Nuclear Size Estimates from X-Ray Fine Structure Measurements

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I. INTRODUCTION

THE fact that the energies of some of the bound atomic electrons are affected to a slight extent by the finite size of the nucleus has long been recognized. One of the first theoretical considerations of the problem was given by Rosenthal and Breit[1] in the early nineteen thirties in an attempt to relate observed isotope shifts to changes in nuclear radius. Employing first-order perturbation calculations with relativistic wave functions they found that only the energy levels of $s_{\frac{1}{2}}$ and $p_{\frac{1}{2}}$ electrons were affected an appreciable amount by the spatial extent of nuclear charge, all other electronic wave functions being zero at the origin. The direction of the displacement of the levels was found to be upwards (decreased binding) because the potential for an electron inside the nuclear volume is much smaller than that due to a point nuclear charge.

While giving the correct order of magnitude for the energy displacement, the perturbation method cannot be relied upon for accurate values since, although the energy displacement is itself quite small, the perturbing potential near the origin is very large. Broch[2] and others[3] have shown that the correction factor to be applied to the perturbation formula results in an appreciable change in the case of the heavier elements, being about 0.75 for Pb($Z=82$). They also demonstrated the significant result that the perturbation formula, while giving an incorrect value of level displacement, does give accurate information on how the energy shift depends on nuclear size and shape for a given value of Z.

The total difference in energy between electron levels for a point nucleus and for a nucleus of finite size cannot be obtained directly from isotope shifts. What is observed experimentally is the small change in energy due to slight differences in nuclear radius between different isotopes. It is possible, however, to determine the total change in electron energy through precise measurements of the L_{II}-L_{III} x-ray level splitting in heavy elements. In this case the $L_{II}(2p_{\frac{1}{2}})$ level undergoes a small displacement due to nuclear size while the $L_{III}(2p_{\frac{3}{2}})$ level remains essentially unperturbed. The radial probability distributions for electrons in these two states are similar enough in shape so that rather accurate calculations may be made of the energy difference assuming a pure Coulomb field. A comparison of experimental measurements of the x-ray fine structure with the calculated values would then reveal any systematic discrepancy which could be interpreted in terms of a non-Coulomb field caused by the finite nucleus.

Schawlow and Townes[4] made such a comparison. Using Broch's[2] modified perturbation method they evaluated the energy level shift due to finite nuclear size for five values of Z between 60 and 95, and found that the shift is strongly Z dependent and can be represented by an exponential in Z. The total fine structure splitting was evaluated for a point nucleus using the formula of Christy and Keller[5] and compared with measured values obtained from the tables of Cauchois and Hulubei.[6] The resulting discrepancy showed just the expected type of systematic deviation for large values of Z. The correction factor necessary to minimize the difference between theory and experiment could be interpreted in terms of a uniformly charged nucleus of radius $R=r_0A^{\frac{1}{3}}$ with $r_0=(2.1\pm0.2)\times10^{-13}$ cm.

Schawlow and Townes emphasize that the finite size of the nucleus is not the only thing which modifies the fine structure splitting. Certain quantum electrodynamic effects cause deviations from a Coulomb field which, for large Z, are of the same order as the nuclear size effect. The large value of r_0 obtained by them, when compared with the value currently accepted as most accurate, namely, $r_0=1.2\times10^{-13}$ cm, indicates the need for explicit corrections for these quantum electrodynamic effects.

These effects are:

(1) The anomalous magnetic moment of the electron, which causes a constant fractional change in the fine structure splitting.

(2) The polarization of the vacuum which, in the case of the fine structure splitting, has the effect of an attractive potential (opposite to that of the nuclear size effect).

(3) Radiative reaction effects (Lamb shift) which correspond to a repulsive potential (in the same direction as the nuclear size effect).

[1] J. E. Rosenthal and G. Breit, Phys. Rev. **41**, 459 (1932); G. Breit, *ibid.* **42**, 348 (1932).

[2] E. K. Broch, Arch. Math. Naturvidenskab **48**, 25 (1945).

[3] See K. W. Ford and D. L. Hill, Phys. Rev. **94**, 1630 (1954) for references to other theoretical work on this problem.

[4] A. L. Schawlow and C. H. Townes, Phys. Rev. **100**, 1273 (1955).

[5] R. F. Christy and J. M. Keller, Phys. Rev. **61**, 147 (1942).

[6] Y. Cauchois and H. Hulubei, *Longueurs d'Onde des Emissions X et des Discontinuities d'Absorption X* (Hermann and Cie, Paris, 1947).

A correction for the effect of the anomalous moment of the electron was made by Schawlow and Townes, but since accurate calculations of correction terms for the other two effects for large values of Z are extremely difficult and had not been done at that time no explicit correction was made for them. Subsequent work of Wichmann and Kroll[7] on vacuum polarization in a strong Coulomb field now makes possible a correction to the fine structure splitting for this effect. It also makes possible an empirical evaluation of the magnitude of the Lamb shift term providing the nuclear radius is regarded as known.

Shacklett and DuMond[8] have recently made a series of precision measurements on the x-ray fine structure splitting for several heavy elements and have found that the large value of nuclear radius deduced by Schawlow and Townes on the basis of older x-ray data can be ascribed in part to inaccuracies in the data. With no correction for effects (2) and (3), a comparison of the theory with the new experimental measurements yields a significantly smaller value of r_0, namely, $r_0=1.08\times10^{-13}$ cm. When corrections for vacuum polarization and a nuclear radius with $r_0=1.2\times10^{-13}$ cm are included, the remaining discrepancy has the sign and magnitude expected for the Lamb shift effect.

A brief resume of the theoretical work on the nuclear size effect and the other associated corrections is presented in order to provide a more coherent background for description of the latest experimental measurements and the results obtainable from them.

II. THEORETICAL CONSIDERATIONS

First-Order Perturbation Method

The energy level shift caused by finite nuclear size may be obtained to first order by the perturbation integral

$$\Delta E=4\pi\int_0^\infty P(r)(V+Ze^2/r)r^2dr, \tag{1}$$

where $P(r)$ is the probability density of an electron near a point nucleus and V is the potential energy of the electron inside the nucleus. Ford and Hill[3] show that the potentials may be replaced by the charge densities which produce them (by Poisson's equation) so that (1) may be put into a form which shows the dependence of ΔE on the nuclear charge distribution somewhat more explicitly. Let the potential resulting from the unperturbed $p_{\frac{1}{2}}$ electron be represented by $V_e(r)$ which is normalized to be zero at the origin. Then

$$\Delta E=4\pi\int_0^\infty \rho_n(r)V_e(r)r^2dr, \tag{2}$$

where $\rho_n(r)$ is the density of nuclear charge. Because of the large relativistic effects the $p_{\frac{1}{2}}$ Dirac wave function becomes infinite at the origin, varying as $r^{\sigma-1}$ $[\sigma=(1-\alpha^2Z^2)^{\frac{1}{2}}$, and $\alpha=e^2/\hbar c]$; the corresponding electron charge density $\rho_e(r)$ varies as $r^{2\sigma-2}$. The potential may then be written[3]

$$V_e(r)=[4\pi Ae/2\sigma(2\sigma+1)]r^{2\sigma}, \tag{3}$$

where A is a normalization constant. The perturbation integral (2) now takes the form

$$\Delta E=\phi(Z)\langle r^{2\sigma}\rangle, \tag{4}$$

where

$$\langle r^{2\sigma}\rangle=(Ze)^{-1}\int_0^\infty r^{2\sigma}\rho_n(r)4\pi r^2dr, \tag{5}$$

and represents the average value of $r^{2\sigma}$ weighted by the nuclear charge distribution. For example, if $\rho_n(r)$ is constant over the nuclear volume, then $\langle r^{2\sigma}\rangle=R^{2\sigma}\times[3/(2\sigma+3)]$ where R is the nuclear radius. This form shows the explicit dependence upon the size and shape of the nuclear charge distribution.

More Exact Calculations

The technique of Broch,[2] as used by Schawlow and Townes, avoids the difficulty inherent in the perturbation method, namely, that the perturbing potential is so large near $r=0$. Broch writes down the two-component radial wave equations for a $p_{\frac{1}{2}}$ electron in a central field;

$$dF/dy-F/y=[1-(E-V)/mc^2](G/2\gamma) \tag{6a}$$

$$dG/dy+G/y=[1+(E-V)/mc^2](F/2\gamma), \tag{6b}$$

where $y=2Zr/a_0$, $\gamma=\alpha Z$, and a_0 is the Bohr radius of hydrogen. A pair of similar equations is written down for a point nucleus:

$$dF_0/dy-F_0/y=[1-(E_0-V_0)/mc^2](G_0/2\gamma) \tag{7a}$$

$$dG_0/dy+G_0/y=[1+(E_0-V_0)/mc^2](F_0/2\gamma). \tag{7b}$$

Now if Eq. (6a) is multiplied through by G_0, Eq. (6b) by $-F_0$, Eq. (7a) by $-G$, and Eq. (7b) by F, we obtain by addition

$$d(FG_0-F_0G)/dy=(\Delta V-\Delta E)(FF_0+GG_0)/2\gamma mc^2, \tag{8}$$

where $\Delta V=V-V_0$ and $\Delta E=E-E_0$. Integration from $y=0$ to $y=\infty$ yields

$$\Delta E\int_0^\infty (FF_0+GG_0)dy=\int_0^{y_0}\Delta V(FF_0+GG_0)dy \tag{9}$$

using the fact that $\Delta V=0$ outside the nuclear radius y_0 and that the wave functions vanish at $y=0$ and at $y=\infty$. The integral on the left can be approximated by assuming

$$\int_0^\infty (FF_0+GG_0)dy\approx\int_0^\infty (F_0^2+G_0^2)dy=2Z/a_0, \tag{10}$$

[7] E. H. Wichmann and N. M. Kroll, Phys. Rev. **101**, 843 (1956).
[8] R. L. Shacklett and J. W. M. DuMond, Phys. Rev. **106**, 501 (1957).

allowing Eq. (9) to be written in the form

$$\Delta E=\int_0^R \Delta V(FF_0+GG_0)dr. \quad (11)$$

This is exact except for the small error made in the approximation of Eq. (10). Equation (11), which differs from Eq. (1) in the form of $P(r)$ and in a normalization constant, is still not useful for calculational purposes because F and G are unknown inside the nucleus and cannot be approximated by F_0 and G_0 as in Eq. (10), since the perturbing potential ΔV is not small.

Broch circumvents this difficulty by noting that $\Delta V=0$ for $y>y_0$ and integrating Eq. (8) from y_0 to infinity rather than from zero to infinity. The result is

$$(FG_0-F_0G)_{y0}=\frac{\Delta E}{2\gamma mc^2}\int_{y_0}^{\infty}(FF_0+GG_0)dy, \quad (12)$$

where another good approximation is possible by assuming as in Eq. (10) that

$$\int_{y_0}^{\infty}(FF_0+GG_0)dy\approx\int_0^{\infty}(FF_0+GG_0)dy\approx 2Z/a_0, \quad (13)$$

this being valid because of the smallness of y_0 compared to atomic dimensions. The energy shift therefore becomes

$$\Delta E=\hbar c(FG_0-F_0G)_{y0} \quad (14)$$

with the wave functions obtained by solving the corresponding set of differential Eqs. (6) or (7). The solutions are matched and evaluated at $y=y_0$ and substituted into (14).

Schawlow and Townes[4] have evaluated ΔE for five heavy elements assuming a uniformly charged nucleus with $r_0=1.5\times10^{-13}$ cm as well as a "shell model" (same radius) with the charge all on the surface. The results for the uniform model expressed as a fraction of the total fine structure splitting ($\Delta E/h\Delta\bar{\nu}$) are presented in the first column of Table I. These results are subject to a correction since they were obtained using approximate normalization of the point nucleus wave functions. Wertheim and Igo[9] have obtained a more accurate normalization factor; their work shows that the values of $\Delta E/h\Delta\bar{\nu}$ in Table I should be multiplied by a factor of about 0.75.

Recent experiments in high-energy electron scattering[10] and mesonic x-rays[11] indicate that the nuclear charge distribution is approximately uniform with a rather diffuse boundary having a root-mean-square radius given by $R=1.2\times10^{-13}A^{\frac{1}{3}}$ cm. It is therefore necessary to modify the values of $\Delta E/h\Delta\bar{\nu}$ in Table I to correspond to this smaller radius as well as to consider the effect of a diffuse boundary. Ford and Hill[3] have shown that the perturbation method is sufficiently accurate for calculating the relative effect of different models on ΔE and that the x-ray fine structure is rather insensitive to the details of the proton distribution near the nuclear surface as long as the amount of "diffuseness" is small. The results of Schawlow and Townes have therefore been corrected for a radius of $r_0=1.2\times10^{-13}$ cm using Eq. (4) retaining the assumption of a uniform nuclear charge density. These modified values of $\Delta E/h\Delta\bar{\nu}$ are also shown in Table I where the normalization correction factor of 0.75 has been included. Schawlow and Townes noted that the numerical values of $\Delta E/h\Delta\bar{\nu}$ have an exponential Z dependence given by

$$\Delta E/h\Delta\bar{\nu}=De^{b(Z-60)}; \quad (15)$$

the values in the second column of Table I are therefore well represented by Eq. (15) with $D=54\times10^{-6}$ and $b=0.0878$.

TABLE I. The fractional change in fine structure ($\Delta E/h\Delta\bar{\nu}$) due to a uniformly charged nucleus. The values in the first column are taken from reference 4 which were calculated using $r_0=1.5\times10^{-13}$ cm and approximate normalization. The values in the second column have been adjusted for $r_0=1.2\times10^{-13}$ cm and corrected for normalization.

Z	$r_0=1.5\times10^{-13}$ cm	$r_0=1.2\times10^{-13}$ cm
60	1.09×10^{-4}	5.46×10^{-5}
70	2.51×10^{-4}	1.28×10^{-4}
81	6.35×10^{-4}	3.32×10^{-4}
90	1.38×10^{-3}	7.40×10^{-4}
95	2.17×10^{-3}	1.18×10^{-3}

Calculation of the Fine Structure Splitting

The calculation of the energy difference between the L_{II} and L_{III} x-ray levels has been carried out with a fair degree of accuracy by Christy and Keller.[5] Their purpose was to obtain a value of the fine structure constant α through a comparison of measured values of the splitting with the theoretical values. Assuming a point nucleus and employing relativistic wave functions they allowed for electron-electron interactions using the Breit Hamiltonian and perturbation methods.

The $2p_{\frac{1}{2}}$–$2p_{\frac{3}{2}}$ spin doublet splitting of electron energy levels in a pure Coulomb field is given in units of the rydberg by the familiar Sommerfeld formula, designated here as $(2/\alpha^2)S(\alpha Z)$. The first-order perturbation correction for the electron screening effects was computed allowing for the interactions of the electrons of the K and L shells and the effect of the M shell on the $2p_{\frac{1}{2}}$ and $2p_{\frac{3}{2}}$ states. Christy and Keller show that the corrections to be added to the Sommerfeld formula may be expressed in the form,

$$-2\alpha^2Z^3f(\alpha Z)+BZ^2-0.0356\alpha^4Z^5, \quad (16)$$

where $f(\alpha Z)$ is a slowly varying function of order unity which can be obtained for specific values of Z by interpolation from a table in their paper. The second

[9] M. S. Wertheim and G. Igo, Phys. Rev. **98**, 1 (1955).
[10] Hofstadter, Hahn, Knudsen, and McIntyre, Phys. Rev. **95**, 512 (1954).
[11] V. L. Fitch and J. Rainwater, Phys. Rev. **92**, 789 (1953).

term in Eq. (16) represents an estimate of the contribution of second-order effects; B is an unknown constant whose magnitude is adjusted by least squares when theory and experiment are compared. The third term is an estimate of the correction needed to compensate for omission of higher order terms in the interaction Hamiltonian. The complete expression for the calculated fine structure splitting in rydbergs may be written in the form

$$\Delta\nu/R=\phi(\alpha Z)+BZ^2, \tag{17}$$

where $\phi(\alpha Z)$ is the sum of the Sommerfeld formula and the first and third terms of Eq. (16).

Christy and Keller claim only about 1% accuracy in the screening corrections since all electron shells were not included in the calculations. While the errors are most probably of low enough Z dependence that they would not mask the nuclear size effect, the Schawlow-Townes method could yield more definitive results if these errors could be minimized. A different approach is being undertaken by S. Cohen[12] who is making relativistic self-consistent wave-function calculations for several heavy elements, but these computations have not progressed to the point where they can be applied to this problem.

Quantum Electrodynamic Corrections

The effects other than nuclear size will be enlarged upon briefly here.

(1) The spin-orbit interaction which produces the fine structure splitting may be regarded classically as arising from two effects[13]; the Larmor precession which is proportional to μ_0, the electron magnetic moment, and electromagnetic in origin, and the Thomas precession which is proportional to $-\mu_0/2$ and relativistic in origin. In considering the effect of the anomalous magnetic moment upon the fine structure splitting, the correction should be applied only to that portion which is of electromagnetic origin since the anomalous part of μ_0 arises from the interaction of the electron-positron field with the electromagnetic field. The correction to the fine structure is therefore

$$\Delta\nu\sim\left[\mu_0\left(1+\frac{\alpha}{2\pi}\right)-\frac{\mu_0}{2}\right]=\frac{\mu_0}{2}\left(1+\frac{\alpha}{\pi}\right), \tag{18}$$

where $(1+\alpha/2\pi)$ is the anomalous moment correction to first order. The theoretical value of the splitting is increased by this factor, but when theory and experiment are compared it is more convenient to decrease the experimental values by the same factor.

(2) The phenomenon known as vacuum polarization arises from creation of virtual electron-positron pairs in the strong field near the nucleus. This induced charge causes deviations from Coulomb's law which modifies the energy levels of electrons which have a finite probability of being within a Compton wavelength of the nucleus. Uehling[14] first calculated the potential due to this induced charge to first order in αZ as well as the displacement of the $1s$ and $2s$ electronic levels using perturbation techniques. The perturbation method breaks down, however, both for the calculation of higher order terms in the potential and for the calculation of the effect on the electron energy levels when αZ is close to one. Wichmann and Kroll[7] made a more detailed study of vacuum polarization which avoids a perturbation expansion. They find that the correction to the Uehling potential for the case of the $2p_{\frac{1}{2}}$ and $2p_{\frac{3}{2}}$ level displacements is small enough to neglect. They also made an accurate evaluation of the contribution of vacuum polarization to the splitting of the two levels. Expressed as a fraction of the fine structure the correction has the form

$$\delta_p{}^{(1)}/(\Delta\nu/R)=Ve^{c(Z-60)}, \tag{19}$$

where $\delta_p{}^{(1)}$ is the contribution in rydbergs obtained from Table I of reference 7, $V=173\times10^{-6}$, and $c=0.0462$. This effect is thus of the same order of magnitude as the nuclear size effect, and therefore the two might be expected to partially cancel one another.

(3) The third effect is due to the interaction of the electron with its own radiation field. This effect for the most part, causes the $2S_{\frac{1}{2}}$ level displacement in hydrogen (generally known as the Lamb shift). The correction in this case has been calculated with high accuracy, but for the heavy elements the usual difficulties are encountered because the expansion parameter αZ is nearly unity. For this reason there exists no quantitative evaluation of the Lamb shift for x-ray levels. However, Wichmann, who has been working on the problem, estimates that the effect on the fine structure splitting is about the same order as the vacuum polarization effect but of opposite sign.[15]

III. EXPERIMENTAL CONSIDERATIONS

Measurement of the Fine Structure Splitting

The L_{II}–L_{III} energy level difference can be measured in several different ways. Figure 1 is an x-ray energy level diagram (not complete or to scale) showing the various pairs of transitions whose energy difference equals the desired fine structure splitting. Of the six possibilities shown only one seems feasible for high precision work—the L_{α_2} and L_{β_1} lines which have a common termination on the M_{IV} level. Having the highest intensities of the usable L transitions, these appear to be the logical choice. The K transitions, while much more intense, are of such high energy (about 100 kev) that the additional experimental problems offset the advantage gained in intensity.

[12] S. Cohen, Bull. Am. Phys. Soc. Ser. II, 2, 309 (1957).

[13] The author acknowledges helpful discussion and correspondence concerning this correction with Professor Christy, Professor DuMond, and Professor Townes.

[14] E. A. Uehling, Phys. Rev. 48, 55 (1935).

[15] E. H. Wichmann (private communication).

The measurement of transition energy involves the measurement of the wavelength of the radiation which in turn means the measurement of a Bragg angle. In order to estimate the precision needed in the angle measurements let us assume that the desired precision in the fine structure splitting is 10% of the predicted nuclear size effect and that elements between $Z=70$ and $Z=94$ are to be studied using a calcite crystal as an x-ray grating. Calculations based on the Bragg law and Eq. (15) yield the result that for the smaller values of Z the angles should be measured to better than 0.1 sec of arc and for the larger values of Z an accuracy of 0.5 sec is needed. This indicates that the greatest possible care must be taken in the measurements, because one or two tenths of a second is about the limit of accuracy in determining Bragg angles using present day x-ray spectrometers.

The experimental work on the latest precision measurement of the x-ray fine structure has been described elsewhere.[8,16]

X-Ray Spectroscopy Techniques

The Bragg angles of the $L_{\alpha 2}$ and $L_{\beta 1}$ x-ray lines of the elements W, Pt, Bi, Th, U, and Pu were measured using DuMond's[17] two-crystal spectrometer. The calcite crystals used were carefully ground and etched on their reflecting surfaces so that the highest resolution consistent with reasonable luminosity could be obtained.

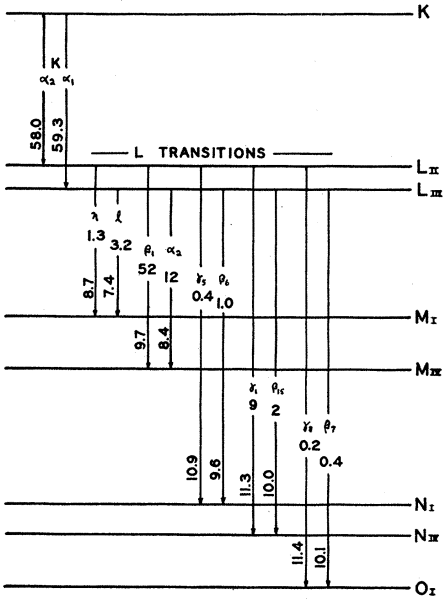

FIG. 1. X-ray energy level diagram (not complete or to scale) showing the transitions giving the L_{II}–L_{III} separation. The numbers alongside the lines are the approximate energies in kilovolts for $W(Z=74)$; the numbers beneath the L-line designations are the intensities relative to an intensity of 100 for $L_{\alpha 1}$.

[16] R. L. Shacklett, U. S. Atomic Energy Commission Special Technical Report No. 23, California Institute of Technology, July 1956 (unpublished).

[17] J. W. M. DuMond and D. Marlow, Rev. Sci. Instr. 8, 112 (1937).

The resolution of the two-crystal spectrometer is not determined by a slit system; the crystals themselves determine the resolution, the incident radiation being collimated according to wavelength by the first crystal and analyzed by the second crystal. Lead slits and stops are used but only to confine the radiation in the desired direction.

Each crystal of the spectrometer is mounted on a vertical axis such that its reflecting surface is parallel to the axis of revolution. Rotation is accomplished by worm-wheel drives utilizing worm gears which have been specially lapped and optically calibrated. In the precision measurement of the Bragg angle of an x-ray line the first crystal (crystal A) is adjusted so that the incoming radiation[18] is incident at the Bragg angle. The body of the spectrometer is then rotated so that it brings the axis of crystal B into the beam of radiation reflected from crystal A. With B also adjusted to approximately the Bragg angle (the "antiparallel" position) the radiation undergoes a second reflection and is detected by an x-ray detector, in this case a sodium iodide crystal and photomultiplier tube. Crystal B is then rotated through small angular increments (five to twenty seconds at a time, depending on the width of the line) with the intensity of radiation as measured by the detector recorded at each position. The center of the curve that is obtained by plotting intensity *versus* angle represents the center of the x-ray line. Crystal B is then rotated around until its reflecting surface is parallel to that of crystal A. This "parallel position" is one which allows two successive reflections of a relatively wide band of wavelengths only if the crystals are exactly parallel within the angular width of their diffraction patterns. The parallel position is thus a reference position from which the Bragg angle θ of the x-ray line can be measured, the angular difference between the two positions being $180°-2\theta$.

In obtaining the spectrometer reading corresponding to the center of the x-ray line profile at the antiparallel position the intensity values were fitted to a theoretical curve which took into consideration the perturbing effects of the vertical divergence of the radiation. Although a rather laborious process the method is worthwhile since the center position can be determined to about 1/400 of the full width of the line at half-maximum height.

Experimental Results

Several corrections must be made to the angle determined from the difference between the parallel and antiparallel positions: (1) worm-wheel errors, (2) temperature above 18°C, (3) shift due to asymmetry in crystal diffraction pattern.[8] Four independent runs were made for each line, and the results averaged with equal weight to give a mean Bragg angle for each of the

[18] The source radiation is obtained by x-ray fluorescence.

TABLE II. Measured values of the fine structure splitting.

Element	Rydbergs	Electron volts	Relative S.D. (ppm)
$_{74}$W	98.275±0.004	1337.02±0.05	37
$_{78}$Pt	125.599±0.008	1708.76±0.11	64
$_{83}$Bi	168.511±0.007	2292.58±0.10	42
$_{90}$Th	249.363±0.009	3392.56±0.12	34
$_{92}$U	277.934±0.009	3781.27±0.13	33
$_{94}$Pu	309.435±0.011	4209.83±0.15	36

12 lines. The standard deviation of the mean in each case averages about 0.2 sec. This uncertainty in Bragg angle represents an uncertainty in the fine structure splitting of about 20% of the nuclear size effect in the case of W ($Z=74$) and about 4% in the case of Pu ($Z=94$). The wavelengths computed from the Bragg angles have an average relative standard deviation of about five parts per million. Table II gives the values of the fine structure splitting in rydbergs and in electron volts calculated from the difference in transition energies of the L_{α_2} and L_{β_1} x-ray lines. Shown also are the relative standard deviations which average about 40 parts per million representing an increase in accuracy of about a factor of 10 over the values available to Schawlow and Townes.

IV. COMPARISON WITH THEORY

The calculation of the theoretical splitting for the case of a point nucleus using Christy and Keller's formula (17) involves interpolating $f(\alpha Z)$ from a table. The logarithm of $f(\alpha Z)$ when plotted as a function of $1-\sigma$ is almost linear. A power series based on Lagrange interpolation was used and has the following form:

$$\log[10f(\alpha Z)]=C_0+C_1x+C_2x^2-C_3x^3+C_4x^4, \quad (20)$$

where $x=100(1-\sigma)$, $C_0=0.6794690$, $C_1=6.83439\times10^{-3}$, $C_2=2.648\times10^{-5}$, $C_3=5.26\times10^{-7}$, $C_4=1.09\times10^{-8}$. The remaining terms appearing in the function $\phi(\alpha Z)$ can be calculated with arbitrary precision.

Schawlow and Townes have shown that the comparison of the theoretical and experimental values of the fine structure splitting can best be illustrated by a plot of their relative deviations. In the case of a point nucleus (no nuclear size or quantum electrodynamic corrections) the relative deviation may be expressed in the form,

$$\frac{\text{theoret-exptl}}{\text{exptl}}=\frac{BZ^2}{\Delta\nu/R}-\left(1-\frac{\phi(\alpha Z)}{\Delta\nu/R}\right), \quad (21)$$

where $\Delta\nu/R$ represents the values of the fine structure splitting appearing in Table II modified to correct for the anomalous moment of the electron [Eq. (18)]. The constant B is adjusted by least squares to minimize the error-squared sum obtaining $B=4.71648\times10^{-4}$. The plot of the deviations in Fig. 2 (open circles) shows clearly that a large systematic error remains which varies with Z in approximately exponential fashion.

In order to include the nuclear size effect in Eq. (21) we subtract a term having the form of Eq. (15). Assuming for the moment that the discrepancy indicated in Fig. 2 can be attributed entirely to a uniformly charged nucleus of unknown radius we adjust the values of B and D by least squares (with $b=0.0878$) obtaining $B=4.81722\times10^{-4}$ and $D=44\times10^{-6}$. The resulting deviations are also plotted in Fig. 2 using solid circles. The standard deviation of D computed from these deviations is 3.5 ppm or about 8%. If this value of D is interpreted in terms of nuclear size we obtain, using Eq. (4),

$$r_0=(1.08\pm0.05)\times10^{-13}\text{ cm} \quad (22)$$

for the nuclear radius constant. The fact that this value of r_0 is smaller than 1.2×10^{-13} cm suggests that the net effect of vacuum polarization and the Lamb shift is to increase the fine structure splitting and hence cause the nucleus to appear smaller.

If we now make explicit corrections for a nuclear radius with $r_0=1.2\times10^{-13}$ cm and for vacuum polarization using Eq. (19) the plot of relative deviations would show the magnitude and possible Z dependence of an additional correction term. With B again adjusted by least squares to 4.71095×10^{-4} the deviations shown in Fig. 3 (open circles) suggest that the correction term could be exponential in Z. In the absence of a quantitative evaluation of the Lamb shift for heavy nuclei it is tempting to suggest that this remaining discrepancy

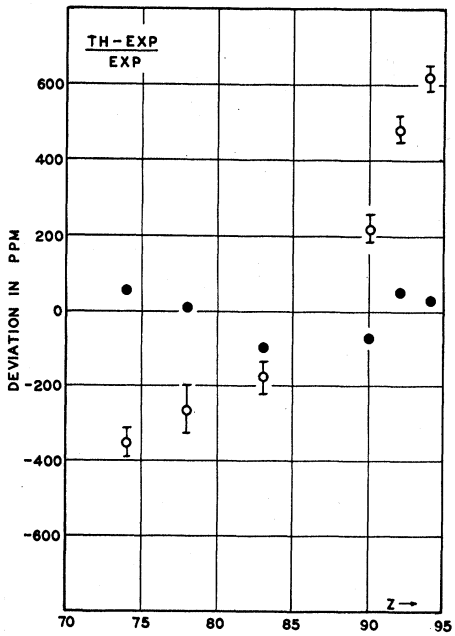

FIG. 2. Relative deviation of theory and experiment without the nuclear size or quantum electrodynamic corrections (open circles). Solid circles show the effect of adjusting by least squares the nuclear radius parameter D to correspond to $r_0=1.08\times10^{-13}$ cm. The relative standard deviation of the experimental measurements is shown on the open circles.

between theory and experiment might be caused by this effect.

There is, of course, the possibility that errors in the point nucleus formula (17) arising because of the approximate nature of Christy and Keller's calculations may be strongly Z dependent. Schawlow and Townes considered the magnitude of the error involved in assuming that the second-order screening effects are proportional to a constant times Z^2 and conclude that it is only a few percent of the nuclear size effect. Quantum electrodynamic effects in the electron-electron interaction have been estimated to order Z^5 by Christy and Keller and are included in Eq. (16); higher order terms may be assumed to be of much smaller magnitude.

We shall assume that the required correction term is exponential in Z and has the form $Le^{a(Z-60)}$. Adjusting B, a, and L by least squares we obtain $B=4.76450\times10^{-4}$, $a=0.115$, and $L=(10.9\pm0.9)\times10^{-6}$. The remaining root-mean-square deviation is 33 parts per million which is even smaller than the average experimental error; the assumption of an exponential term for the correction rather than another type of Z dependence therefore seems to be reasonable. The points are plotted as solid circles in Fig. 3.

The equation which gives the theoretical fine structure splitting may therefore be written out in full in the form

$$\Delta\nu/R=[2\alpha^{-2}S(\alpha Z)-2\alpha^2Z^3f(\alpha Z)-0.0356\alpha^4Z^5+BZ^2]\times[1+\alpha/\pi-De^{b(Z-60)}+Ve^{c(Z-60)}-Le^{a(Z-60)}], \quad (23)$$

with

$$B=4.76450\times10^{-4}$$
$$D=54\times10^{-6}, \qquad b=0.0878$$
$$V=173\times10^{-6}, \qquad c=0.0462$$
$$L=10.9\times10^{-6}, \qquad a=0.115.$$

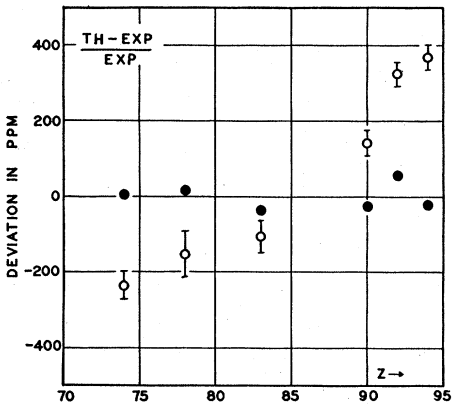

FIG. 3. Relative deviation of theory and experiment when corrections for a nuclear radius of $r_0=1.2\times10^{-13}$ cm and vacuum polarization are included (open circles). Solid circles show the effect of addition of the empirical (Lamb shift) correction term. The relative standard deviation of the experimental measurements is shown on the open circles.

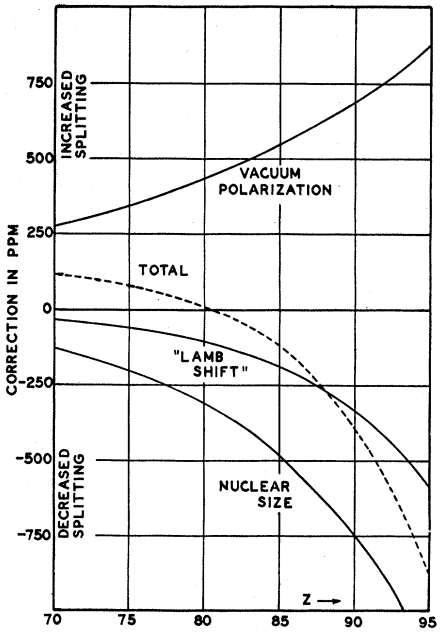

FIG. 4. The effect of the various correction terms on the fine structure splitting as given by the Christy-Keller point-nucleus formula.

The effect of the various correction terms on the fine structure splitting as given by the Christy-Keller formula is shown in Fig. 4.

V. CONCLUSIONS

The effect of finite nuclear size on the $2p_{\frac{1}{2}}$ level of heavy elements, while relatively small, is measurable with reasonable accuracy through precise measurements of x-ray fine structure splitting. Quantum electrodynamic effects which give rise to deviations from a Coulomb field near the nucleus produce similar systematic discrepancies between the calculated splitting for the case of a point nucleus and the measured values. Since the magnitude of one of these effects (the Lamb shift) can only be estimated at present, the method developed by Schawlow and Townes is not now applicable in a direct measurement of nuclear size. If instead, the nucleus is assumed to have an approximately uniform charge distribution of radius $R=1.2\times10^{-13}A^{\frac{1}{3}}$ cm as indicated by other more accurate experiments, it is possible to deduce from a comparison of the measured fine structure with the theoretical values corrected for nuclear size and vacuum polarization that the Lamb shift has a very strong Z dependence, similar to that of vacuum polarization and opposite in sign. A more accurate evaluation of the fine structure splitting for the case of a point nucleus such as might be obtained by a careful self-consistent field approach would make the results of the comparison of theory and experiment more definite than they are now and would also encourage greater precision in x-ray measurements.