Run	${\scriptstyle \scriptstyle {\scriptstyle {f  u}_{+}}\ { m (Mc/sec)}}$	$\nu_{-}$ (Mc/sec)	$     \nu_+ - \nu $ (Mc/sec)	H (G)	μ <i>ι</i> (nm)
1, 2 3, 4 5, 6	1148.29805(25) 1148.29795(25) 1148.29815(25)	$\begin{array}{c} 1148.13395(25)\\ 1148.13425(25)\\ 1148.13405(25)\end{array}$	$\begin{array}{c} 0.16410(35) \\ 0.16370(35) \\ 0.16410(35) \end{array}$	$\begin{array}{c} 188.806(40) \\ 188.801(40) \\ 188.665(50) \end{array}$	$\begin{array}{r} -1.1402(25) \\ -1.1374(25) \\ -1.1395(25) \end{array}$
				Average value: $\mu_I = -1.1395(30)^{a}$	

TABLE II. Data analysis.

a Value does not include diamagnetic correction.

isotope. The results are

# $^{39}\Delta^{42} = +0.20(25)\%$ $^{40}\Delta^{42} = -0.25(25)\%$ , $^{41}\Delta^{42} = +0.42(25)\%$

These results do not serve as a critical test of the current theories of the distribution of nuclear magnetism.

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# Spin and Nuclear Moments of 245-Day Zn<sup>65</sup>; Redetermination of the hfs of $Zn^{67}$ and $\tau({}^{3}P_{1})$ of Zinc\*

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The hfs of the  $(4s4p)^{3}P_{1}$  state of 245-day Zn<sup>65</sup> and of Zn<sup>67</sup> has been determined by the optical doubleresonance technique. The nuclear spin I, and the hfs splittings are:  $Zn^{65}$ : I = 5/2,  $\nu(7/2-5/2) = 1875.475(6)$ Mc/sec,  $\nu(5/2-3/2) = 1334.123(6)$  Mc/sec;  $Zn^{67}$ :  $\nu(7/2-5/2) = 2111.300(3)$  Mc/sec,  $\nu(5/2-3/2)$ = 1551.565(4) Mc/sec. The hfs coupling constants, corrected to second order for interaction with the  ${}^{3}P_{2}$  and  ${}^{3}P_{0}$  states, are: Zn<sup>65</sup>: A(65) = +535.163(2) Mc/sec, B(65) = +2.870(5) Mc/sec; Zn<sup>67</sup>: A(67) = +609.086(2)Mc/sec, B(67) = -18.782(8) Mc/sec. If quadrupole shielding effects are neglected, the corresponding moments of Zn<sup>65</sup> are:  $\mu(65) = +0.7692(2) \mu_N$  and  $\hat{Q}(65) = -0.024(2)$  b. The value given for  $\mu(65)$  includes an estimated correction of 0.09(8)% for the hfs anomaly. The ratio of the Zn65 and Zn67 quadrupole moments is Q(65)/Q(67) = -0.1528(3); this result is independent of the shielding corrections. The Zn<sup>65</sup> spin and magnetic moment are consistent with a  $(2p_{3/2})^4(1f_{5/2})^3$  neutron assignment with some configuration mixing. The small quadrupole moment is compatible with the zero moment expected for a half-filled f shell. In the course of this work, the lifetime of the (4s4p)  ${}^{3}P_{1}$  state of zinc was redetermined and found to be  $\tau({}^{3}P_{1}) = 20(2) \mu$ sec. This result includes a large correction for the effects of wall collisions. The theory of wall broadening of optical double-resonance lines is developed.

#### I. INTRODUCTION

HIS is the third paper in a series devoted to the nuclear spins and moments of the radioactive isotopes of the group II elements.<sup>1,2</sup> The  $(ns^2)$  <sup>1</sup>S<sub>0</sub>

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atomic ground state of these elements is diamagnetic and exhibits no hfs. The paramagnetic, metastable (nsnp) <sup>3</sup> $P_1$  state can be readily studied by optical double resonance and exhibits both magnetic dipole and electric quadrupole hfs. Previously we have reported on Cd<sup>109</sup> (Ref. 1) and Cd<sup>107</sup> (Ref. 2). Here we report on 245-day Zn65.

The determination of the nuclear moments in a series of odd neutron Group II isotopes will complement the large body of information that already exists on odd proton nuclei.

The neutron assignments in  $Zn^{65}$  and  $Zn^{67}$  are  $(2p_{3/2})^4$  $(1f_{5/2})^3$  and  $(2p_{3/2})^4(1f_{5/2})^5$ , respectively. The proton as-

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signment is  $(2p_{3/2})^2$ ; the remainder of the nucleons form closed twenty-eight particle neutron and proton shells. The half-filled f shell in Zn<sup>65</sup> should exhibit a zero quadrupole moment while the single f hole in Zn<sup>67</sup> produces a large positive quadrupole deformation.

In the course of level-crossing studies in Zn<sup>65</sup> and  $Zn^{67}$  it became apparent that the  $Zn^{67}(4s4p)$  <sup>3</sup> $P_1$  hfs intervals were not known with sufficient precision.3 These were originally measured by Böckmann et al.4 In addition, precise knowledge of the  ${}^{3}P_{1}$  natural radiative lifetime is necessary for the full interpretation of both level-crossing and double-resonance work. Previous measurements of  $\tau({}^{3}P_{1})$ ,  $\tau({}^{1}P_{1})$ , and their ratio were inconsistent.<sup>5–7</sup> The Zn<sup>67</sup>( $^{3}P_{1}$ ) hfs and  $\tau(^{3}P_{1})$  were redetermined as part of this work, and  $\tau({}^{1}P_{1})$  was determined by level crossing and is reported elsewhere.<sup>3</sup>

#### II. THEORY OF WALL BROADENING OF OPTICAL DOUBLE-RESONANCE LINES

To determine  $\tau({}^{3}P_{1})$  accurately from the doubleresonance linewidth we must correct for wall collision broadening. In previous work<sup>8</sup> this correction was small and approximation methods were used. For the present case the corrections are much larger, and an exact treatment is required. The order of magnitude of wall effects can be obtained from the following simple physical argument. A typical zinc atom travels a distance  $\bar{v}\tau$  in a lifetime, where  $\bar{v}$  is the average velocity. Therefore, approximately half of those atoms within  $\bar{v}\tau$  of the walls collide with the walls and are lost, assuming complete quenching on the walls. If there is a uniform distribution of excited atoms in the cell, and if  $\bar{v}\tau$  is much less than D, the diameter of the spherical cell, the fraction of atoms quenched is  $A\bar{v}\tau/2V$ , where A is the surface area and V is the volume. For a spherical cell this is just  $3\overline{v}\tau/D$ . In zinc  $\overline{v}\approx 4\times 10^4$  cm/sec and  $\tau\approx 2\times 10^{-5}$  sec, so  $\bar{v}\tau \approx 0.8$  cm as compared with  $\bar{v}\tau \approx 0.07$  cm in cadmium. Thus, for the size cells used (2–3 cm in diam)  $3\bar{v}\tau/D\approx 1$ for zinc, and the effect of wall collisions is substantial. An exact theory of wall broadening will now be developed.

Let  $T_W$  be the time for a particular excited atom to reach the walls, assuming straight line motion; i.e., because of the low density,  $n = 10^{12}$  atoms/cc, we neglect atomic collisions which alter the path of an atom. The expression for the double-resonance signal can be written, in analogy with the expression of Bitter and



FIG. 1. Coordinate system for evaluating the effect of wall collisions.

Brossel,9

$$I(\omega) = K \left\langle \int_{0}^{T_{w}} P(F, m, m', t) e^{-t/\tau} \frac{dt}{\tau} \right\rangle_{\mathrm{av}}, \qquad (1)$$

where the average is taken over all cell parameters, as well as over velocity. The function P(F,m,m',t) is the probability that an atom, initially in the state (F,m)will at time t find itself in the state (F,m') under the influence of an rf magnetic field of strength  $H_1$  and frequency  $\omega$  in the plane perpendicular to the static field  $\hat{\mathbf{H}}_{0.10}$  The rate of creation of excited atoms in the cell is  $K = nV\phi\sigma_{opt}$ , where n is the density of zinc,  $\phi$  is the photon flux falling on the cell, and  $\sigma_{opt}$  is the cross section for the absorption of a photon. If one assumes a uniform distribution of excited atoms in the cell, then the probability of finding a particle in  $d\mathbf{V}$  at  $(r,\theta,\phi)$ , see Fig. 1, is just  $r^2 \sin\theta dr d\theta d\phi / V$ , and the probability of having its velocity directed into dA is just  $dA \cos\theta/4\pi r^2$ . Noting that in the coordinate system of Fig. 1 the equation of a sphere is  $r = D \cos\theta$ ,  $0 \le \theta \le \pi/2$ , we obtain

$$I(\omega) = K \int_{0}^{\infty} f(v) dv \int_{A} \int_{0}^{2\pi} \int_{0}^{\pi/2} \int_{0}^{D} \frac{\cos\theta}{\int_{0}^{r/v}} P e^{-t/\tau} \times \frac{dt}{\tau} \frac{dA \cos\theta}{4\pi r^{2}} \frac{r^{2} \sin\theta dr d\theta d\phi}{V}, \quad (2)$$

<sup>&</sup>lt;sup>8</sup> A. Landman and R. Novick, Phys. Rev. 134, A56 (1964).

<sup>&</sup>lt;sup>4</sup> V. K. Böckmann, H. Krüger, and E. Rechnagel, Ann. Physik 20, 250 (1957).

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<sup>&</sup>lt;sup>7</sup> A. Filippov, Sow. Phys. 1, 289 (1932). <sup>8</sup> F. W. Byron, Jr., M. N. McDermott, and R. Novick, Phys. Rev. (to be published).

<sup>&</sup>lt;sup>9</sup> F. Bitter and J. Brossel, Phys. Rev. 86, 308 (1952). <sup>10</sup> E. Majorana, Nuovo Cimento 9, 43 (1932).

since  $T_W = r/v$ . Setting  $x = \cos\theta$  and performing the trivial integrations on dA and  $d\phi$ , we get, after some obvious changes of variable  $(y=t/\tau, z=r/v\tau)$ ,

$$I(\omega) = K \int_{0}^{\infty} f(v) dv \left(\frac{3v\tau}{D}\right) \int_{0}^{1} x dx$$
$$\times \int_{0}^{Dx/v\tau} dz \int_{0}^{z} P e^{-y} dy, \quad (3)$$

where  $f(v) = (4/\pi^{1/2}v_p^3)v^2 \exp(-v^2/v_p^2)$  is the Maxwellian velocity distribution with  $v_p = (2RT/M)^{1/2}$ .

In terms of the dimensionless variables  $B = \gamma H_1 \tau$ ,  $\delta = (\omega - \gamma H_0)\tau$ ,  $p^2 = B^2 + \delta^2$ , and  $y = t/\tau$ ,  $P(\frac{1}{2}, \frac{1}{2}, -\frac{1}{2}, t)$  and P(1,0,1,t) can be written as

$$P(\frac{1}{2}, \frac{1}{2}, -\frac{1}{2}, t) = (B^2/p^2) \sin^2 \frac{1}{2} py,$$
  

$$P(1,0,1,t) = 2(\delta^2/p^2) P(\frac{1}{2}, \frac{1}{2}, -\frac{1}{2}, t)$$
  

$$+ \frac{1}{2} (B^2/p^2) P(\frac{1}{2}, \frac{1}{2}, -\frac{1}{2}, 2t). \quad (4)$$

Thus, using an obvious notation for the corresponding

$$I(\omega),$$

$$I_{1/2}(p) = K \frac{B^2}{p^2} \int_0^\infty f(v) dv \left(\frac{3v\tau}{D}\right) \int_0^1 x dx$$

$$\times \int_0^{Dx/v\tau} dz \int_0^z e^{-y} \sin^2\frac{1}{2}py dy, \quad (5)$$

and it is a simple matter to show that

$$I_1(p) = (2\delta^2/p^2)I_{1/2}(p) + (2B^2/p^2)I_{1/2}(2p).$$
(6)

Thus it is only necessary to do one integral to obtain the signal in both cases. Integrating Eq. (5) by parts several times, we find

$$I_{1/2}(p) = \frac{KB^2}{2p^2} \int_0^\infty f(v)dv$$
$$\times \int_0^{D/v\tau} \left[ 2 - 3\left(\frac{v\tau}{D}\right)x + \left(\frac{v\tau}{D}\right)^3 x^3 \right] e^{-x} \sin^2\frac{1}{2}pxdx, \quad (7)$$

which is manifestly positive, as it must be. Using this expression it is trivial to demonstrate, by differentiating  $I_{1/2}(p)$  with respect to  $\tau/D$ , that as  $\tau/D$  increases, the intensity always decreases, as is required on physical grounds. A straightforward integration of Eq. (7) yields

$$I_{1/2}(p) = \frac{KB^2}{2(p^2+1)} \int_0^\infty f(v) dv \left\{ 1 - \frac{3}{2} \frac{v\tau}{D} + 3\left(\frac{v\tau}{D}\right)^3 - 3\frac{v\tau}{D} \frac{1}{p^2+1} + 3\left[\frac{1}{p^2+1} + 3\left[\frac{1}{p^2+1} + \frac{8}{(p^2+1)^3}\right]\left(\frac{v\tau}{D}\right)^3 - 3\left[\frac{2}{p(p^2+1)^2}\left(\sin\frac{pD}{v\tau} + p\cos\frac{pD}{v\tau}\right) + \frac{p^2-1}{p^2(p^2+1)^2}\left(\cos\frac{pD}{v\tau} - p\sin\frac{pD}{v\tau}\right) + \frac{p^2+1}{p^2}\right]\left(\frac{v\tau}{D}\right)^2 e^{-D/v\tau} + 3\left[\frac{2}{p(p^2+1)^3}\left((p^2-1)\sin\frac{pD}{v\tau} - 2p\cos\frac{pD}{v\tau}\right) + \frac{p^2-1}{p^2(p^2+1)^3}\left((p^2-1)\cos\frac{pD}{v\tau} + 2p\sin\frac{pD}{v\tau}\right) - \frac{p^2+1}{p^2}\right]\left(\frac{v\tau}{D}\right)^3 e^{-D/v\tau}\right\}, \quad (8)$$

and the related expression

$$I_{1/2}(0) = \frac{KB^2}{2} \int_0^\infty f(v) dv \left\{ 1 - \frac{9}{2} \frac{v\tau}{D} + 30 \left(\frac{v\tau}{D}\right)^3 - 3 \left[ 10 \left(\frac{v\tau}{D}\right)^3 + 10 \left(\frac{v\tau}{D}\right)^2 + \frac{7}{2} \left(\frac{v\tau}{D}\right) + \frac{1}{2} \right] e^{-D/v\tau} \right\}.$$
 (9)

The remaining integrals cannot be evaluated in terms of simple functions. To determine the half-width we must solve the equation  $I_1(\delta_{1/2}) = \frac{1}{2}I_1(0)$ , which is equivalent to

$$I_{1/2}(\delta_{1/2}) = \frac{1}{2} I_{1/2}(0) , \qquad (10a)$$

in the limit of small rf amplitude (*B* tends to zero). The lifetime measurements were made with small rf amplitude and extrapolated to zero. Detailed calculations show that for a fairly large region of p values (the region, of course, depending on  $v\tau/D)I_{1/2}(p)$  can be satisfac-

torily approximated by evaluating the velocity average integrals exactly in the terms where  $\exp(-D/v\tau)$  does not appear, and elsewhere by setting  $v=\bar{v}$ . This gives, for example,

$$I_{1/2}(0) = \frac{KB^2}{2} \left\{ 1 - \frac{9}{2} \frac{\bar{v}\tau}{D} + 15\pi \left(\frac{\bar{v}\tau}{D}\right)^3 - 3 \left[ 10 \left(\frac{\bar{v}\tau}{D}\right)^3 + 10 \left(\frac{\bar{v}\tau}{D}\right)^2 + \frac{7}{2} \frac{\bar{v}\tau}{D} + \frac{1}{2} \right] \exp(-D/\bar{v}\tau) \right\}.$$
 (11)

An approximation of this form was made by Dumont.<sup>5</sup> However, great care must be taken lest it turn out that what is a satisfactory approximation to  $I_{1/2}(p)$  near the half-maximum intensity points is quite unsatisfactory for computing  $I_{1/2}(0)$ , thus introducing important errors in determining the half-width.

This approximation is clearly exact for  $\bar{v}\tau/D\ll 1$ . In this case the exponentials are negligible, and to lowest

 $KB^2/$ 

order in  $\bar{v}\tau/D$ 

$$I_{1/2}(p) = \frac{KB^2}{2(p^2+1)} \left\{ 1 - \frac{3}{2} \frac{\bar{v}\tau}{D} - \frac{3}{D} \frac{\bar{v}\tau}{p^2+1} \right\},$$
$$I_{1/2}(0) = \frac{KB^2}{2} \left\{ 1 - \frac{9}{2} \frac{\bar{v}\tau}{D} \right\}.$$

and

Of course, if 
$$\bar{v}\tau/D=0$$
, then we get just  $I_{1/2}(p)=KB^2/2(p^2+1)$ , the well-known Bitter-Brossel result. In this

case, the half-width is readily found to be

$$\Delta_{1/2} = \frac{1}{\pi \tau} \left( 1 + \frac{3 \bar{v} \tau}{2D} \right),$$

which is in exact agreement with Dumont's result in this limit for a cubical cell; namely,

$$\Delta_{1/2} = \frac{1}{\pi \tau} \left( 1 + \frac{3 \bar{v} \tau}{2L} \right),$$

where L is the length of the side of the cube. This is certainly reasonable, since for  $\bar{v}\tau/D$  much less than typical cell dimensions, one would expect by the argument given at the beginning of this section that  $\Delta_{1/2}$ should have the form

$$\Delta_{1/2} = \frac{1}{\pi \tau} \left( 1 + \alpha \frac{A \bar{v} \tau}{V} \right),$$

where  $\alpha$  is a geometry-independent constant. Since A/V is equal to 6/D for a sphere and 6/L for a cube, we see that  $\alpha = \frac{1}{4}$  for both a cubical and spherical cell.

In the case of zinc  $\bar{v}\tau = 0.8$  cm, and we require an exact numerical solution of Eq. (10a). To obtain it, it is useful to rewrite Eqs. (8) and (9). If we let  $x = v_{p}\tau/D$ and  $a = \pi \Delta_{1/2} D/v_p$ , where  $\Delta_{1/2}$  is the half-width in cps at zero rf field, the equation to be solved becomes

$$I_{1/2}(a,x) - \frac{1}{2}I_{1/2}(0,x) = 0, \qquad (10b)$$

where

$$I_{1/2}(a,x) = \frac{2KB^2}{(\pi)^{1/2}} \frac{1}{[(ax)^2 + 1]} \left[ \frac{(\pi)^{1/2}}{4} - \frac{3}{4}x + 3x^3 - \frac{3}{2}x \frac{1}{(ax)^2 + 1} + 3x^3 \left\{ \frac{1}{(ax)^2 + 1} + \frac{8}{[(ax)^2 + 1]^3} \right\} - 3\left\{ \frac{3 - (ax)^2}{(ax)[(ax)^2 + 1]^2} s_2(a,x) + \frac{3(ax)^2 - 1}{(ax)^2[(ax)^2 + 1]^2} c_2(a,x) + \left[ \frac{1}{(ax)^2} + 1 \right] f_2(x) \right\} + 3\left\{ \frac{4[(ax)^2 - 1]}{ax[(ax)^2 + 1]^3} s_3(a,x) + \frac{[(ax)^2 - 2ax - 1][(ax)^2 + 2ax - 1]}{(ax)^2[(ax)^2 + 1]^3} c_3(a,x) - \left[ \frac{1}{(ax)^2} + 1 \right] f_3(x) \right\} \right].$$
(12)

$$I_{1/2}(0,x) = \frac{2KB^2}{(\pi)^{1/2}} \left\{ \frac{(\pi)^{1/2}}{4} - \frac{9}{4} + 30x^3 - 3\left[10f_3(x) + 10f_2(x) + \frac{7}{2}f_1(x) + \frac{1}{2}f_0(x)\right] \right\}.$$
(13)

The functions  $f_n(x)$ ,  $s_n(a,x)$ , and  $c_n(a,x)$  are defined by one of the values actually found experimentally, we get

$$f_n(x) = x^n \int_0^\infty e^{-1/(xy)} e^{-y^2} y^{n+2} dy, \qquad (14a)$$

$$s_n(a,x) = x^n \int_0^\infty \frac{a}{y} e^{-y^2} y^{n+2} dy, \quad (14b)$$

$$c_n(a,x) = x^n \int_0^\infty \frac{a}{y} \cos^2 e^{-1/(xy)} e^{-y^2} y^{n+2} dy.$$
(14c)

Thus, one takes the measured value of  $\Delta_{1/2}$ , calculates *a* from it (knowing, of course, D and  $v_p$ ) and solves Eq. (10b) numerically to obtain x. Tabulated values of  $f_n(x)$ ,  $s_n(a,x)$ , and  $c_n(a,x)$ , obtained with the aid of a computer, were used. This then gives  $\tau$ .

In the light of what has been said concerning approximation techniques, it is interesting to compare  $I_{1/2}(0)$ calculated from Eq. (13) with  $I_{1/2}(0)$  calculated by using the approximation given in Eq. (11). For x=0.29,

$$\{I_{1/2}(0)\}_{\text{exact}} = \frac{KB^2}{2}(0.20), \quad \{I_{1/2}(0)\}_{\text{approx}} = \frac{KB^2}{2}(0.74),$$

whereas the difference between  $\{I_{1/2}(p)\}_{exact}$  and  $\{I_{1/2}(p)\}_{approx}$  for some intermediate value like p=2will only be a few percent. This justifies our previous remark that care must be used in solving Eq. (10b) for x by approximate methods, and the exact equations are required for the evaluation of the zinc  ${}^{3}P_{1}$  lifetime.

#### **III. EXPERIMENTAL**

#### A. Sensitivity

The principal difference between this experiment and the previous ones on cadmium arises from the fact that the lifetime of the  ${}^{3}P_{1}$  state in zinc is about eight times longer than the corresponding lifetime in cadmium and is comparable to the time required for an atom to traverse the resonance cell.

A50

A51

The fact that many of the excited atoms strike the walls before they have an opportunity to radiate produces both line broadening and a reduction in sensitivity. From Eqs. (12) to (14) it is simple to show that the collisions reduce the saturated signal by a factor of about one-half of what one would get with  $\bar{v}\tau/D=0$ . Using the known lifetimes,  $\tau(Zn) = 20(2) \times 10^{-6}$  sec and  $\tau$ (Cd) = 2.39(4)×10<sup>-6</sup> sec,<sup>8</sup> we find that the effective cross section for photon absorption in zinc is a factor of ten smaller than the corresponding cadmium cross section. The over-all effect of the wall collisions and the reduced cross section is to increase the minimum number of detectable atoms by a factor of twenty relative to cadmium.

The light sources used in this work were constructed and operated in the same manner as the cadmium lamps reported in Ref. 1. Under the best operating conditions the intensity of the zinc lamp in the center of the 3076 Å intercombination line is about a factor of 2.5 less than the corresponding output of the best cadmium lamp.<sup>1</sup> The signal-to-noise ratio in optical double-resonance experiments is limited by the fluctuations in the shot noise resulting from instrumentally scattered light.<sup>1</sup> This ratio varies as the square root of the light intensity.

The net effect of the light source, wall collisions, and reduced cross section is a thirty-twofold increase in the minimum number of detectable zinc atoms relative to cadmium. In practice we have found that with both the even (I=0) and odd (I=5/2) stable zinc isotopes there is a five-hundredfold increase in the minimum number of detectable zinc atoms relative to cadmium. The reason for this additional factor of sixteen is not understood.

#### B. Production and Identification of Zn<sup>65</sup>

The isotope production and scattering cell preparation techniques are identical with those described previously.2 High-purity copper was bombarded with 11-MeV protons for 400 µA-h, producing 245-day Zn<sup>65</sup> by the reaction  $Cu^{65}(p,n)Zn^{65}$ . It was hoped that this procedure would yield a sample free of stable zinc. Counting measurements indicated that 4.3 mCi, or  $2.6 \times 10^{15}$  atoms of Zn<sup>65</sup>, were produced, in good agreement with the yield calculated from (p,n) cross sections given by Wing and Huizenga.<sup>11</sup> Approximately 70% of the activity was successfully distilled into the quartz resonance cell. In spite of our precautions an analysis of the double-resonance spectrum indicated that the sample was contaminated with twenty times as much stable zinc as 245-day Zn<sup>65</sup>.

The chemical identity of the sample is assured by resonance fluorescence.<sup>1</sup> Only 38-min Zn<sup>63</sup> and 245-day Zn<sup>65</sup> are produced in the irradiation, and the Zn<sup>63</sup> is allowed to decay in the cyclotron. There are no other long-lived odd-mass isotopes of zinc except stable Zn<sup>67</sup>. Double-resonance lines arising from this isotope can be identified by comparing the observed spectrum with that obtained from a sample of stable material. Observations of the spectrum assigned to Zn<sup>65</sup> over a period of months indicated a long half-life.

# C. Double-Resonance Apparatus and Method

The apparatus was essentially the same as that used previously,<sup>1,2</sup> with the following modifications. Helmholtz coils capable of producing 500 G were installed, and a Polacoat polarizer<sup>12</sup> was used in the detection arm since this material has better transmission than Polaroid HN38 at the zinc intercombination line. As before, unpolarized light directed along the field was used to excite the atoms; this produced only  $\sigma$  excitation  $(\Delta M = \pm 1)$ . The detector polarizer was oriented to accept only  $\pi$  light.

Initial observations of Zeeman transitions in Zn<sup>65</sup> indicated that the quadrupole interaction was of the order of a few megacycles, and that Zeeman observations would not suffice to determine B(65) adequately. We, therefore, undertook to observe direct  $(\Delta F = 1)$ hyperfine transitions. In both the Zeeman and highfrequency measurements the rf magnetic field is oriented at right angles to the constant magnetic field, so that only  $\Delta M = \pm 1$  transitions are effected. The high-frequency transitions were identified by their Zeeman effect. Transitions were observed in pairs  $[F,M] \leftrightarrow [F', (M \pm 1)] \text{ and } [F,M] \leftrightarrow [F', -(M \pm 1)].$ The members of these pairs have equal and opposite linear Zeeman displacements.

In the direct measurement of the hfs intervals an Airborne Instruments Corp. Type-124A cavity oscillator, cathode modulated at 280 cps, was used as a source of radio-frequency power. The frequency was measured with a Hewlett-Packard Model-524C cycle counter used in conjunction with a Model-508B transfer oscillator. The same high-frequency apparatus was used in the redetermination of the Zn<sup>67</sup> hfs.

As in the case of cadmium<sup>8</sup> the lifetime of the  ${}^{3}P_{1}$ state was determined by observing the width of Zeeman transitions in the even isotopes. This is discussed in greater detail in the following section.

#### IV. OBSERVATIONS

#### A. Zn<sup>65</sup> Spin and Hyperfine Intervals

Low-field Zeeman transitions were observed in the Zn<sup>65</sup> cell at fields corresponding to  $g_F/g_J = 2/7$ , 4/35, 2/5 and no others. Such a Zeeman spectrum can only be produced by spin 5/2 nuclei. Unfortunately, stable  $Zn^{67}$  has I=5/2, and it was necessary to extend the observations into the intermediate-field region to separate the Zn<sup>65</sup> and Zn<sup>67</sup> spectra. At magnetic fields of about 100 G the individual Zeeman transitions were

<sup>&</sup>lt;sup>11</sup> J. Wing and J. R. Huizenga, Phys. Rev. 128, 280 (1962).

<sup>&</sup>lt;sup>12</sup> M. N. McDermott and R. Novick, J. Opt. Soc. Am. 51, 1008 (1961).



FIG. 2. Intermediate-field Zeeman transitions in the  $F = \frac{7}{2}, F = \frac{5}{2}$ , and  $F = \frac{3}{2}$  states of Zn<sup>65</sup>.

resolved. At this field twice as many resonances were observed as could be accounted for by  $Zn^{67}$  alone, which implied the presence of a second isotope with spin 5/2. The position of the extra resonances suggested a hfs about 10% smaller than that of  $Zn^{67}$ . These results definitely establish a nuclear spin, I=5/2, for  $Zn^{65}$  in agreement with the earlier assignment based on the log ft value of the nuclear decay.<sup>13</sup>

Transitions observed with circularly polarized light,<sup>1,2</sup> indicated that the Zn<sup>65</sup> hfs is normal. Thus, the nuclear magnetic moment is positive, and the last odd neutron is in a state with  $I = l - \frac{1}{2}$ . This is in agreement with the  $f_{5/2}$  shell model assignment.

Figure 2 shows typical Zeeman transitions in each of the three hyperfine states. The intermediate-field dependence of these transitions gave  $\nu(7/2-5/2) = 1876.3(10)$  Mc/sec, and  $\nu(5/2-3/2) = 1332.6(15)$  Mc/sec. This leads to the following preliminary values of the hyperfine interaction constants: A(65) = +535.1(3) Mc/sec, and B(65) = +3.3(10) Mc/sec. It is apparent that B(65) is too small to be reliably determined by observation of intermediate-field Zeeman transitions. In addition second-order perturbations of the hfs from the  ${}^{3}P_{2}$  and  ${}^{3}P_{0}$  fine structure are of the order of 0.5 Mc/sec and must be fully evaluated.

Direct hyperfine measurements were undertaken to obtain a precise value of the quadrupole interaction constant. Precise observations were made on the following pairs of transitions:  $(7/2, \pm 3/2) \leftrightarrow (5/2, \pm 1/2)$  and  $(5/2, \pm 1/2) \leftrightarrow (3/2, \mp 1/2)$ . Each transition was observed at two magnetic fields. The measurements lead to the following values for the zero-field intervals:

$$\nu(7/2-5/2) = 1875.475(6)$$
 Mc/sec,  
 $\nu(5/2-3/2) = 1334.123(6)$  Mc/sec.

The assigned error is twice the standard deviation.

# B. Redetermination of Zn<sup>67</sup> Hyperfine Intervals

The hfs in the  ${}^{3}P_{1}$  state of Zn<sup>67</sup> has been measured by Böckmann, Krüger, and Rechnagel<sup>4</sup> with an estimated precision of about 0.1 Mc/sec. To determine the atomic constants in the  ${}^{3}P_{1}$  state and the nuclear moments of Zn<sup>65</sup>, more precise values were needed. In this case, the pairs of transitions  $(7/2, \pm 1/2) \leftrightarrow (5/2, \pm 1/2)$  and  $(5/2, \pm 1/2) \leftrightarrow (3/2, \pm 1/2)$  were studied. Each transition was observed at three different fields. The measurements give for the zero-field intervals in Zn<sup>67</sup>:

$$\nu(7/2-5/2) = 2111.300(3)$$
 Mc/sec,  
 $\nu(5/2-3/2) = 1551.565(4)$  Mc/sec.

The assigned error is twice the standard deviation. The results are more precise than but consistent with the previous values obtained by Böckmann *et al.*:  $\nu(7/2-5/2)=2111.13(12)$  Mc/sec and  $\nu(5/2-3/2)=1551.54(10)$  Mc/sec.<sup>4</sup> Figure 3 shows typical hyperfine transition resonances for Zn<sup>65</sup> and Zn<sup>67</sup>.

## C. Lifetime of the ${}^{3}P_{1}$ State

Before an analysis of the second-order hfs corrections for interaction with the  ${}^{3}P_{2}$  and  ${}^{3}P_{0}$  fine structure states can be made, an accurate knowledge of the natural radiative lifetime of the  $(4s4p){}^{3}P_{1}$  state is necessary.<sup>14</sup> This can be accomplished by observing the width of the I=0 Zeeman resonance. Unfortunately, the lifetime



FIG. 3. Low-field hyperfine transitions in Zn<sup>65</sup> and Zn<sup>67</sup>.

<sup>14</sup> A. Lurio, M. Mandel, and R. Novick, Phys. Rev. **126**, 1758 (1962).

<sup>&</sup>lt;sup>13</sup> P. Avignon, Ann. Phys. 1, 10 (1956).

is so long that the width of these resonances is doubled by wall collisions, and we must make substantial correction for this effect. Coherence narrowing and atomatom collision broadening were avoided by working at temperatures of 250°C and less. Foreign gas broadening was avoided by using cells sealed at a vacuum of about  $1 \times 10^{-8}$  Torr. Under these conditions the natural linewidth is distorted only by wall collisions and possible field inhomogeneities. With the exception of wall effects, the method of determining the lifetime is the same as that used in cadmium.8 The homogeneity of the static field was checked carefully with a proton nuclear magnetic-resonance magnetometer, and it was found to be uniform to one mG over the cell volume at typical operating fields. The long lifetime of the  ${}^{3}P_{1}$  state insures that there will be considerable motional averaging which will reduce the resulting broadening to less than one kilocycle. The half-width was measured at a static field twice that of the typical operating field, and no significant discrepancy was observed.

A series of measurements at different rf amplitudes were made on three different spherical cells one inch in diameter at a temperature of 234°C. It is necessary to extrapolate the observations to zero rf amplitude to eliminate the effects of rf power broadening. Figure 4 shows a typical plot of the line width *versus* rf amplitude, the intercept of this curve yielding the line width at zero rf field.<sup>8</sup> The half-widths at zero rf amplitude but with wall collision broadening  $\Delta_{1/2}$  for the three spherical cells of inner diameter D are as follows:

$$\begin{aligned} \Delta_{1/2} &= 30.0(12) \text{ kc/sec for } D = 2.48 \text{ cm}, \\ \Delta_{1/2} &= 29.6(6) \text{ kc/sec for } D = 2.40 \text{ cm}, \\ \Delta_{1/2} &= 28.4(6) \text{ kc/sec for } D = 2.48 \text{ cm}. \end{aligned}$$

These results were averaged to yield a combined result of

$$\Delta_{1/2} = 29.3(8)$$
 kc/sec for  $D = 2.44(4)$  cm.

The theory of wall collision broadening has been developed in Sec. II. Unfortunately, the theory yields a complicated equation in which the natural lifetime occurs implicitly, and it is necessary to resort to numerical calculations. The parameters of the theory are  $x=v_p\tau/D$  and  $a=\pi\Delta_{1/2}D/v_p$ , where  $v_p=(2RT/M)^{1/2}$  is the Maxwellian most probable velocity. In the present case of a 2.44 cm diam cell at 507°K,  $v_p=3.59\times10^4$  cm/sec and a=6.27(21). The integrals which appear in the equation for x were calculated numerically on an IBM 7090 computer for the range  $0.20 \le x \le 0.60$  in steps of 0.01. The numerical values of these functions are substituted into Eq. (10b), which is then solved numerically. This leads to x=0.287 and  $\tau(^{3}P_{1})=1.95\times10^{-5}$  sec.

To check the validity of the wall collision corrections, measurements were made on a 1.93-cm-diam cell at a temperature of 244°C. The half-width was found to be  $\Delta_{1/2}=32.2(20)$  kc/sec. Thus,  $v_p=3.63\times10^4$  cm/sec and



FIG. 4. Rf broadening of the spin-zero Zeeman resonance in stable zinc as observed in a 2.48-cm diam bulb. For the purpose of extrapolation the square of the half-width is plotted against the square of the rf amplitude.

a=5.36(32). Carrying out the same analysis as before we find x=0.378 and  $\tau=2.01\times10^{-5}$  sec, in good agreement with the value for the larger cell.

The final value for the natural radiative lifetime of the  $(4s4p)^{3}P_{1}$  state of zinc is  $\tau=2.0(2)\times10^{-5}$  sec. The assigned error is twice that arising from the uncertainty in  $\Delta_{1/2}$  to allow for possible incomplete motional averaging of the slight inhomogeneities in the static magnetic field.

This value of the lifetime is in disagreement with the value determined by Dumont<sup>5</sup> who found  $\tau({}^{3}P_{1})$  $\approx 3.2 \times 10^{-5}$  seconds using the double-resonance method. The source of the disagreement is not certain, but we noted in Sec. II that Dumont used a questionable approximation in his calculation to correct for the wall broadening. The ratio of the present result to the lifetime of the lowest  ${}^{1}P_{1}$  state in zinc, determined by Landman<sup>3</sup> to be 1.38(6)×10<sup>-9</sup> sec, yields  $\tau(P_1)/\tau(P_1)$  $=0.70(7)\times10^{-4}$ . This ratio is in agreement with Filippov's measurements' based on the "hook" method, which is expected to be quite accurate for the determination of ratios of this sort. He found  $A({}^{3}P_{1})/A({}^{1}P_{1})$  $= \tau ({}^{1}P_{1}) / \tau ({}^{3}P_{1}) = 0.67 \times 10^{-4}$ . In the case of cadmium where the wall collision corrections are much smaller the corresponding lifetimes have been directly determined by optical resonance techniques,<sup>8,15</sup> and their

<sup>&</sup>lt;sup>15</sup> R. Novick and A. Lurio, Bull. Am. Phys. Soc. 7, 258 (1962).

Constant	Value of constant for zinc		
$c_1$	0.5656(6)		
$c_2$	0.8247(4)		
$\xi$	1.01		
$\eta$	1.03		
heta	1.095		
$(1-\delta)(1-\epsilon)$	0.981		

TABLE I. Constants used in evaluating the secondorder hfs corrections.

ratio is in excellent agreement with Filippov's result.<sup>7</sup> This agreement strengthens our belief that the "hook" method is reliable for the determination of the lifetime ratios.

We also note that at the temperatures used in this measurement,  $T \leq 517^{\circ}$ K, the density *n* was always less than about  $1.5 \times 10^{12}$  atoms/cc. Using a collision broadening cross section<sup>16</sup> of  $\sigma_{coll} = 1.6 \times 10^{-14}$  cm<sup>2</sup>, we find  $n\bar{v}\sigma_{\rm coll}/\pi \approx 400$  cps, i.e., only about 1% of the measured widths, so the effect of collisions can safely be neglected. The cross section for the absorption of a photon is  $\sigma_{\text{opt}} \approx 10^{-15} \text{ cm}^2$ , so  $n\sigma_{\text{opt}}L \approx 4 \times 10^{-3}$  (using L = 2.5 cm, a typical cell dimension). This indicates strongly that we can neglect effects arising from the trapping of radiation, such as coherence narrowing,<sup>17</sup> etc.

#### V. CALCULATION OF HFS INTERACTION CONSTANTS

Off-diagonal interactions between the  ${}^{3}P_{1}$ ,  ${}^{3}P_{2}$ , and  ${}^{3}P_{0}$  states produce a modification of the apparent interaction constants. In particular, the second-order dipole interaction will modify the apparent quadrupole interaction. Since B is of the order of 2 Mc/sec and  $A^2/\Delta E$  is approximately 0.5 Mc/sec, it is clear that we must correct for these effects to determine Q(65)reliably.

The effect of the second-order interaction of the  ${}^{3}P_{2}$ and  ${}^{3}P_{0}$  states on the hfs in the  ${}^{3}P_{1}$  state is calculated by the method of Lurio *et al.*,<sup>14</sup> using an iterative procedure. Calculation of the individual electron interaction constants may be found in Ref. 18. Preliminary values of A and B are calculated neglecting the second-order interaction, corrections based on these values are obtained and applied to A and B, and the procedure is repeated. One iteration was sufficient.

The constants used in evaluating the second-order hfs corrections are given in Table I and are defined in Ref. 14. The constants are taken from Lurio<sup>18</sup> except for  $c_1$  and  $c_2$  which are calculated from  $\tau({}^3P_1)$  determined as part of this work and  $\tau({}^{1}P_{1})$  determined by Landman.<sup>3</sup> The uncorrected and corrected hfs coupling constants are given in Table II. The uncertainties in the corrections are less than the experimental error in

TABLE II. Values of the hfs coupling constants before and after second-order corrections.

hfs	Zn <sup>65</sup>		Zn <sup>67</sup>		
coupling	uncorrected	corrected	uncorrected	corrected	
constant	(MC/Sec)		(MC/sec)		
A	535.117(2)	535.163(2)	609.208(2)	609.086(2)	
B	2.445(4)	2.870(5)	-19.331(7)	-18.782(8)	

A, but increase the error in B slightly. The values are in agreement with the results of level-crossing studies.<sup>3</sup>

#### VI. NUCLEAR MOMENTS OF Zn<sup>65</sup>

The magnetic moment can be determined from A(65)if we neglect the effects of the finite nuclear size. That is, we assume the ratio of the nuclear g factors of  $Zn^{65}$ and  $Zn^{67}$  to be equal to the ratio of A(65) to A(67). In so doing we neglect the hyperfine anomaly defined as

$$\Delta = \{ [A(65)g_I(67)] / [A(67)g_I(65)] \} - 1.$$
(15)

The effect of finite nuclear size appears in two ways. the distribution of charge and the distribution of magnetism. The first effect is sensitive to differences in the size of the nuclei which are not known, and can only be estimated assuming a constant density of nuclear matter. Using the formulas of Breit and Rosenthal,<sup>19</sup> we obtain  $\Delta(B-R) = -0.006\%$ .

The second effect was first estimated by Bohr and Weisskopf,<sup>20</sup> and is severely model-dependent. Using the calculations of Stroke, Blin-Stoyle, and Jaccarino,<sup>21</sup> we find  $\Delta(B-W) = -0.001\%$  for the single-particle model. If we include first-order configuration mixing and use the parameters of Noya, Arima, and Horie,<sup>22</sup> the experimental values of  $\mu(65)$  and  $\mu(67)$ , and the free-nucleon g values,<sup>23</sup> we obtain  $\Delta(B-W) = +0.031\%$ . The final result is then  $\Delta = +0.03(3)\%$ , where we have neglected the Breit-Rosenthal correction and included an uncertainty based solely on the uncertainty of the factors used in the calculation of  $\Delta(B-W)$ .<sup>21,22</sup> In view of the uncertainty in the theoretical estimate of the hfs anomaly we must allow for an uncertainty of at least 0.08% in obtaining the nuclear moment from the observed hfs.

The magnetic moment of Zn<sup>67</sup> may be obtained from the results of Dharmatti and Weaver,24 Ting and Williams,<sup>25</sup> Yasaitis and Smaller,<sup>26</sup> and Boyne and

- <sup>23</sup> This is the most consistent way of performing the calculation [H. H. Stroke (private communication)].
   <sup>24</sup> S. S. Dharmatti and H. E. Weaver, Jr., Phys. Rev. 85, 927
- (1952). <sup>25</sup> Y. Ting and D. Williams, Phys. Rev. 89, 595 (1953). Smaller Phys. Rev. 82, 750 (1953).

  - <sup>26</sup> E. Yasaitis and B. Smaller, Phys. Rev. 82, 750 (1951).

<sup>&</sup>lt;sup>16</sup> F. W. Byron, Jr., and H. M. Foley, Phys. Rev. (to be published).

<sup>&</sup>lt;sup>17</sup> J. P. Barrat, J. Phys. Rad. 20, 541, 633, 657 (1959). 18 A. Lurio, Phys. Rev. 126, 1768 (1962).

 <sup>&</sup>lt;sup>19</sup> G. Breit and J. E. Rosenthal, Phys. Rev. 41, 459 (1932).
 <sup>20</sup> A. Bohr and V. Weisskopf, Phys. Rev. 77, 94 (1950).
 <sup>21</sup> H. Stroke, R. J. Blin-Stoyle, and V. Jaccarino, Phys. Rev. <sup>11</sup> H. SLIOKE, K. J. BIIN-Stoyle, and V. Jaccarino, Phys. Rev.
123, 1326 (1961).
<sup>22</sup> H. Noya, A. Arima, and H. Horie, Progr. Theoret. Phys. (Kyoto) Suppl. 8, 33 (1958).
<sup>23</sup> This is the most consistent wave for the statement of the statemen

Franken.<sup>27</sup> The measurements are combined with a net diamagnetic correction of 0.247(13)% for zinc ions relative to protons in water, calculated by Dickenson,<sup>28</sup> to give  $\mu(67)=0.87575(8)\mu_N$ . It is estimated that the diamagnetic correction is reliable only to about 5%. Thus

$$\mu(65) = \frac{A(65)}{A(67)} \mu(67) \frac{1}{[1.0003(3)]}$$
$$= +0.7692(2)\mu_N.$$

This result includes the calculated hfs anomaly, and the entire uncertainty in the result arises from the uncertainties in the evaluation of the anomaly.

The ratio of the quadrupole moments of  $Zn^{65}$  and  $Zn^{67}$  is just the ratio of the quadrupole coupling constants

$$\frac{Q(\text{Zn}^{65})}{Q(\text{Zn}^{67})} = \frac{B(\text{Zn}^{65})}{B(\text{Zn}^{67})} = -0.1528(3).$$

The calculation of the absolute value of Q is based on our knowledge of  $\langle r^{-3} \rangle_{av}$  for the  $p_{3/2}$  electron. Lurio<sup>18</sup> has estimated this using measurements in the <sup>3</sup> $P_2$  state of Zn<sup>67</sup> and finds  $Q(Zn^{67}) = 0.16b$ . Then

$$Q(\text{Zn}^{65}) = -0.024(2)$$
b.

The quoted error includes the range of values resulting from various estimates of  $\langle r^{-3} \rangle_{av}$ . Shielding corrections have been neglected.

#### VII. DISCUSSION OF THE Zn<sup>65</sup> MOMENTS

The most consistently successful model for calculating the nuclear moments of light and medium mass nuclei is the configuration mixing model of Noya, Arima, and Horie.<sup>22</sup> If we use the value for their nuclear-pairing energy parameter which best fits the nuclear binding energy data in this region, C=30 MeV, we find  $\mu(65)$  $=0.792 \,\mu_N$  for a  $(2p_{3/2})^4 (1f_{5/2})^3$  neutron configuration. This is in excellent but probably somewhat fortuitous agreement with the experimental value, since the same model gives  $\mu(\text{Zn}^{67}) = 1.023 \mu_N$ .

The extreme single-particle model predicts a zero quadrupole moment for odd neutron nuclei. In the configuration mixing model, proton configurations which have a nonvanishing quadrupole moment are mixed into the shell model configuration by the two-particle interactions of the protons with the odd neutrons. The outer neutron shell of Zn<sup>65</sup> is half-filled, all other neutron shells being filled, predicting a zero quadrupole moment, which is in agreement with the very small quadrupole moment observed. If C=30 and  $r_0=1.2\times10^{-13}$  cm are adopted, the configuration mixing model predict Q(67)=0.14b, indicating that collective effects are not important.

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 <sup>&</sup>lt;sup>27</sup> H. S. Boyne and P. A. Franken, Phys. Rev. **123**, 242 (1961).
 <sup>28</sup> W. C. Dickenson, Phys. Rev. **77**, 736 (1950).