(F)$  = signal strength for the transition ( $^3P_1FM$ )  $\leftrightarrow$  ( $^3P_1FM-1$ ).  $x_1$ ,  $x_2$ , and  $|P|$  are defined in Table I.

from precise measurements on eight of the individual Zeeman resonances. Each resonance was observed in more than one field in the general range of 80 to 220 G. The data were analyzed in terms of the Hamiltonian given in Eq. (6). The interaction constants were adjusted for a "best fit" to the observed spectrum (see above). The results are

$$\begin{aligned} A_{109} &= -1,148.6 \pm 2.0 \text{ Mc/sec,} \\ B_{109} &= -167.3 \pm 2.0 \text{ Mc/sec.} \end{aligned} \quad (19)$$

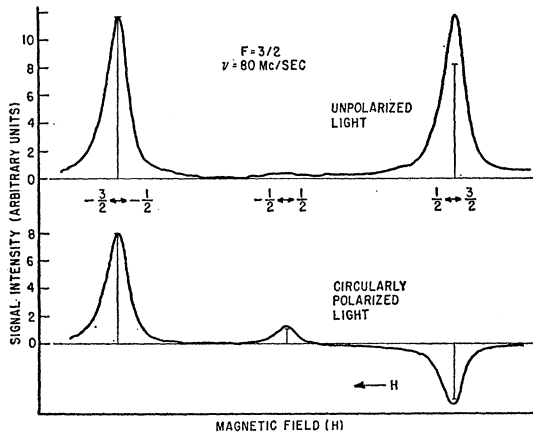


FIG. 10. Intermediate-field Zeeman resonances in the  $F=3/2$  state of  $\text{Cd}^{109}$  as observed with unpolarized and  $\sigma^+$  circularly polarized exciting light. The vertical bars indicate the theoretical intensities normalized to the high-field resonance (Table III). Note that the field increases from right to left.

The errors quoted are three times the standard deviation for each constant.

### III. NUCLEAR MOMENTS

#### A. Magnetic Dipole Moment

The magnetic moment can be obtained to within the effects of a possible hyperfine anomaly by assuming the ratio of the nuclear  $g$  factors of  $\text{Cd}^{109}$  and  $\text{Cd}^{111}$  is equal to the ratio of  $A_{109}$  to  $A_{111}$ .

The  $\text{Cd}^{111}$  moment may be obtained from the results of Proctor and Yu,<sup>27</sup> Lindström,<sup>28</sup> and Boyne and Franken.<sup>29</sup> Combining these results with an estimated net diamagnetic correction of +0.475% for cadmium relative to protons in water, we find  $\mu_{111} = -0.59501(8) \mu_N$ . The dipole interaction constant ( $A_{111}$ ) in the  $(5s5p)^3P_1$  state of  $\text{Cd}^{111}$  has been determined to be  $A_{111} = -4123.81(1) \text{ Mc/sec}$  by optical double resonance.<sup>30</sup> Using these values and our value for  $A_{109}$  we find

$$\mu_{109} = 5(A_{109}/A_{111})\mu_{111} = -0.8286(15) \mu_N. \quad (20)$$

The uncertainty quoted for the moment is based solely

<sup>27</sup> W. G. Proctor and F. C. Yu, Phys. Rev. **79**, 35 (1950).

<sup>28</sup> G. Lindström, Arkiv Fysik **4**, 1 (1951).

<sup>29</sup> H. S. Boyne and P. A. Franken, Phys. Rev. **123**, 242 (1961).

<sup>30</sup> R. F. Lacey (private communication). A preliminary result appears in Quarterly Progress Report, Research Laboratory of Electronics, Massachusetts Institute of Technology, 1959, p. 49 (unpublished).

TABLE III. Theoretical intensities for intermediate-field Zeeman transitions in  $\text{Cd}^{109}$ ,<sup>a,b</sup>

Transitions:	$\frac{7}{2} \leftrightarrow \frac{5}{2}$	$\frac{5}{2} \leftrightarrow \frac{3}{2}$	$\frac{3}{2} \leftrightarrow \frac{1}{2}$	$\frac{1}{2} \leftrightarrow -\frac{1}{2}$	$-\frac{1}{2} \leftrightarrow -\frac{3}{2}$	$-\frac{3}{2} \leftrightarrow -\frac{5}{2}$	$-\frac{5}{2} \leftrightarrow -\frac{7}{2}$
			<i>Unpolarized excitation</i>				
$F=\frac{7}{2}$	0.0975	0.0364	0.0059	0.0000	0.0123	0.0364	0.0662
$F=\frac{5}{2}$		0.224	0.0443	0.0000	0.0602	0.195	
$F=\frac{3}{2}$			0.0147	0.0000	0.0209		
			<i>Circularly polarized excitation</i>				
$F=\frac{7}{2}$	+0.0975	+0.0474	+0.0148	-0.0032	-0.0095	-0.0073	0.0000
$F=\frac{5}{2}$		+0.0785	+0.0091	+0.0013	+0.0418	+0.116	
$F=\frac{3}{2}$			-0.0191	+0.0049	+0.0378		

<sup>a</sup> The field-dependent corrections have been evaluated at the fields appropriate to the data shown in Figs. 8, 9, and 10.<sup>b</sup> A negative intensity indicates that the  $\pi$  radiation *decreases* when a Zeeman transition occurs.

on the uncertainty in  $A_{109}$  and contains no estimate of effects due to a possible hyperfine anomaly.

### B. Quadrupole Moment

The problem of deriving the quadrupole moment from the value of the quadrupole interaction reduces in all cases to an evaluation of  $\langle 1/r^3 \rangle_{\text{av}}$  for the interacting electrons. In the case of the alkaline earths, it is first necessary to estimate the degree of mixing of the pure  $LS$  (or  $jj$ ) states. If we neglect possible configuration mixing we can obtain the coupling coefficients from the observed lifetimes of the  $^3P_1$  and  $^1P_1$  states. Since the radiative lifetime of the pure  $^3P_1$  state is extremely long, it is very sensitive to a slight admixture of the short-lived  $^1P_1$  state. If we let  $\psi(^3P_1')$  be the wave function for the state designated as  $^3P_1$  in cadmium, then

$$\psi(^3P_1') = \alpha\psi(^3P_1^0) + \beta\psi(^1P_1^0) = c_1(\frac{1}{2}, \frac{3}{2})_1 + c_2(\frac{1}{2}, \frac{1}{2})_1, \quad (21)$$

where

$$\frac{\beta^2}{\alpha^2} = \frac{\tau(^1P_1) \lambda^3(^3P_1 - ^1S_0)}{\tau(^3P_1) \lambda^3(^1P_1 - ^1S_0)},$$

$$c_1 = (1/3)^{1/2} \alpha + (2/3)^{1/2} \beta,$$

$$c_2 = (2/3)^{1/2} \alpha - (1/3)^{1/2} \beta.$$

The measured lifetimes are  $\tau(^1P_1) = 1.66(6) \times 10^{-9}$  sec<sup>31</sup> and  $\tau(^3P_1) = 2.43(6) \times 10^{-6}$  sec,<sup>32</sup> and from them we obtain  $c_1 = 0.5405(8)$  and  $c_2 = 0.8413(6)$ .

To obtain  $\langle 1/r^3 \rangle_{\text{av}}$ , we rely on the similarity between the quadrupole interaction and either the magnetic dipole hyperfine-structure interaction or the fine-structure interaction. In the case of the magnetic-dipole interaction, it is first necessary to derive the value of this interaction for the  $p_{3/2}$  electron alone. This can be done by making use of the measured dipole-interaction constants in the  $^3P_2$  and  $^3P_1$  states. Except for slightly altered nomenclature, the following expressions are

taken from Casimir<sup>33</sup>:

$$A(^3P_2) = \frac{1}{4}a_s + \frac{3}{4}a_{3/2}$$

$$A(^3P_1) = \frac{1}{4}(2c_2^2 - c_1^2)a_s + [(5/4)c_1^2 - (5\sqrt{2}/16)(c_1c_2\xi) + (5/2)\theta c_2^2]a_{3/2}. \quad (22)$$

$a_s$  and  $a_{3/2}$  are the dipole interaction constants for  $s_{1/2}$  and  $p_{3/2}$  electrons, respectively.  $\xi$  and  $\theta$  are relativistic correction factors  $\approx 1$  which we have computed following Schwartz.<sup>34,35</sup> Using the following values for the constants of  $\text{Cd}^{111}$ ,  $A(^3P_2) = 3292.936(2)$  Mc/sec,<sup>36</sup>  $A(^3P_1) = -4123.81(1)$  Mc/sec,<sup>30</sup>  $\theta = 1.290(15)$ ,  $\xi = 1.034(10)$ , and  $c_1$  and  $c_2$  as given above, we obtain  $a_{3/2}(\text{Cd}^{111}) = -265(23)$  Mc/sec. Kopfermann<sup>37</sup> shows that

$$a_{3/2} = \frac{16}{15} \frac{m}{M} g_I \mu_0^2 \langle 1/r^3 \rangle_{\text{av}} F_{3/2}, \quad (23)$$

where  $m/M$  is the electron to proton mass ratio and  $F_{3/2}$  is a relativistic correction factor. Taking  $g_I(111) = -1.190$ , we obtain

$$\langle 1/r^3 \rangle_{\text{av}} = 4.37(\pm 0.38)/(a_0^3 F_{3/2}). \quad (24)$$

The evaluation of  $\langle 1/r^3 \rangle_{\text{av}}$  from the fine structure is complicated by the fact that, for two-electron spectra, spin-other-orbit, spin-spin, and other terms in the Breit Hamiltonian besides the spin-orbit interaction may become important. It is clear from the term separation in the  $5s5p$  configuration of cadmium, that the ordinary intermediate coupling theory which neglects these additional effects is not valid. An approximate allowance for the spin-other-orbit interaction has been made by Wolfe.<sup>38</sup> Following the Wolfe method we find the spin-orbit parameter for the  $p$  electron,<sup>37</sup>

<sup>33</sup> H. B. G. Casimir, *On the Interaction Between Atomic Nuclei and Electrons* (Teyler's Tweede Genootschap, Haarlem, 1936).

<sup>34</sup> C. Schwartz, Phys. Rev. **97**, 380 (1955).

<sup>35</sup> C. Schwartz, Phys. Rev. **105**, 173 (1957).

<sup>36</sup> W. Faust, M. McDermott, and W. Lichten, Phys. Rev. **120**, 469 (1960).

<sup>37</sup> H. Kopfermann, *Nuclear Moments* (Academic Press Inc., New York, 1958), 2nd ed.

<sup>38</sup> H. C. Wolfe, Phys. Rev. **41**, 443 (1932).

<sup>39</sup> See, for example, E. M. Condon and G. H. Shortley, *The Theory of Atomic Spectra* (Cambridge University Press, Cambridge, 1957), Chap. 4, p. 122.

<sup>31</sup> A. Lurio and R. Novick, reported at the Second International Conference on Optical Pumping, Heidelberg, Germany, 1962 (unpublished).

<sup>32</sup> F. Byron, M. McDermott, and R. Novick, Bull. Am. Phys. Soc. **7**, 26 (1962).

$\zeta_p = 1004 \text{ cm}^{-1}$ . Kopfermann<sup>37</sup> gives  $\zeta_p = a_0^3 \alpha^2 R H Z^*$   $\langle 1/r^3 \rangle_{\text{av}}$ , where  $Z^*$  is the effective nuclear charge, roughly equal to  $Z-2$  for a  $p$  electron,  $H$  is a relativistic correction factor  $\approx 1$ , and the other factors have their usual meaning. With  $Z^*=46$ , the value of  $\langle 1/r^3 \rangle_{\text{av}}$  derived from the fine structure is  $\langle 1/r^3 \rangle_{\text{av}} = 3.73/(a_0^3 H)$ . The uncertainty in this estimate cannot be evaluated easily, and we have not attempted to do so.

The relationship between the measured quadrupole interaction constant,  $B$ , and the quadrupole interaction for a  $p_{3/2}$  electron,  $b_{3/2}$ , is given by  $B = [(c_1^2)/2 - c_1 c_2 \sqrt{2} \eta] b_{3/2}$ .<sup>33</sup>  $\eta$  is a factor defined by Schwartz<sup>34</sup> and has the value  $\eta = 1.094(10)$  for the  $^3P_1$  state of cadmium. It follows that  $b_{3/2} = 300(4) \text{ Mc/sec}$ . This, in turn, is related to  $Q$  by  $b_{3/2}(\text{Mc/sec}) = \frac{2}{5} (e^2/h) \langle 1/r^3 \rangle_{\text{av}} R Q \times 10^{-6}$ , where  $R$  is another relativistic correction factor. The value of  $Q$  obtained from the hyperfine structure is  $Q = 0.71 \text{ b}$  for  $F/R = 0.966$ . If an extrapolation of a numerical calculation performed by Schwartz<sup>35</sup> for indium is made, a value of  $H/R = 1.02$  is obtained. The Casimir value for  $H/R = 0.958$ . We then obtain  $Q = 0.87 \text{ b}$  ( $H/R = 1.02$ ) or  $Q = 0.82 \text{ b}$  ( $H/R = 0.958$ ), from the estimate based on the fine structure. To arrive at a final value, we arbitrarily take an average of the three estimates with a weight of 2 assigned to the hfs result. The final result is  $Q = 0.78(10) \text{ b}$ . The uncertainty given is large enough to include all the foregoing estimates.

#### IV. DISCUSSION

##### A. Magnetic Moment

The most consistently successful nuclear model which has been used for deriving magnetic moments in the mass region around cadmium ( $A \approx 112$ ) is the configuration mixing model of Arima and Horie.<sup>40</sup> The application of this model to  $\text{Cd}^{109}$  results in a prediction for the magnetic moment which agrees quite well with our measured value. The ground-state neutron configuration is most likely  $(1g_{7/2})^6 (2d_{5/2})^5$  while the proton configuration is undoubtedly  $(g_{9/2})^8$ . The excitations which we have considered are  $(1g_{9/2})^{10} (1g_{7/2})^6 \rightarrow (1g_{9/2})^9 \times (1g_{7/2})^7$  and  $(2d_{5/2})^5 \rightarrow (2d_{5/2})^4 (2d_{3/2})^1$  for the neu-

trons and  $(1g_{9/2})^8 \rightarrow (1g_{9/2})^7 (1g_{7/2})^1$  for the protons. If the interaction between nucleons is assumed to have a  $\delta$ -function behavior and the energy level spacings of Arima and Horie<sup>40</sup> are employed, a moment of  $\mu = -0.78 \mu_N$  is obtained. This is to be compared with our value for the moment of  $\mu = -0.8286 \mu_N$ . On the other hand, if an harmonic oscillator potential, characterized by the parameter  $C = 30$  as in Noya, Arima, and Horie<sup>41</sup> is used, the moment is predicted to be  $\mu = -0.76 \mu_N$ .

It is interesting to compare this moment with that for the 247-keV state in  $\text{Cd}^{111}$  which presumably also belongs to the  $(2d_{5/2})^5$  configuration. This has been measured<sup>42</sup> to be  $\mu = -0.783(28) \mu_N$ . The slightly smaller value contrasts with the observation<sup>43</sup> that the addition of two neutrons without a spin change usually pushes the moment nearer to the Schmidt value.

We have also calculated a quadrupole moment based on the configuration mixing model<sup>41</sup> with a harmonic oscillator potential for the nucleons. If  $C$  is taken to be  $C = 30 \text{ MeV}$  the quadrupole moment calculated is  $Q_{\text{calc}} = +0.10 \text{ b}$ , to be compared to the experimental value  $Q_{\text{exp}} = +0.78 \text{ b}$ . A value of  $Q_{\text{calc}} = 0.12 \text{ b}$  is obtained for  $C = 40 \text{ MeV}$ . The failure of the model to predict a reasonable value of  $Q$  is almost certainly attributable to the onset of collective effects. These do not strongly affect the magnetic moment.

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<sup>41</sup> H. Noya, A. Arima, and H. Horie, *Progr. Theoret. Phys. (Kyoto) Suppl.* **8**, 33 (1958).

<sup>42</sup> R. M. Steffen and W. Zobel, *Phys. Rev.* **103**, 126 (1956).

<sup>43</sup> A. de-Shalit, *Phys. Rev.* **91**, 1479 (1953). See, also, Table 6, p. 63 of Ref. 37.

<sup>40</sup> A. Arima and H. Horie, *Progr. Theoret. Phys. (Kyoto)* **12**, 623 (1954).