

# Theory of Isotope Shift\*

G. BREIT

*Yale University, New Haven, Connecticut*

## I. INTRODUCTION

### (A) Historical

THE theory of the spectroscopic isotope shift is concerned with nuclear sizes only in a very limited sense because the information furnished by the shift has to do only partially with the nuclear size and is more specifically concerned with the values of certain parameters related to changes in nuclear radii and to changes of nuclear shapes.

The early observations of Merton (1919)<sup>1</sup> regarding the relative displacement of lines emitted by samples of Pb of different geophysical origin and therefore with different isotopic constitution are believed to have given the first indication of the effect under discussion. The observed displacement was much too large to be attributed to the nuclear mass correction used successfully by Bohr in his explanation of the differences in the wavelength of nearly coincident lines of hydrogen and helium.

### (B) Mass Effect and Nuclear Charge Distribution Effect

The differences in wavelengths of lines emitted by isotopes of the same element can arise either as a result of the differences in the masses of the isotopes or on account of differences in the nuclear charge distributions.

The mass effect is usually considered as a sum of two parts. The first of these is the well-known Bohr reduced mass correction. The second, known as the Hughes-Eckart effect, depends on the presence of cross-product terms in the momenta of different electrons. While more difficult to evaluate than the Bohr reduced mass correction, the Hughes-Eckart effect<sup>2</sup> is of the same order as the Bohr reduced mass correction. Both contain the ratio  $m/(MA)$  with  $m$  standing for electron mass,  $M$  for nucleon mass, and  $A$  for mass number. The differences in the effect for isotopes  $A$ ,  $A+1$  thus

contains the factor

$$m/MA(A+1)$$

and decreases rapidly as  $A$  increases. The mass effects are important for light elements such as Li but are negligible for heavy elements such as Tl, Pb, and the rare earths. The charge distribution effect is the only one of interest in nuclear physics. It arises because the nuclear charge is distributed through a finite volume. Laboratory measurements have to do with minute changes in the nuclear charge distribution taking place as a result of adding neutrons to the lightest stable isotope. For an element with mass number  $\sim 200$  the estimated change in nuclear radius resulting from addition of one neutron is  $\sim 1/600$  of the nuclear radius, i.e.,  $\sim 2 \times 10^{-15}$  cm. The smallness of this change is the main reason for the necessity of considering various small changes in nuclear shape and structure as other contributing causes of the observed displacements.

### (C) Relationship to Other Hyperfine-Structure Effects

The isotope shift is usually observed in hyperfine-structure studies. The hyperfine structure of spectroscopic lines is caused either by the interaction of electrons with the nuclear spin or by the charge distribution effect. The former is illustrated in Fig. 1 which shows the production of magnetic field  $\mathcal{H}_e$  by an electron current  $C$ . The nuclear magnetic moment  $\mu_N$  is space quantized in this magnetic field. Most atomic levels are split therefore into  $2I_N+1$  levels where  $I_N$  is the nuclear spin. Figure 2 shows how the effect does not occur. The electron spin does not act in the manner pictured; if it did, the splittings would be  $-\frac{1}{2}$  of those observed for  $s$  terms. In this manner the hyperfine structure shows directly the inadequacy of the elementary nonrelativistic spin picture of the electron and the superiority of the description offered by Dirac's equation. Use of the latter is essential for the theory of the spectroscopic isotope shift. In Fig. 3 is shown a typical pattern for the levels of the three most common isotopes of Pb. The horizontal lines represent the levels. The numbers attached to the lines are mass numbers

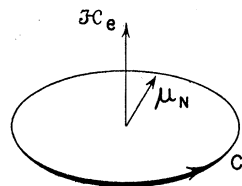


FIG. 1. Illustration of the production of magnetic field  $\mathcal{H}_e$  by current  $C$  caused by the circulation of the Dirac electron. The nuclear magnetic moment  $\mu_N$  is space quantized with respect to the magnetic moment.

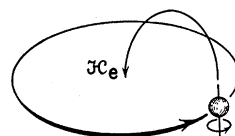


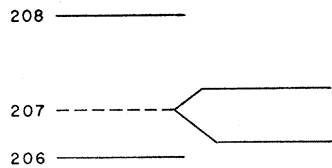
FIG. 2. Production of magnetic field at the nucleus by the magnetic moment of a spinning particle. The direction of the magnetic field at the nucleus is opposite to that of preceding figure. Experimental evidence speaks against the literal employment of the spin picture and favors the Dirac equation.

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<sup>1</sup> T. R. Merton, *Nature* **104**, 406 (1919).

<sup>2</sup> D. S. Hughes and C. Eckart, *Phys. Rev.* **36**, 694 (1930); John P. Vinti, *Phys. Rev.* **56**, 1120 (1939); **58**, 879 (1940).

FIG. 3. Schematic representation of energy levels of the three most abundant isotopes of Pb. The mass numbers of the isotopes are shown at each energy level. The dotted line represents the weighted mean energy of the levels for the odd isotope.



of corresponding isotopes. The dotted line is the weighted mean energy of the levels of the odd isotope. In speaking of the isotope displacement of  $\text{Pb}^{207}$  with respect to  $\text{Pb}^{206}$  and  $\text{Pb}^{208}$  it is usually understood that this weighted mean is used. The statistical weights  $2F+1$  of the solid level lines of  $\text{Pb}^{207}$  in the diagram are used to obtain the weighted mean. The experimentally observed spectral lines are usually interpreted in terms of such diagrams with the adjustment to no isotope shift at ionization. The theoretical justification for taking the weighted mean in the manner described is that first-order perturbation theory of the nuclear spin hyperfine structure shows this to be the correct procedure as may be seen by Slater's method of sums. The assumption that the effect of  $\mu_N$  is small enough to be treated in first order appears to be justifiable theoretically on account of the smallness of  $\mu_N$ . This assumption is supported by the experimental fact that the ratio of isotope displacements such as

$$\frac{\nu(\text{Pb}^{208}) - \nu(\text{Pb}^{207})}{\nu(\text{Pb}^{207}) - \nu(\text{Pb}^{206})}$$

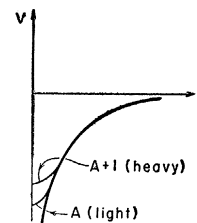
and other similar ratios observed in the spectra of many other elements are derived consistently making use of this assumption independently of the particular atomic level.

## II. NUCLEAR CHARGE DISTRIBUTION EFFECT

### (A) Spherical Nuclei

The effect of distributing the nuclear charge through a finite volume is often referred to as the nuclear volume effect. It has been worked on by Bartlett (1931)<sup>3</sup> who made estimates employing nonscreened and nonrelativistic wave functions. Racah (1932)<sup>4</sup> made relativistic calculations, obtained essentially complete results formally, but vastly overestimated the theoretical value (several hundred times) by employing a somewhat-arbitrary value of electron density at the nucleus. Rosenthal and Breit (1932)<sup>5</sup> and Breit (1932)<sup>6</sup> made more detailed calculations obtaining approximate agreement with experiment, the theoretical effect being usually somewhat greater than the experimental. The

FIG. 4. Modification of the potential energy of an electron in the field of a nucleus caused by the finiteness of the nuclear radius. The thick line refers to a point nucleus; the thin lines give modifications for nuclei of finite radii. The figure shows that the electron energy is expected to increase with the mass number  $A$ .



general nature of the effect is illustrated in Fig. 4 showing the potential energy  $V$  between electron and nucleus plotted against their relative distance  $r$ . The heavy curve represents  $V$  for the case of a nucleus of zero radius. The deviation from this curve for the case of the heavier isotope of mass number  $A+1$  is larger than that for the lighter and the expected position of the energy level is expected to be higher for the heavier isotope. The *observed* isotope shift is only a *small fraction* of the *actual* one since it corresponds to the difference between the effects of the potential-energy curves for the nuclei  $A$  and  $A+1$ .

The change in the energy of a spectroscopic term caused by the nuclear volume effect will be abbreviated as IS and denoted as  $\delta W$ . The change in  $\delta W$  in going from isotope with mass number  $A$  to isotope with mass number  $A+1$  will be denoted by  $\Delta\delta W$  and referred to as RIS, the abbreviation standing for *relative isotope shift*. It is to be understood that the nuclear mass effect is negligible or else that a correction has been made for it.

The calculation of the IS caused by the volume effect cannot be performed quite accurately for all nuclear models by first-order perturbation theory. The reason for this is that the *nuclear field distorts the electron wave function quite appreciably*. The distortion was taken into account in the work of Rosenthal and Breit<sup>5</sup> who obtained a correction factor of the order of  $\frac{1}{2}$  for  $Z=81$  and a Gamow trough. The correction factor has been studied more systematically by Broch<sup>7</sup> and in connection with the isotope shift for x-ray terms by Wertheim and Igo.<sup>8</sup> Since the actual field is less attractive than the Coulomb field, the electron density inside the nucleus is decreased as a result of the wave-function distortion and a change in the nucleus-electron potential caused by an increase in the nuclear radius produces less effect on the electron energy than given by first-order perturbation theory with electron wave functions approximated by those in the field of a nucleus of zero size. The *isotope shift* is therefore *overestimated* if one makes calculations *with undistorted wave functions*.

In IS calculations the potential energy between the nucleus and the electron is usually supposed to be entirely of electrostatic origin. The electron-neutron interaction<sup>9</sup> is equivalent to an attraction of  $\sim 4000$  ev

<sup>3</sup> J. H. Bartlett, *Nature* **128**, 408 (1931).

<sup>4</sup> G. Racah, *Nature* **129**, 723 (1932).

<sup>5</sup> J. E. Rosenthal and G. Breit, *Phys. Rev.* **41**, 459 (1932).

<sup>6</sup> G. Breit, *Phys. Rev.* **42**, 348 (1932).

<sup>7</sup> E. K. Broch, *Arch. Math. Naturvidenskab* **48**, 25 (1945).

<sup>8</sup> M. S. Wertheim and G. Igo, *Phys. Rev.* **98**, 1 (1955).

<sup>9</sup> E. Fermi and L. Marshall, *Phys. Rev.* **72**, 1139 (1947); Rainwater, Rabi, and Havens, *Phys. Rev.* **72**, 634 (1947);

through a distance of  $e^2/mc^2$  while the interaction of the electron with a proton at a distance  $e^2/mc^2$  gives a potential energy of 510 000 ev and the energy is larger inside this distance. These numbers suggest that the electron-neutron interaction has a negligible effect on the RIS. In fact the ratio of the RIS, as usually calculated on the hypothesis of a uniform expansion of the nuclear volume with  $A^{1/3}$  dependence of nuclear radius on mass number, to the direct effect of attraction of electron by neutron is

$$\left| \frac{\Delta\delta W}{\delta_n W} \right| \cong \frac{125Z}{3A} \left( \frac{r mc^2}{e^2} \right)^2,$$

where it has been assumed for simplicity that the nuclear charge is distributed on the nuclear surface and that the newly added neutron is located close to that surface. With the nominal values  $Z=80$ ,  $A=200$ ,  $r=3e^2/mc^2=8.4 \times 10^{-13}$  cm the ratio is 150. The assumption of uniform charge distribution exaggerates the ratio but not seriously enough to make one doubt the insignificance of the effect of the direct electron-neutron interaction.

The *grain structure* of the nuclear charge distribution is customarily *neglected* in the estimates. Similarly changes in the average proton density having the character of a decrease in one range of radial distances compensated by a change in the opposite direction in another range of distances are usually not considered except insofar as they are connected with a general expansion of the nuclear size. The only exception is that of effects of intrinsic quadrupole moments discussed later. Some of the omitted effects doubtless take place on the shell theory of nuclear structure<sup>10</sup> but until recently the theory has not been sufficiently developed to make the calculation of such effects worthwhile.

For a uniform distribution of nuclear charge through a spherical nuclear volume the potential energy curve inside the nucleus is a parabola somewhat as in Fig. 4. It is sometimes easier to make calculations using a constant potential inside the nucleus and assuming continuity of the potential at the nuclear surface. Such a potential corresponds to distributing all of the nuclear charge on the spherical nuclear surface. This model is sometimes called the *top slice model*. It exaggerates the absolute value of the IS. Neglecting wavefunction distortion, the energy change caused by a nuclear potential

$$V_N = \left[ -\frac{n+1}{n} + \frac{1}{n} \left( \frac{r}{r_0} \right)^n \right] \frac{Ze^2}{r_0} \quad (1)$$

L. L. Foldy, Phys. Rev. **83**, 688 (1951); Hamermesh, Ringo, and Wattenberg, Phys. Rev. **85**, 483 (1952); Hughes, Harvey, Goldberg, and Stafne, Phys. Rev. **90**, 497 (1953); Crouch, Krohn, and Ringo, Phys. Rev. **102**, 1321 (1956).

<sup>10</sup> M. G. Mayer, Phys. Rev. **74**, 235 (1948), **75**, 1969 (1949); Haxel, Jensen, and Zuess, Z. Physik **128**, 295 (1950).

for  $s$  terms of one electron has the value

$$\delta W = \frac{4\pi R_\infty}{Z} a_H^3 \psi^2(0) \frac{1+\rho}{\Gamma^2(2\rho+1)} \frac{(n+1)y_0^{2\rho}}{2\rho(2\rho+n+1)(2\rho+1)}, \quad (l=0) \quad (1.1)$$

where

$$y_0 = 2Zr_0/a_H \quad (1.2)$$

and

$$a_H = \hbar^2/me^2 \quad (1.3)$$

is the Bohr radius for hydrogen,  $R_\infty$  is the Rydberg,  $\psi^2(0)$  is the nonrelativistic electron density at the nucleus, and

$$\rho = (1-\alpha^2 Z^2)^{1/2}, \quad (l=0), \quad (1.4)$$

where  $\alpha$  is the fine structure constant. The nuclear potential in Eq. (1) is adjusted to join continuously to the Coulomb potential  $-Ze^2/r$  at  $r=r_0$ . For  $r < r_0$  it is represented by a parabola of  $n$ th order. Uniform charge density corresponds to  $n=2$ , the top slice model to  $n=\infty$ , no change from the point particle potential,  $-Ze^2/r$ , to  $n=-1$ . Relativistic effects enter through the difference  $\rho-1$ . If  $\rho=1$  there is no effect of  $n$  in (1.1). The reason for this is that the nonrelativistic electron density is practically uniform within nuclear dimensions. Only the leading term in an expansion in powers of  $r_0$  is included in (1.1) which is therefore slightly inaccurate. The relativistic effects in (1.1) are large primarily because of the entrance of  $y_0^{2\rho}$  which brings in the factor

$$2\rho y_0^{2\rho-1}$$

in  $\Delta\delta W$ . Since for  $r_0=10^{-12}$  cm and  $Z=80$ ,  $\rho=0.81$  and  $y_0 \cong 300$  there enters the factor  $\sim (300)^{0.62}$  relativistically in comparison with the nonrelativistic factor  $\sim (300)^1$  corresponding to roughly a factor 6 in favor of the relativistic result. The origin of this effect is the steep increase of electron density at small  $r$  which varies as  $r^{2\rho-2}$ . For  $l>0$

$$\delta W = \frac{2k(k-\rho)}{\Gamma^2(2\rho+1)} \frac{(n+1)y_0^{2\rho}}{2\rho(2\rho+n+1)(2\rho+1)} \times \{ [(l+1)^2 - Z^2\alpha^2]^{1/2} - 1 - (l^2 - Z^2\alpha^2)^{1/2} \}^{-1} \delta \quad (2)$$

where  $k = -1, +1, -2, +2, \dots$  for  $s_{1/2}, p_{1/2}, p_{3/2}, d_{3/2}, \dots$  terms,

$$\rho = [k^2 - Z^2\alpha^2]^{1/2} \quad (2.1)$$

and  $\delta$  is the doublet splitting for the particular  $l$ . Relativistic effects on the relation between  $\delta$  and  $\langle r^{-3} \rangle$  are neglected in (2). Since the IS is more readily observed for  $l=0$  the approximations made in deriving (2) do not appear to be very important. For  $k^2 > 1$  the value of  $\rho$  is practically  $|k|$  and the IS is therefore appreciable only for  $s_{1/2}$  and  $p_{1/2}$  terms. Most of the more reliable evidence comes from  $s$  terms.

In order to evaluate (1.1) it is necessary to know  $\psi^2(0)$ . In (2) the corresponding step is not necessary,

the normalization of the wave function being furnished by the empirical value of  $\delta$ . Since the principal evidence regarding IS comes from observations on  $s$  terms the evaluation of  $\psi^2(0)$  is unfortunately necessary. This is sometimes carried out employing some electron screening approximation such as is in the Fermi-Thomas-Hartree or Hartree-Fock models. A less laborious and apparently equally successful method is furnished by the Landé-Goudsmit-Fermi-Segrè formulas. These give<sup>11</sup>

$$\psi^2(0) \cong \frac{Z_i Z_0^2}{\pi a_H^3 (n^*)^3} = 2.16 \times 10^{24} \frac{Z_i Z_0^2}{(n^*)^3} \text{ cm}^{-3}, \quad (3)$$

where  $Z_{ie}$  = internal effective nuclear charge,  $Z_{0e}$  = external atomic charge

$$n^* = n - \Delta$$

is the effective quantum number obtained by equating the term value energy to  $-R_\infty Z_0^2 / (n^*)^2$ , and  $n$  is the principal quantum number while  $\Delta$  is the quantum defect. Equation (3) is similar to that derived by Fermi and Segrè in connection with magnetic hyperfine structure theory. In their result the  $1/n^{*3}$  of Eq. (3) is changed as follows:

$$\frac{1}{n^{*3}} \rightarrow -\frac{d}{2dn} \frac{1}{n^{*2}} = \frac{1}{n^{*3}} \left( 1 - \frac{d\Delta}{dn} \right). \quad (3.1)$$

The considerations of Fermi-Segrè involve the employment of the JWKB approximation for atomic wave functions and cannot claim to establish (3) employing modification (3.1) with complete accuracy. Some work in collaboration with John F. Wild<sup>12</sup> indicates that the formula may in some cases be better without the correction term  $-d\Delta/dn$ . This numerical work was carried out employing Fermi-Thomas fields and integrating the nonrelativistic equation numerically. There is, on the other hand, some empirical evidence in hyperfine-structure measurements supporting the validity of the correction factor  $1 - (d\Delta/dn)$ . This evidence is based on the occurrence of  $\psi^2(0)$  in Fermi's formula for the magnetic coupling of an  $s$  electron to the nuclear spin. Thus F. M. Kelly<sup>13</sup> finds agreement for the magnetic moment of  $^{79}\text{Au}$ <sup>197</sup> to better than 1% from  $6s$  and  $7s$  level of Au I. For the first, the factor  $1 - (d\Delta/dn)$

is 1.424 and for the second 1.038. Other evidence is found in the work of Crawford and Schawlow<sup>14</sup> on isotope shift and that of Schawlow, Hume, and Crawford<sup>15</sup> on the hyperfine structure of Pb<sup>207</sup>. Basing itself on the corrections used in the former of these references which in turn make use of the effect of the finite nuclear size on the magnetic electron-nucleus coupling introduced by Rosenthal and Breit,<sup>5</sup> the second reference reconciles the value  $\mu = 0.588 \pm 0.001$  nm of the magnetic moment of Pb<sup>207</sup> obtained by the nuclear-induction method<sup>16</sup> with the spectroscopic value which without corrections is 18% too low. The correction for electron wave function distortion is 15% and is obtained on the same basis as the correction determined for Tl III. In addition a 3% correction is used for the distribution of the nuclear magnetism through a finite rather than infinitesimal volume.<sup>17</sup> In this way the Goudsmit-Fermi-Segrè formula is in a sense verified through direct measurement of  $\mu$ . The Fermi-Segrè factor  $1 - (d\Delta/dn)$  is 1.16 for  $5d^{10}6s$  of Pb IV. The hyperfine interval factor  $a_{6s}$  has been deduced from the levels of Pb III using the measurements of Crooker<sup>18</sup> and Crawford making allowance for inter-configuration interactions. The accuracy of the interval factor thus derived is estimated to be  $\pm 2\%$ . Other evidence for the validity of the Fermi-Segrè factor  $1 - d\Delta/dn$  is found in the paper by Crawford and Schawlow. Thus for  $^{55}\text{Cs}$ <sup>133</sup> the directly measured nuclear moment  $g$  value,<sup>19,20</sup>  $g(I) = 0.7315$  is 3.9% higher than the value obtained<sup>21</sup> from the hyperfine splitting  $\Delta(\nu_{6s})$  which is 0.7025. When the correction for the change in the electron wave function caused by the nuclear size effect is applied, the corrected hyperfine-structure value becomes  $g(I) = 0.728$  which differs from the directly measured value by 0.4%. For  $6s$  of La III the hyperfine-structure value<sup>22</sup>  $g(I) = 2.65$  is 4.2% lower than the induction-method value,<sup>20</sup> 2.761. The correction for finite nuclear radius is 4.1% reducing the discrepancy to 0.1%. In the case of Tl, Crawford and Schawlow obtain good consistency from different terms of Tl, I, II, III, finding a mean  $\mu = 1.58$  nm as compared with a magnetic-induction value<sup>23</sup> of 1.628 nm. The lowest among the hyperfine values is 1.54 and the highest 1.64. Among these there is available the somewhat rare comparison with a moment based on the

<sup>11</sup> The formulas appear to have been used first for IS calculations in the simple form<sup>6</sup> reproduced in Eq. (3) of the text which makes no correction for  $d\Delta/dn$  and is the same as that used by S. Goudsmit, Phys. Rev. **37**, 663 (1939) who employed Casimir's results for the purely hydrogenic case. The transition to the case of screened electrons was justified somewhat more completely by Breit<sup>6</sup> making use of the relation  $\langle (l+1)r^{-3} \rangle = 2\pi\psi^2(0)$  occurring in hyperfine-structure formulas which were shown to apply to general central fields as in G. Breit, Phys. Rev. **37**, 51 (1931). The inclusion of the factor  $1 - d\Delta/dn$  occurs first in E. Fermi and E. Segrè, Z. Physik **82**, 729 (1933) and the derivation of the factor in E. Fermi and E. Segrè, Mem. reale accad. Italia Classe sci. fis. mate nat. **4**, 131 (1933).

<sup>12</sup> J. F. Wild and G. Breit (to be published); John F. Wild, dissertation, Yale University, 1957.

<sup>13</sup> F. M. Kelly, Proc. Phys. Soc. (London) **A65**, 250 (1952).

<sup>14</sup> M. F. Crawford and A. L. Schawlow, Phys. Rev. **76**, 1310 (1949).

<sup>15</sup> Schawlow, Hume, and Crawford, Phys. Rev. **76**, 1876 (1949).

<sup>16</sup> W. G. Proctor, Phys. Rev. **76**, 684 (1949).

<sup>17</sup> G. Breit and L. A. Wills, Phys. Rev. **44**, 470 (1933); F. Bitter, Phys. Rev. **76**, 150 (1949); H. Kopfermann, *Kernmomente* (Akademische Verlagsgesellschaft, Leipzig, 1940), p. 17; A. Bohr and V. F. Weisskopf, Phys. Rev. **77**, 94 (1950); A. Bohr, Phys. Rev. **81**, 331 (1951).

<sup>18</sup> A. M. Crooker, Can. J. Research **A14**, 115 (1936).

<sup>19</sup> Kusch, Millman, and Rabi, Phys. Rev. **55**, 1176 (1939).

<sup>20</sup> W. H. Chambers and D. Williams, Phys. Rev. **76**, 461 (1949).

<sup>21</sup> S. Millman and P. Kusch, Phys. Rev. **58**, 438 (1940).

<sup>22</sup> M. F. Crawford and N. S. Grace, Phys. Rev. **47**, 536 (1935); H. Wittke, Z. Physik **116**, 547 (1940).

<sup>23</sup> H. L. Poss, Phys. Rev. **72**, 637 (1947).

coupling of a  $6p_{3/2}$  electron which agreed within 2% with the mean from six determinations. Kelly, Kuhn, and Pery<sup>24</sup> have calculated the splitting of the ground state,  $4s_{3/2}$ , of Ca II for the isotope  $^{40}\text{Ca}^{43}$  making use of the values  $g_I = -\mu/I = -0.3758$  nm and of  $I=7/2$  obtained by Jeffries.<sup>25</sup> The splitting measured by them is  $0.109 \pm 0.002$   $\text{cm}^{-1}$  which is 4% smaller than the calculated  $0.113$   $\text{cm}^{-1}$ . They have included the correction for the volume distribution of nuclear charge which is  $\sim 1\%$  and they find that in this case the effect of the volume distribution of nuclear magnetism is negligible. In the isoelectronic spectrum of K calculation gave  $0.0165$   $\text{cm}^{-1}$ ; direct measurement  $0.0154$   $\text{cm}^{-1}$ . There appears to be evidence here that the Goudsmit, Fermi, Segrè formula gives in this case a systematically too-high value of  $\psi^2(0)$ . In Wild's calculations the ratio of the Fermi-Segrè value to the exact one is 1 for the  $11s$  state provided one sets  $Z_i = 52.78$  while for the  $6s$  state the ratio is 1.03 for the same  $Z_i$ . Changing  $Z_i$  to  $Z = 55$ , as is done in most calculations with  $s$  electrons, increases these ratios by 4% making the calculated value 1.07 for  $6s$ . If the factor  $1 - (d\Delta/dn)$  is neglected and  $Z_i = 52.78$  is used, the Goudsmit formula agrees within 0.5% with the directly computed values. The value  $Z_i = 52.78$  fits in with the screening constant  $\sigma_2$  derived from x-ray data. The theoretical numbers were obtained by means of the Fermi-Thomas field and  $n^*$ ,  $\Delta$  are theoretical rather than experimental values. It is not justifiable, therefore, to use the calculated values literally. It appears of interest, however, that the employment of the same approximation on a model of the actual atom gives approximately the same differences as the employment of the molecular-beam or magnetic-induction values for comparison with the observed hyperfine structure. For collections of material on nuclear moments from magnetic hyperfine structure and from direct measurement one may refer to the article by Mack<sup>26</sup> and the books by Kopfermann<sup>27</sup> and Ramsey.<sup>28</sup> A discussion of the effect of  $d\Delta/dn$  in the case of  $\text{Xe}^{129,131}$  is found in the work of A. Bohr, J. Koch, and E. Rasmussen.<sup>29</sup> For accurate work an evaluation of radial integrals and relativistic corrections in hyperfine-structure reference is made to the papers by C. Schwartz<sup>30</sup> and for treatment of data on isotope shift to the work of Crawford, Gray, Kelly, and Schawlow.<sup>31</sup>

Derivation of the Fermi-Segrè formula makes use of the JWKB approximation in order to define a phase

which can be considered as a continuous function of the energy. The JWKB approximation has rather limited accuracy failing for some  $r$  altogether and the derivation is therefore questionable. The whole method is furthermore applicable only to the central field model of the atom. Since the actual atom is a many-body system, there is a further question regarding the soundness of the derivation. The tests by means of hyperfine-structure measurements and their comparison with direct nuclear magnetic moment measurements which have been referred to above are *not wholly convincing* because of the possibility of interconfiguration interactions. These do not affect the isotope shift in quite the same way as they affect the magnetic hyperfine structure coupling especially because of the effect of the screening by the valence electron of inner closed shell and it is not easy and probably impossible therefore to settle the question on a wholly empirical basis. It is possible,<sup>32</sup> however, to extend the Fermi-Segrè derivation in such a way as to avoid both of the assumptions. Instead of dealing with the JWKB phase, the logarithmic derivative of the radial function of the valence electron at  $r=0$  is used. The Coulomb field is rounded-off inside the nucleus. The reciprocal of the logarithmic derivative, the  $\mathcal{R}$  function of Wigner, can be used for the calculation of a phase shift in a problem in which the space is extended to negative  $r$  for the valence electron. For the negative  $r$  the potential energy of the electron is continued with the same negative value as it has at  $r=0$ . In this manner one can borrow the results of nuclear reaction theory without specializing the considerations to one-particle systems. The theorems proved by Wigner<sup>33</sup> regarding  $\mathcal{R}$  functions mean, then, that the phase shift varies monotonically with energy. The kinks in Wigner's curves appear in the spectroscopist's language as perturbations. In between the kinks the results are essentially like those of Fermi and Segrè. This argument shows that *in a statistical sense the Fermi-Segrè results are correct* but it does not go far enough to predict how close one is to a kink and is in this sense qualitative only. On the other hand, it appears to extend the range of applicability of the Fermi-Segrè results. It is understandable from this viewpoint that the  $1 - d\Delta/dn$  correction factor does not always improve the agreement even though at times it improves it very much.

In the calculation of the IS one should be concerned with the change of the energy of the whole electron system rather than with the change of the energy of the valence electron alone. For this reason the effect of closed electron shells cannot be neglected and especially so for closed  $(ns)^2$  configurations.<sup>6</sup> Thus, if a  $6p$  electron is excited to a higher energy state, the screening which it exerts on the  $(6s)^2$  shell changes and the  $6s$  electrons are coupled more closely to the nucleus.

<sup>24</sup> Kelly, Kuhn, and Pery, Proc. Phys. Soc. (London) **A67**, 450 (1954).

<sup>25</sup> C. D. Jeffries, Phys. Rev. **90**, 1130 (1953).

<sup>26</sup> J. E. Mack, Revs. Modern Phys. **22**, 64 (1950).

<sup>27</sup> H. Kopfermann, *Kernmomente* (Akademische Verlagsgesellschaft, Leipzig, 1940).

<sup>28</sup> N. F. Ramsey, *Nuclear Moments* (John Wiley and Sons, Inc., New York, 1953).

<sup>29</sup> Bohr, Koch, and Rasmussen, Arkiv Fysik **4**, 455 (1951).

<sup>30</sup> C. Schwartz, Phys. Rev. **97**, 380 (1955); **99**, 1035 (1955); **105**, 173 (1957).

<sup>31</sup> Crawford, Gray, Kelly, and Schawlow, Can. J. Phys. **A28**, 138 (1950).

<sup>32</sup> G. Breit, Rydberg Centennial Conference on Atomic Spectroscopy, June (1954); Lunds Univ. Arsskr. **50**; Kgl. Fysiograf. Sällskap. i Lund Handl. **65**, 85.

<sup>33</sup> E. P. Wigner, Ann. Math. **53**, 36 (1951).

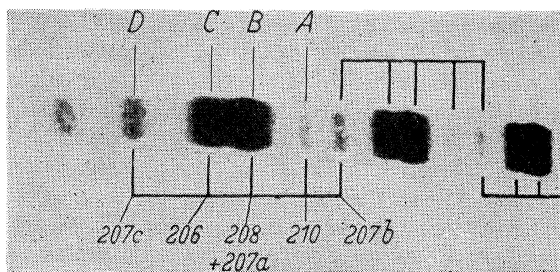


FIG. 5. The hyperfine structure of Pb I  $\lambda 4058A$  obtained with Fabry-Perot etalon by Brix *et al.*<sup>35</sup> The measurements have a bearing on the intrinsic nuclear quadrupole moment effects,<sup>39,40</sup> the shell-structure effect<sup>48</sup> and the single particle wave function effect.<sup>49</sup>

It was necessary<sup>6</sup> to invoke this effect in order to explain the general features of the shifts in Pb I and it has since been used extensively by Crawford.<sup>34</sup>

In summary it is seen that the determination of the effective coupling constant between the electron system and the nucleus is not very simple and that one should not rely on it to better than a few percent. Since the experimental data usually provide displacements for several spectroscopic terms it is unlikely, however, that conscientiously determined values of the effective  $\Delta r_0/r_0$  are wrong by more than a few percent and systematic errors of 50% in cases for which there are checks available appear improbable.

The determination of ratios of relative isotope shifts for the same element is free of errors in the determination of  $\psi^2(0)$  and effects of screening of inner shells by the valence electron. They depend only on the validity of first-order perturbation theory as applied to the small perturbing potential representing the differences between the "electronic configuration"-"nucleus" field as one goes from isotope to isotope. There is no doubt regarding this validity, the difference in the shape of the electron wave functions inside the nucleus being very slight even though these functions differ appreciably from those for a pure Coulomb field. Observations enabling one to draw conclusions of this type are illustrated in Fig. 5 which reproduces a Fabry-Perot exposure of  $\lambda 4058$  with RaD sample obtained by Brix, Buttler, Houtermans, and Kopfermann.<sup>35</sup> The faint line caused by Pb<sup>210</sup> is one of the sources of information regarding an anomaly in the isotope shift which will be discussed later on. From such observations one can deduce the values of relative isotope displacements for the pairs (206,207) (207,208) (208,210).

Crawford and Schawlow (1949)<sup>14</sup> on applying corrections for screening of  $(6s)^2$  and wave-function distortion to Hg II, Tl III, and Pb IV found that a uniform charge distortion and the nuclear radius formula

$$r_0 = 1.5 \times 10^{-13} A^{\frac{1}{2}} \text{ cm}$$

<sup>34</sup> M. F. Crawford, Phys. Rev. **99**, 1637 (1955); other papers by the same author.

<sup>35</sup> Brix, Buttler, Houtermans, and Kopfermann, Z. Physik **133**, 192 (1955). Additional measurements on Pb<sup>210</sup> have been made by Manning, Anderson, and Watson, Phys. Rev. **78**, 417 (1950).

give an isotope shift  $\cong 2 \times$  observed. Brix and Kopfermann (1949)<sup>36</sup> and Humbach (1952)<sup>37</sup> reach the same conclusion. It is obviously difficult to draw an inference from this regarding the validity of the nuclear radius formula. The latter may be a good representation of the average nuclear size without giving an accurate account of the minute changes taking place on the addition of a few neutrons. It appeared, therefore, understandable that Schawlow and Townes (1952)<sup>38</sup> found no significant difference between the uniform density model and experiment when they compared the total energy displacement  $\delta W$ , i.e., the IS, for the fine structure of x-ray terms. The charge distribution favored by experiment using a somewhat large nuclear radius appeared to be intermediate between the top slice and the uniform types. The large reliance which must be made on the accuracy of the calculation of the x-ray term energy difference has to be remembered in this connection. Absolute calculations are well known to be more difficult than relative ones especially in a case involving many-body interactions.

### (B) Deformed Nuclei

In an attempt to account for the RIS of Nd, Sm, and Eu, Brix and Kopfermann<sup>39</sup> introduced the effect of the ellipsoidal nuclear shape into the theory. This effect has received additional consideration<sup>40</sup> and has proved very useful in correlating observations on the RIS with the collective model of nuclear structure. The nucleus is usually assumed to be deformed retaining its density. The theory has been considered explicitly by Ford<sup>41</sup> and by Wilets *et al.*,<sup>42a</sup> and independently by Bodmer.<sup>42b</sup>

<sup>36</sup> P. Brix and H. Kopfermann, Z. Physik **126**, 344 (1949).

<sup>37</sup> W. Humbach, Z. Physik **133**, 589 (1952).

<sup>38</sup> A. L. Schawlow and C. H. Townes, Science **115**, 284 (1952). The possibility of using smaller nuclear radii for the x-ray fine structure has been examined experimentally and theoretically by R. L. Shacklett and J. W. M. DuMond, Phys. Rev. **106**, 501 (1957) and has been reported on by Shacklett at the Stanford University Conference on Nuclear Sizes and Density Distributions, December 18 (1957). It appears likely from this report that the smaller radii indicated by electron scattering data account satisfactorily for the improved x-ray measurements. These have to do with the charge distribution of a nucleus rather than the change in the distribution caused by the addition of a neutron. Radiative corrections play a relatively large role in this work.

<sup>39</sup> P. Brix and H. Kopfermann, Nachr. Akad. Wiss. i. Göttingen, Math.-physik. Kl. **2**, 31 (1947).

<sup>40</sup> P. Brix and H. Kopfermann, Festschr. Akad. Wiss. i. Göttingen, Math.-physik. Kl. **17** (1951); Kopfermann, Stuedel, and Thulke, Z. Physik **138**, 309 (1954) (Ru); Hindmarsh, Kuhn, and Ramsden, Proc. Phys. Soc. (London) **A67**, 478 (1954) (Sn, Cd); G. Noeldeke and A. Stuedel, Z. Physik **137**, 632 (1954); Brix, Buttler, Kopfermann, and Houtermans, Z. Physik **133**, 192 (1952) (Pb 210).

<sup>41</sup> K. W. Ford, Phys. Rev. **90**, 29 (1953).

<sup>42a</sup> Wilets, Hill, and Ford, Phys. Rev. **91**, 1488 (1953).

<sup>42b</sup> A. R. Bodmer, Proc. Phys. Soc. (London) **47**, 622 (1954). In connection with the discussion in the text and especially Eq. (6), reference should be made to the relation of isotope shift to electron scattering also studied by Bodmer [Proc. Phys. Soc. (London) **46**, 1041 (1953)] in relation to the electron scattering study of H. Feshbach [Phys. Rev. **84**, 1026 (1951)]. In these studies  $\langle r^2 \rangle \cong \langle r^{2p} \rangle$  appears as the parameter characterizing the nuclear charge distribution.

Taking the nuclear shape to be such that the radius at an angle  $\theta$  with the symmetry axis is

$$r(\theta) = r_0[1 + \beta P_2(\cos\theta)]N_\beta, \quad (4)$$

where  $N_\beta$  is a normalization constant and comparing the deformation effect with that of a uniform spherical charge distribution,

$$\frac{\delta W_\beta}{\delta W_r} = \frac{\beta \partial W / \partial \beta}{\delta r_0 \partial W / \partial r_0} \approx \frac{\rho(2\rho+3)}{5} \beta^2 \quad (4.1)$$

so that the ratio of relative isotope shifts due to the two causes is

$$\frac{\Delta \delta W_\beta}{\Delta \delta W_r} \approx \frac{3}{10} (2\rho+3) \frac{A \partial(\beta^2)}{\partial N}, \quad (4.2)$$

where  $N$ ,  $A$  are, respectively, the neutron and mass numbers. Only the leading terms in the nuclear radius are included in these formulas and they are concerned with  $s$  electrons exclusively. If one assumes that for  $\Delta N = 2$ ,  $\Delta \beta^2 = 0.005$  for  $Z = 62$ ,  $A = 150$  the ratio in (4.2) has a value 0.54. The quadrupole moment connected with  $\beta^2$  is that corresponding to the intrinsic nuclear deformation<sup>43</sup> rather than to the measured quadrupole moment.

The part of the calculations of Wilets, Hill, and Ford which makes use of the consideration of the electron in the field of the nucleus and leads them to the calculation of contributions of the three regions corresponding to the electron distance  $r$  being greater than the major semiaxis, smaller than the minor semiaxis and the intermediate region is not necessary for an understanding of the phenomenon. The origin of the effect is understood more clearly, as is realized by them in connection with compressibility considerations, by noting that for undistorted electron wave functions the IS is a double integral representing the mutual electrostatic energy between the electron and the nuclear charge distributions. It can be evaluated by determining first the electrostatic potential caused by the mean electron charge distribution and integrating the product of the nuclear charge density and this potential. Approximating the electron density by

$$\sum_\mu \psi_\mu^* \psi_\mu = \frac{B r^{2\rho-2}}{4\pi Z e^2}, \quad (5)$$

where  $B$  is a constant and  $Z$  the nuclear charge, the electrostatic potential is readily seen to be

$$\varphi = \frac{B}{Ze} \frac{r^{2\rho}}{2\rho(2\rho+1)} + C, \quad (5.1)$$

where  $C$  is a constant and  $-e$  is the charge on the electron. The IS thus depends on the mean value  $\langle r^{2\rho} \rangle$  weighted in proportion to the nuclear charge

density. In the notation of Eqs. (4.1), (4.2) the ratio of isotope shift for the deformed volume to that for the spherical volume is

$$(\delta W_r + \delta W_\beta) / \delta W_r = \left( \int r^{2\rho} d\mathbf{r} \right)_\beta / \left( \int r^{2\rho} d\mathbf{r} \right)_{\beta=0} \quad (5.2)$$

Here the subscript  $\beta$  means that the integration is taken over the volume enclosed by the surface  $r = r(\theta)$  of (4). The requirement of incompressibility of nuclear matter gives

$$N_\beta^3 [1 + 3\beta^2/5] = 1, \quad (5.3)$$

while

$$\left( \int r^{2\rho} d\mathbf{r} \right)_\beta = N_\beta^{2\rho+3} \left( \int r^{2\rho+2} (1 + \beta P_2)^{2\rho+3} d\mathbf{r} \right)_{\beta=0}, \quad (5.4)$$

as is readily seen by mapping the distorted on to the undistorted volume. To within the order  $\beta^2$  the integral in the right-hand side of the last equation divided by its value for  $\beta = 0$  is

$$1 + \frac{1}{5}(\rho+1)(2\rho+3)\beta^2, \quad (5.5)$$

while according to (5.3)

$$N_\beta^{2\rho+3} = 1 - \frac{1}{5}(2\rho+3)\beta^2, \quad (5.6)$$

resulting in

$$(\delta W_r + \delta W_\beta) / \delta W_r = 1 + \frac{1}{5}\rho(2\rho+3)\beta^2. \quad (5.7)$$

Comparison of (5.5) and (5.6) shows that  $\delta W_\beta / \delta W_r$  is composed of two parts the first of which is  $(\rho+1)/\rho$  times and the second  $-1/\rho$  times the answer. The first arises from the deformation consisting in changing  $r \rightarrow r[1 + \beta P_2(\cos\theta)]$ ; the second, from the contraction of this deformed distribution which is represented by (5.3) and is a consequence of the assumed incompressibility of nuclear matter. It is apparent that a quantitative application of (4.1) which is equivalent to (5.7) relies very heavily on this hypothesis, about one-half of the effect having been removed by the contraction. It is also clear that the measurement of the RIS is not a direct determination of the electric quadrupole moment. Taken literally but without corrections for wave-function distortion it is a measure of

$$\Delta \langle r^{2\rho} \rangle \propto \text{RIS}, \quad (6)$$

where  $\Delta$  as previously is the change caused by going from isotope to isotope. In principle, any electric multiple contributes to the left-hand side of (6). The electric quadrupole is essentially a measure of

$$\langle r^2 P_2(\cos\theta) \rangle \quad (6.1)$$

and is not identical with (6). The connection of (6.1) with (6) is not unique. The connection is apparent from (5.1). The potential caused by the electron increases with  $r$  roughly quadratically. On account of the non-linear dependence on  $r$ , displacing half of an element of

<sup>43</sup> A. Bohr and B. R. Mottelson, Phys. Rev. **89**, 316 (1953).

charge out and half in through the same radial distance increases the energy and produces the RIS.

On the incompressible fluid assumption Eq. (4.2) indicates that the RIS caused by the intrinsic quadrupole moment should be large when the deformation parameter varies rapidly with  $N$ . Thus in the explanation of the anomalously large shift between  ${}_{63}\text{Eu}^{151}$  and  ${}_{63}\text{Eu}^{153}$  a large change in  $\beta^2$  is supposed to take place between  $N=88$  and  $N=90$ . This view is supported by measurements of the quadrupole moment  $Q$ . For the shift between  ${}_{62}\text{Sm}^{150}$  and  ${}_{62}\text{Sm}^{152}$  both of which have no nuclear spin there is no observed  $Q$  but it is supposed that  $Q_0$ , the intrinsic  $Q$ , performs a jump as  $N$  changes from 88 to 89. The intrinsic moment  $Q_0$  is a property of the deformed nucleus and in a classical mechanics picture precesses together with the axis of symmetry around the total angular momentum  $I$ . If  $I=0$  the average component of a vector along the axis of symmetry is zero in any direction because the projection on the spin axis vanishes. The quadrupole moment cannot be observed directly in this case therefore. The relation between  $Q$  and  $Q_0$  is

$$Q_0 = \frac{I+1}{I} \frac{2I+3}{2I-1} Q. \quad (6.2)$$

The deformation parameter  $\beta=0$  for closed shells. Thus disregarding deforming effects of the protons at the magic neutron numbers  $N=82$  and  $126$  the nucleus is expected to be spherically symmetric. According to data on rotational levels the deformation  $\beta^2$  reaches a maximum in between. *The quadrupole contribution to the RIS may be expected, therefore, to be great close to the magic numbers.* It is positive just after the shell is closed and negative as closure of the shell is approached. This expectation is in agreement with observation on the  $\text{Pb}^{208}-\text{Pb}^{206}$  and  $\text{Pb}^{210}-\text{Pb}^{208}$  shifts for which the heavier pair shows the larger shift. Therefore *just after the shell is closed the RIS should be large* since the quadrupole effect adds itself to the effect of general expansion of the nuclear volume and just before closure the shift should be small since the quadrupole effect is of the opposite sign.

There has been some theoretical expectation of the deformation of even-even nuclei being larger than that of the odd-even kind and there has been some support for this view in observations on energy levels. Wilets, Hill, and Ford<sup>42a</sup> use this fact in their explanation of the even-odd staggering such as has been illustrated in the case of  $\text{Pb}^{206,207,208}$  in Fig. 3. The statement

$$(\beta^2)_{2n+1} < \frac{1}{2} [(\beta^2)_{2n} + (\beta^2)_{2n+2}], \quad (7)$$

which expresses the belief that for an odd number  $2n+1$  of neutrons in a shell the deformation is anomalously small in comparison with deformations for  $2n$  and  $2n+2$  neutrons is equivalent to

$$(\beta^2)_{2n+1} - (\beta^2)_{2n} < (\beta^2)_{2n+2} - (\beta^2)_{2n+1}, \quad (7.1)$$

which, when combined with the arguments regarding the effects of  $\beta^2$  on  $\langle r^{2p} \rangle$ , implies the even-odd staggering phenomenon. The argument holds either at the beginning or at the end of the formation of a closed shell. According to recent evidence the difference in the deformations of even-even and odd-even nuclei is not as definite as it appeared to be<sup>44</sup> so that the explanation of the odd-even staggering which has just been presented does not appear as relevant as it did a few years ago.

The relation of the above explanation to the collective model has been studied by K. W. Ford<sup>41</sup> who has correlated the isotope shift data with that on the position of first excited states of even-even nuclei and has shown that there is a strong parallelism in the two phenomena. The distortion parameter needed for the explanation of energy levels is too large however in comparison with indications from isotope shift data. Ford is especially concerned with the largeness of the theoretical prediction for  $\beta^2$ . In an attempt to account for the observations of Arroe on the anomalously large isotope shift of Ce, Ford<sup>45</sup> re-examined the evidence, coming to the conclusion that there is a systematic discrepancy between the mechanical moments of inertia as determined from spacings of energy levels and the data on electrical deformations as derived from isotope shift quadrupole moments and other electromagnetic evidence, the moments of inertia required by energy spacings being  $4 \pm 1$  times too large. It will be recalled in this connection that the correlation between  $Q_0$  and experiment which has been achieved by Wilets, Hill, and Ford required the use of two arbitrary parameters, one of which took care of the difference between the mechanical and electrical moments and another which involved a shift of the theoretical curve down by 0.5 and which was attributed to effects of compressibility. The agreement obtained after these adjustments was sufficient to establish the correlation in major trends beyond any doubt but nevertheless leaving some quantitative questions. Sunyar<sup>46</sup> has made additional measurements on  $E2$  and  $(E2) + M1 \gamma$  transitions ( $\text{Sm}^{152}$ ,  $\text{Gd}^{154}$ ,  $\dots$ ,  $\text{Po}^{210}$ ) and finds close parallelism in the behavior of the electrical and mechanical deformation parameters obtaining values of  $\beta^2$  from energy spacing which are about 5 times those from the mean lives. The existence of a discrepancy had already been noted by Bohr and Mottelson.

Since there appears to be agreement between the electric sources of information regarding  $\beta^2$ , it is appealing to consider the explanation of the RIS as satisfactorily settled from a phenomenologic viewpoint. One might wish to relegate the consideration of nuclear level spacings to another chapter and hope that whatever is wrong in the theory does not affect the isotope shift. Such optimism appears to be premature however

<sup>44</sup> The writer is indebted to Dr. L. Wilets for this remark.

<sup>45</sup> K. W. Ford, Phys. Rev. **95**, 1250 (1954).

<sup>46</sup> A. W. Sunyar, Phys. Rev. **98**, 653 (1955).



because most electromagnetic interaction data have to do with  $\gamma$ -ray transition probabilities and with related Coulomb excitation transitions so that relevant matrix elements have little relation to  $\langle r^{2\rho} \rangle$  which is not directly concerned with nuclear angular properties. Only data on monopole transitions depend on spherically symmetric averaging of the nuclear charge distribution. As long as the collective model contains serious discrepancies of the type mentioned, it is thus difficult to be sure that the nuclear deformation associated with the intrinsic nuclear quadrupole moment constitutes the main part of the electron-nucleus interaction determining the RIS.

The *polarization* of the nucleus by the *electron* has been studied theoretically by Breit, Arfken, and Clendenin.<sup>47</sup> Because of the large increase in the electron mass when the electron is inside a heavy nucleus, the effect is appreciably larger than one might suppose. Another reason for considering this effect is that a systematic difference can be expected to exist between odd-even and even-even nuclei regarding polarizability on account of the difference in level densities of such nuclei. The calculations have been performed for electric-monopole and electric-dipole effects. The dipole effect appears to be too small to be of real interest. The monopole effect can be made comparable with the odd-even staggering if sufficiently drastic departures from central field approximations to nuclear wave functions are assumed. It was also found necessary to suppose that differences in densities and transition probabilities of perturbing nuclear levels in the region of a few Mev are not compensated for as the nuclear excitation is increased to 10 or 20 Mev. Irregularities in positions of levels of even isotopes are not explicable by the polarization view in a natural manner but the absence of non-negligible effects has not been definitely established. It appears possible that the smallness of the value of the observed RIS is caused by partial cancellation of the effect of progressive changes in the nuclear radius by the polarization effect.

Suggestions have been made regarding another possible cause of odd-even staggering.<sup>48</sup> The Schmidt model which accounts with some success for the observed nuclear magnetic moments implies that unpaired nucleons are geometrically outside closed shells to an appreciable degree. It may be expected, therefore, that the addition of an odd neutron to  $\text{Pb}^{206}$  to form  $\text{Pb}^{207}$  produces somewhat less than one-half of the expansion effect on the effective radius of the nuclear charge distribution which is produced when a neutron pair is added to form  $\text{Pb}^{208}$ . Although this explanation

is very different from that of Wilets, Hill, and Ford, the common feature of both is the difference in charge distributions of even-even and odd-even nuclei. This difference is common to all explanations which do not make use of the polarization of nuclei by electrons.

Another suggestion based on the individual particle model and the shell structure theory was made<sup>49</sup> in connection with the anomalously large shift  $\text{Pb}^{210} - \text{Pb}^{208}$ . Two  $2g_{9/2}$  neutrons added to  $\text{Pb}^{208}$  were supposed to form  $\text{Pb}^{210}$  while the two neutrons which have to be added to  $\text{Pb}^{206}$  to form  $\text{Pb}^{208}$  were taken to be  $1i_{13/2}$ . Since  $g$  neutrons are more penetrating than  $i$  neutrons, the nuclear charge core may be supposed to be expanded relatively more by their addition. Wave functions of neutrons outside the nuclear core have been estimated to be nearly of the individual particle type, the mean distance between two neutrons being sufficiently large in this region in comparison with the range of the nucleon-nucleon force. Estimates<sup>49</sup> indicated that the observed ratios  $[(210) - (208)] / [(208) - (206)] \cong 1.5$  could be approximately accounted for on this view. The picture used in this explanation fits in naturally with the smallness of the part of the RIS which is usually attributed to the progressive change of the nuclear radius as has been previously pointed out.<sup>5, 6, 14, 36, 37</sup> The wave function of the last neutron is located inside the proton distribution only partly and it is to be expected that the charge distribution expands only partially. A related viewpoint has been previously expressed by Kopfermann<sup>27</sup> but without specific reference to properties of wave functions.

More recently Wilets<sup>50</sup> arrived at similar conclusions as a result of studies of the compressibility of nuclear matter. The latter do not fit the requirements set upon them by the isotope shift<sup>51</sup> which favor a compressibility of  $\sim 70$  Mev while calculations of Brueckner and Gammel<sup>52</sup> favor  $\sim 170$  Mev more closely in agreement with the incompressible fluid model. Since Wilets' calculations on neutron and proton distributions favor a larger neutron than proton radius for the heavy elements, it appeared natural to him to suppose that the addition of a neutron does not affect seriously the proton distribution. The only essential part of this argument is that the added neutron remains mainly on the outside of the proton distribution. The explanations just considered agree in keeping the added neutron somewhat outside the proton distribution. Eventually the wave-function approach is likely to be the more informative.

<sup>49</sup> G. Breit, Phys. Rev. **86**, 254 (1952).

<sup>50</sup> L. Wilets, private communication. For compressibility studies referred to, compare reference<sup>52</sup> and R. Berg and L. Wilets, Proc. Phys. Soc. (London) **A68**, 229 (1955); R. A. Berg and L. Wilets, Phys. Rev. **101**, 201 (1956); L. Wilets, Phys. Rev. **101**, 1805 (1956).

<sup>51</sup> K. W. Ford and D. L. Hill, Ann. Rev. Nuclear Sci. **5**, 46 (1955).

<sup>52</sup> K. A. Brueckner and J. L. Gammel, "The properties of nuclear matter" (paper in preprint form).

<sup>47</sup> Breit, Arfken, and Clendenin, Phys. Rev. **78**, 390 (1950).

<sup>48</sup> P. Brix and H. Kopfermann, Nachr. Acad. Wiss. i. Göttingen, Math.-physik. Kl. **2**, 31 (1947); M. Fierz, Nachr. Acad. Wiss. i. Göttingen, Math.-physik. Kl. **3**, 1 (1947); G. Breit, Phys. Rev. **78**, 470 (1950); also, compare G. Breit, Phys. Rev. **79**, 891 (1950) for an acknowledgment of the priority of publications by Brix and Kopfermann and by Fierz which were overlooked by the author.

Recent work of Nilsson<sup>53</sup> and of Mottelson and Nilsson<sup>54</sup> suggests that the actual situation regarding the addition of neutrons may be much more complicated. Nevertheless, it appears difficult to exclude the possibility that differences in the penetrating power of neutrons with different values of  $L$  matter in approximately the manner just referred to. Consequently there is more certainty in the statement that the intrinsic quadrupole effects can account for an appreciable part of the  $[(210)-(208)]/[(208)-(206)]$  anomaly in Pb in the manner suggested by Willets, Hill, and Ford than in the inference that there are no other important effects entering the explanation of this anomaly.

Even though the quantitative features of the intrinsic quadrupole moment explanation are far from certain, there is no doubt regarding the existence of correlations between expectation and fact. It is noteworthy in this connection that in the case of ground-state configurations Mottelson and Nilsson<sup>54</sup> have found evidence of correlations in changes of  $\beta$  as calculated on the unified model and observed changes in  $Q_0$ . Their calculations reproduce the large change in  $\beta^2$  at  $N=88, 90$  which is suggested by isotope shifts of Sm and Eu. The values of  $Q_0$  used by them come, however, from other electromagnetic evidence and the difficulty of reconciling the electrical and mechanical moments of inertia is not resolved by their work.

The radiative correction<sup>55</sup> to the magnetic moment of the electron can be expected to have a non-negligible effect on the spectroscopic isotope shift. The magnitude of the effect was estimated as  $\sim 0.05$  of the progressive change of radius effect. The nature of the effect is the interaction of the radiative correction to the electron moment with the electric field of the nucleus. While relatively small this effect adds to other somewhat uncertain contributions and contributes to the difficulty of deriving unique conclusions from the experimental observations.

### III. CONCLUDING REMARKS

There is little doubt about the existence of effects of changes in the distribution of nuclear charge density on the electronic energy levels and the approximate agreement of observed effects with progressive changes in

<sup>53</sup> S. G. Nilsson, Kgl. Danske Videnskab, Selskab, Mat.-fys. Medd. **29**, 16 (1955).

<sup>54</sup> B. R. Mottelson and S. G. Nilsson, Phys. Rev. **99**, 1615 (1955).

<sup>55</sup> G. Breit and W. W. Clendenin, Phys. Rev. **85**, 689 (1952).

radii to be expected on general views. The e is good correlation with anomalies to be expected on the basis of values obtained for the intrinsic quadrupole moments from sources other than atomic spectroscopy. The large numbers of causes that have been estimated to contribute amounts comparable with those observed makes it difficult to draw unique conclusions regarding the changes in the shape of intrinsic nuclear charge distributions which occur with the successive addition of neutrons to a nucleus. The main reason for the difficulty in obtaining a unique interpretation is the smallness ( $\sim 1/600$ ) of the estimated fractional change in the nuclear radius which takes place when a neutron is added to a heavy nucleus. It appears possible however that with the improvement of theories of nuclear structure the spectroscopic isotope shift will become a significant test of their validity.

A characteristic feature of the available theories is their one-sidedness and the schematic character of the treatment of the nucleus. In many of the theoretical attempts it is assumed that the addition of a neutron to a volume occupied by nuclear matter will result in an expansion of the charge distribution and that the expansion can be estimated approximately by assuming incompressibility of nuclear matter. Such a hypothesis has not been well established and the emphasis on the strong influence of the geometry of the arrangement of nucleons has not been well founded. The added neutron is not completely localizable inside the proton charge distribution and while it is outside of that distribution it exerts a force on the protons and neutrons at smaller distances than itself. The discussions of proton and neutron charge distributions and their compressibilities inside a nucleus may be good enough for a reproduction of the action of the nucleus as a whole without being exact enough for the understanding of the small effects in the charge distribution which take place as the result of adding one neutron. The representation of the nucleus as made of "nuclear matter" has never been justified in a satisfactory manner and the applicability of such concepts as compressibility is questionable. It appears therefore that the problem of the isotope shift will have to be treated on its own merits much more so than in the past. The changes in the nuclear wave function resulting from the addition of a neutron will have to be examined more directly than heretofore and the changes of nuclear charge distribution will then be derivable without the unjustified employment of concepts of the properties of nuclear matter in bulk.

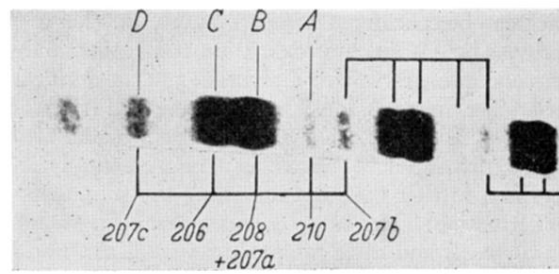


FIG. 5. The hyperfine structure of  $\text{PbI } \lambda 4058.4$  obtained with Fabry-Perot etalon by Brix *et al.*<sup>36</sup> The measurements have a bearing on the intrinsic nuclear quadrupole moment effects,<sup>39,40</sup> the shell-structure effect<sup>48</sup> and the single particle wave function effect.<sup>49</sup>