THE
PHYSICAL REVIEW

A journal of experimental and theoretical physics established by E. L. Nichols in 1893

SECOND SERIES, VOL. 100, NO. 5 DECEMBER 1, 1955

Effect on X-Ray Fine Structure of Deviations from a Coulomb Field near the Nucleus*

A. L. SCHAWLOW† AND C. H. TOWNES
Columbia University, New York, New York
(Received May 20, 1955)

Deviations from a Coulomb field near the nucleus of very heavy atoms appear to produce changes as large as about one part in 400 on the fine structure splitting of the $2p$ electronic level. Theoretical evaluation of these changes is discussed and available x-ray data analyzed to yield an experimental determination of their magnitude. A nuclear radius as large as $2\times10^{-13}A^{\frac{1}{3}}$ cm must be assumed if the observed effects come only from deviations in the Coulomb field due to the finite nuclear size. The preferred interpretation is that most of the contribution to the observed change in fine structure comes from quantum electrodynamic effects (Lamb shift) which produce deviations from a Coulomb field near the nucleus. This appears to provide a method for study of the Lamb shift for large Z and possibly also of nuclear size.

INTRODUCTION

A BRIEF description of a method for determining deviations from a Coulomb field near a nucleus from x-ray data has already been given and interpreted in terms of an effective nuclear radius.[1] Two recent new experiments[2–5] on the distribution of charge in nuclei have made appropriate a more detailed discussion of this method so that a comparison of the several types of information now available can be made and certain deviations from a Coulomb field which are not directly connected with the nuclear size can be pointed out.

The "isotope shift" in atomic spectra is a well-known phenomenon. In heavy elements it is due to the finite size of nuclei and the variation in electronic energy levels with the small differences in effective nuclear size occurring in two or more isotopes. Usually the entire change in electronic energy between electron levels for the ideal point nucleus and those for a nucleus of finite size cannot be experimentally determined, so that one must be content with measuring only the small differences in effective nuclear size through the isotope shift.

Precise measurements of the fine structure splitting between the $2p_{\frac{1}{2}}$ and $2p_{\frac{3}{2}}$ levels of heavy atoms allow an experimental determination of the entire change in electron energy due to finite nuclear size, plus that due to any other deviation from a Coulomb field occurring in the immediate vicinity of the nucleus. Such deviations may be due to vacuum polarization or to other electrodynamic effects, usually called "Lamb shifts," in the very strong fields near the nucleus.

Sensitivity of the fine structure to the above effects is due to the following circumstances. Although the $2p_{\frac{3}{2}}$ wave function is essentially zero everywhere inside or within a few nuclear radii of the nucleus, the $2p_{\frac{1}{2}}$ wave function for a heavy atom has an appreciable value in this region, because of what can be regarded as relativistic mixing of the $2p_{\frac{1}{2}}$ and $2s_{\frac{1}{2}}$ functions. This modifies the spacing between the L_{II} and L_{III} levels by a small amount which increases very rapidly with increasing Z. Fortunately, screening effects are small enough for the $2p_{\frac{1}{2}}$ and $2p_{\frac{3}{2}}$ wave functions that they can be allowed for. Hence the fine structure splitting assuming a pure Coulomb field can be calculated with some accuracy and compared with experimental measurements. The discrepancy between calculated and measured values increases with Z in the way expected for the effect of a deviation from a Coulomb field in the vicinity of the nucleus and very much more rapidly than any terms expected from screening or most other electronic effects. Vacuum polarization and other deviations from a Coulomb field which occur in the very strong electric fields near the nucleus of course give

* Work supported by the U. S. Atomic Energy Commission.
† Now at Bell Telephone Laboratories, Inc., Murray Hill, New Jersey.
[1] A. L. Schawlow and C. H. Townes, Science **115**, 284 (1952).
[2] V. L. Fitch and J. Rainwater, Phys. Rev. **92**, 789 (1953).
[3] L. N. Cooper and E. M. Henley, Phys. Rev. **92**, 801 (1953).
[4] Hofstadter, Fechter, and McIntyre, Phys. Rev. **92**, 978 (1953).
[5] L. I. Schiff, Phys. Rev. **92**, 988 (1953).

similar effects. The rapid variation of this discrepancy then allows isolation of the sum of effects due to nuclear size and the Lamb shift from less interesting effects. The increasing amount of information on the nuclear charge distribution from other experiments[2-5] may allow a sufficiently accurate determination of effects due to nuclear size so that the x-ray fine structure shift can be used to determine higher order electrodynamic corrections for the case of large Z.

The distribution of electrons in $2p_{\frac{1}{2}}$ and $2p_{\frac{3}{2}}$ states of a heavy atom are indicated in Figs. 1 and 2. In Fig. 1, the probability of finding an electron at a given distance r from the nucleus is plotted as a function of r. The similarity of these probability distributions is important in allowing the difference between their energies to be calculated with some accuracy, even in the presence of screening. Figure 2 gives $|\psi|^2$, the electron probability density, as a function of r. Here it is evident that the two wave functions differ radically near the nucleus. A $2p_{\frac{1}{2}}$ wave function gives the electron a very large probability of being found in the immediate vicinity of the nucleus, while for a $2p_{\frac{3}{2}}$ wave function this probability is very small.

CALCULATION OF THE CHANGE IN ELECTRONIC ENERGY DUE TO FINITE NUCLEAR SIZE

Although the change in electronic energy due to finite nuclear size is very small compared with the total electron energy, it cannot be accurately calculated by a first-order perturbation technique. In the small region inside the nucleus the perturbing potential is so enormous that a first-order calculation is wrong by about 30%. The almost exact method of treatment used here has been published by Broch[6]; however, it will be outlined below since it is not well known or readily available. The two-component relativistic wave equations may be written[7]

$$\frac{d\phi_1}{dy}-\frac{j}{y}\phi_1=\frac{1}{2a}\left(1-\frac{E-V}{mc^2}\right)\phi_2, \tag{1}$$

$$\frac{d\phi_2}{dy}+\frac{j}{y}\phi_2=\frac{1}{2a}\left(1+\frac{E-V}{mc^2}\right)\phi_1, \tag{2}$$

where j is a quantum number which is $+1$ for $p_{\frac{1}{2}}$ states and -2 for $p_{\frac{3}{2}}$ states, $y=2Zr/a$, r=distance from the center of the nucleus, a=Bohr radius of hydrogen, and E and V are the total energy and potential energy respectively. The corresponding equations for a hypothetical point nucleus are

$$\frac{d\phi_2^{(0)}}{dy}-\frac{j}{y}\phi_1^{(0)}=\frac{1}{2a}\left(1-\frac{E^{(0)}-V^{(0)}}{mc^2}\right)\phi_2^{(0)}, \tag{3}$$

$$\frac{d\phi_2^{(0)}}{dy}+\frac{j}{y}\phi_2^{(0)}=\frac{1}{2a}\left(1+\frac{E^{(0)}-V^{(0)}}{mc^2}\right)\phi_1^{(0)}. \tag{4}$$

[6] E. K. Broch, Arch. Math. Naturvidenskab 48, 25 (1945).
[7] J. E. Rosenthal and G. Breit, Phys. Rev. 41, 459 (1932).

If we multiply Eq. (1) by $\phi_2^{(0)}$, Eq. (2) by $-\phi_1^{(0)}$, Eq. (3) by $-\phi_2$ and Eq. (4) by ϕ_1 and add, we obtain

$$\frac{d}{dy}(\phi_1\phi_2^{(0)}-\phi_2\phi_1^{(0)})=\frac{1}{2a}(\phi_1\phi_1^{(0)}+\phi_2\phi_2^{(0)})\left(\frac{\Delta V-\Delta E}{mc^2}\right), \tag{5}$$

where $\Delta V=V-V^{(0)}$ and $\Delta E=E-E^{(0)}$. Integrating from y_0 to ∞, and remembering that outside the nucleus $\Delta V=0$,

$$2Z\alpha mc^2(\phi_1\phi_2^{(0)}-\phi_1^{(0)}\phi_2)_{y0}=\Delta E\int_{y0}^{\infty}(\phi_1\phi_1^{(0)}+\phi_2\phi_2^{(0)})dy. \tag{6}$$

But

$$\int_{y0}^{\infty}(\phi_1\phi_1^{(0)}+\phi_2\phi_2^{(0)})dy\approx\int_0^{\infty}(\phi_1\phi_1^{(0)}+\phi_2\phi_2^{(0)})dy\approx\frac{2Z}{a}\int_0^{\infty}[(\phi_1^{(0)})^2+(\phi_2^{(0)})^2]dr=\frac{2Z}{a}; \tag{7}$$

hence, from (6) and (7),

$$\Delta E=\frac{hc}{2\pi}(\phi_1\phi_2^{(0)}-\phi_1^{(0)}\phi_2)_{y0}. \tag{8}$$

The energy level shift, ΔE, due to the finite nuclear size is obtained by substituting the values of ϕ_1, ϕ_2, $\phi_1^{(0)}$, and $\phi_2^{(0)}$ at the nuclear radius, y_0, from the solutions of Eqs. (1), (2), (3), and (4) into Eq. (8).

Near the nucleus, $1-E/mc^2\ll V/mc^2$, so that one may assume $E=mc^2$ and the solutions of (1) and (2) for $y\geq y_0$ are:

$$\phi_1/a=C_1[J_{2\gamma}(2y^{\frac{1}{2}})+C_2J_{-2\gamma}(2y^{\frac{1}{2}})] \tag{9}$$

and

$$\phi_2=C_1[A_{2\gamma}(2y^{\frac{1}{2}})+C_2A_{-2\gamma}(2y^{\frac{1}{2}})], \tag{10}$$

where the J's are Bessel functions, C_1 and C_2 are constants, $\gamma=(j-Z^2\alpha^2)^{\frac{1}{2}}$, $A_{2\gamma}=(j-\gamma)J_{2\gamma}+y^{\frac{1}{2}}J_{2\gamma+1}$, and $A_{-2\gamma}=(j-\gamma)J_{-2\gamma}-y^{\frac{1}{2}}J_{-2\gamma-1}$. For a point nucleus, $C_2=0$; its value for a finite nucleus is determined by matching at $y=y_0$ these solutions to those for the region inside the nucleus. The latter may be obtained for any assumed intra-nuclear potential V as a power series since y_0 is small ($y_0\leq 0.05$). That is, inside the nucleus, the solutions may be written

$$\phi_1=\alpha y+\beta y^3+\gamma y^5+\delta y^7+\cdots, \tag{11}$$

$$\phi_2=by^2+cy^4+dy^6+ey^8+\cdots \tag{12}$$

It is through influence on these solutions, and hence on the constant C_2, that the nuclear charge distribution enters the problem.

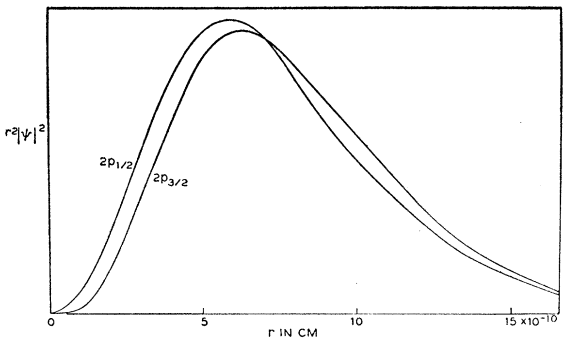

FIG. 1. Probability $r^2\ |\psi|^2$ of finding a $2p$ electron a given radius r from the nucleus with $Z=85$. (Note: "10^{-10}" should read "10^{-11}".)

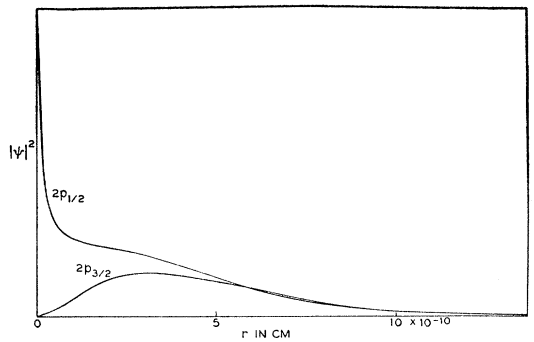

FIG. 2. The electron density $|\psi|^2$ for a $2p$ electron when $Z=85$. (Note: "10^{-10}" should read "10^{-11}".)

If expressions (11) and (12) are substituted into Eqs. (1) and (2), the coefficients may all be evaluated for any particular nuclear charge distribution in terms of one constant, α. Then $\phi_1/a\phi_2$ at $y=y_0$ is uniquely determined by the solutions inside the nucleus. Equating it to the corresponding quantity expressed in terms of the solutions outside the nucleus,

$$C_2=\frac{(\phi_1/a\phi_2)_{y=y_0}A_{2\gamma}(2y_0^{\frac{1}{2}})-J_{2\gamma}(2y_0^{\frac{1}{2}})}{J_{-2\gamma}(2y_0^{\frac{1}{2}})-(\phi_1/a\phi_2)_{y=y_0}A_{-2\gamma}(2y_0^{\frac{1}{2}})}. \quad (13)$$

Substituting the solutions (9) and (10) into (8), the energy level displacement is

$$\Delta E=Ze^2C_1^2C_2y_0^{\frac{1}{2}}[J_{2\gamma+1}(2y_0^{\frac{1}{2}})J_{-2\gamma}(2y_0^{\frac{1}{2}}) +J_{2\gamma}(2y_0^{\frac{1}{2}})J_{-2\gamma-1}(2y_0^{\frac{1}{2}})], \quad (14)$$

and using the properties of the Bessel functions

$$\Delta E=-2Ze^2C_1^2C_2\frac{\gamma}{\Gamma(1+2\gamma)\Gamma(1-2\gamma)}. \quad (15)$$

The normalization constant, C_1 may be obtained from $\Delta\nu$, the fine structure splitting, when the principal quantum number n is large.[7] It is

$$C_1^2=\frac{h\Delta\nu}{Ze^2\{[(l+1)^2-a^2]^{\frac{1}{2}}-1-(l^2-a^2)^{\frac{1}{2}}\}}, \quad (16)$$

so that from (15)

$$\Delta E=\frac{2C_2}{[(l+1)^2-a^2]^{\frac{1}{2}}-1-(l^2-a^2)^{\frac{1}{2}}} \times\frac{\gamma h\Delta\nu}{\Gamma(1+2\gamma)\Gamma(1-2\gamma)}. \quad (17)$$

ΔE can hence be obtained as a given fraction of the fine structure splitting $\Delta\nu$ if C_2 is calculated from (13). It must be noted that expression (16) is not especially accurate for the present case, where $n=2$. However, it gives a convenient first approximation to C_1^2.

Somewhat more accurate values of C_1^2 might be obtained by using Dirac wave functions with an appropriately screened value of the effective nuclear charge Ze. C_1^2 may be still better determined by obtaining the wave function for a $2p_{\frac{1}{2}}$ electron in the field of a nucleus of charge Ze and a screening charge of the other electrons with a Fermi-Thomas distribution. From a calculation of the latter type, Wertheim and Igo[8] have recently obtained a normalization factor for the $2p_{\frac{1}{2}}$ electron when $Z=92$. Their value for C_1^2 is 0.76 of its value for a hydrogenic wavefunction with $Z=92$, or 0.74 times the value given in (16). K. M. King is engaged in a still more precise calculation of the $2p_{\frac{1}{2}}$ wave function which, however, is not yet complete. For convenience, values of C_1^2 from (16) or of ΔE given by (17) will be used in much of the following discussion, and an appropriate correction for the normalization will be applied later.

The shift of the $p_{\frac{3}{2}}$ level due to the finite nuclear size is negligible in comparison with that of the $p_{\frac{1}{2}}$ level, so that the calculated displacement of the $p_{\frac{1}{2}}$ level is the entire change in the fine structure splitting due to nuclear size.

MAGNITUDE OF EFFECTS OF NUCLEAR SIZE

Before discussing results obtained by the somewhat complex method of calculation described above, a few general observations about the nature of the expected results may be useful. The perturbing energy term due to a finite nuclear size has the following form

$$\epsilon=\int\rho_n(V-V_0)dv, \quad (18)$$

where ρ_n is the density of nuclear charge, V the potential due to the electron charge distribution, and V_0 its value at the center of mass of the nucleus. If the electron charge density is a constant ρ_e over the nucleus, then (18) has the form

$$\epsilon=-\frac{2\pi}{3}\int\rho_n\rho_e r^2dv, \quad (19)$$

[8] M. S. Wertheim and G. Igo, Phys. Rev. 98, 1 (1955).

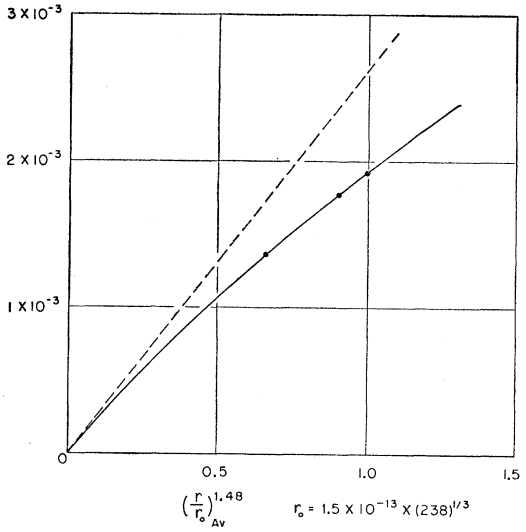

FIG. 3. Ratio of nuclear size effect to fine structure ($\Delta E/h\Delta\nu$) as a function of $(r^{1.48})_{Av}$ for $Z=92$. Normalization was obtained by using a screening factor of 5.5. Dotted line corresponds to result of first-order perturbation theory.

and the perturbation ϵ is proportional to the mean square radius $(r^2)_{Av}$, since

$$(r^2)_{Av}=\frac{\int \rho_n r^2 dv}{\int \rho_n dv}=\frac{\int \rho_n r^2 dv}{Ze}. \quad (20)$$

Actually, very near the nucleus the Dirac wave function varies as r^{n-1}, where $n=(1-\alpha^2Z^2)^{\frac{1}{2}}$, so that ρ_e varies as r^{2n-2} and therefore $V-V_0$ as r^{2n}. Hence ϵ is proportional to $(r^{2n})_{Av}$. When $Z=92$, this corresponds to $(r^{1.48})_{Av}$ rather than $(r^2)_{Av}$.

If ϵ is sufficiently small, first-order perturbation theory is appropriate and the change in energy ΔE due to the nuclear size should be proportional to $(r^{2n})_{Av}$. If ϵ is not sufficiently small, then ΔE may be expected to be some function of $(r^{2n})_{Av}$ (independent of other details of the radial distribution of charge). Since ϵ represents an increase in energy as $(r^{2n})_{Av}$ increases, the electron wave function at the nucleus will be somewhat decreased, and deviations from first-order perturbation theory can be expected in the direction of making the energy change ΔE somewhat smaller than that given by first-order theory and to depend on $(r^{2n})_{Av}$ somewhat more slowly than linearly.

Calculations of $\Delta E/h\Delta\nu$ for a $2p_{\frac{3}{2}}$ electron have been made by the method described above for five values of Z between 60 and 95 assuming a nuclear radius of $1.5\times10^{-13}A^{\frac{1}{3}}$ and either a uniform charge distribution or a surface charge only. The resulting values are listed in Table I.

A more accurate normalization, using the screening of a Thomas-Fermi electron distribution factor as mentioned above, decreases all values of Table I by a factor of about 0.76. Appropriate values of the effect of other nuclear models with different radii of radial distributions can be obtained by interpolation or extropolation of the information in Table I.

The variation of $\Delta E/h\Delta\nu$ with Z cannot be very easily predicted from qualitative arguments. However, it is found that the results of Table I can be rather accurately represented by the simple expression

$$\Delta E/h\Delta\nu=De^{b(Z-60)}, \quad (21)$$

where $D_1=1.09\times10^{-4}$ and $b_1=0.0858$ for the uniformly charged nucleus. $D_2=1.72\times10^{-4}$ and $b_2=0.0817$ for the charge on the nuclear surface. It must again be remembered that the values of D here come from use of Eq. (16). This normalization is not very accurate, but should give the correct functional form of $\Delta E/h\Delta\nu$.

The very rapid increase in $\Delta E/h\Delta\nu$ with Z given by (21) is a valuable characteristic of this effect, which allows it to be distinguished from less interesting effects which vary more slowly with Z. For Z between 70 and 92, $\Delta E/h\Delta\nu$ increases approximately as Z^7 or ΔE as Z^{11}.

For $Z=92$, Fig. 3 indicates how $\Delta E/h\Delta\nu$ varies with increasing $(r^{1.48})_{Av}$. The dotted line shows the result of a first-order perturbation calculation, which is in considerable error for the case of interest. It may be seen from this figure that, as expected, $\Delta E/h\Delta\nu$ increases somewhat more slowly than linearly with $(r^{1.48})_{Av}$.

THEORY OF THE FINE STRUCTURE SPLITTING

Calculation of the fine structure splitting between the $2p_{\frac{1}{2}}$ and $2p_{\frac{3}{2}}$ (L_{II} and L_{III}) x-ray levels has been described in some detail by Christy and Keller.[9] They allow for interaction between the various atomic electrons by using the Breit Hamiltonian[10] for interelectron interactions. They show that the fine structure may be expanded in the form

$$h\Delta\nu_0=mc^2S(\alpha Z)\times\left[1-\frac{1}{\alpha Z}f_1(\alpha Z)+\frac{1}{(\alpha Z)^2}f_2(\alpha Z)+\cdots\right], \quad (22)$$

TABLE I. The fractional change ($\Delta E/h\Delta\nu$) in fine structure due to finite nuclear size. Nuclear radius is assumed to be 1.5×10^{-13} $A^{\frac{1}{3}}$ cm. Normalization is obtained from expression (16).

Z	For uniformly charged nucleus	For nucleus with surface charge only
60	1.09×10^{-4}	1.73×10^{-4}
70	2.51×10^{-4}	3.92×10^{-4}
81	6.35×10^{-4}	9.56×10^{-4}
90	1.38×10^{-3}	2.03×10^{-3}
95	2.17×10^{-3}	3.01×10^{-3}

[9] R. F. Christy and J. M. Keller, Phys. Rev. **61**, 147 (1942).
[10] G. Breit, Phys. Rev. **34**, 553 (1929).

where $S(\alpha Z)$ is the Sommerfeld expression for the fine structure splitting in a hydrogenic atom and f_1 and f_2 are slowly varying functions of order unity. Christy and Keller calculate $f_1(\alpha Z)$ and approximate the small term involving f_2 by assuming that f_2 is a constant of unknown value. Since $S(\alpha Z)$ is approximately proportional to $(\alpha Z)^4$, (22) may be written

$$\frac{\Delta\nu_0}{R}=\frac{2}{\alpha^2}S(\alpha Z)-2\alpha^2Z^3f(\alpha Z)+BZ^2, \qquad (23)$$

where R is the Rydberg, the first term is the Sommerfeld expression for the fine structure, $f(\alpha Z)$ can be obtained from the work of Christy and Keller, and B is an unknown constant.

Christy and Keller's calculation was made in order to obtain a value of the fine structure constant α by use of the x-ray fine structure measurements and an expression such as (23). No effects due to the finite size of the nucleus were included in their considerations. Bethe and Longmire[11] applied a correction factor $(1+\alpha/2\pi)^{-1}$ to Christy and Keller's value of α in order to allow approximately for quantum electrodynamic effects on the electron moment. The resulting value of α obtained was not in good agreement with other determinations. However, we shall see below that if the effects of finite nuclear size are allowed for, the value of α obtained agrees excellently with that obtained by other methods.

COMPARISON BETWEEN THEORY AND EXPERIMENTAL MEASUREMENTS OF OF X-RAY FINE STRUCTURE

Rather accurate values of x-ray fine structure have been known for some time; the latest values are taken from the compilation by Cauchois and Hulubei.[12] These may now be compared with the above theory with

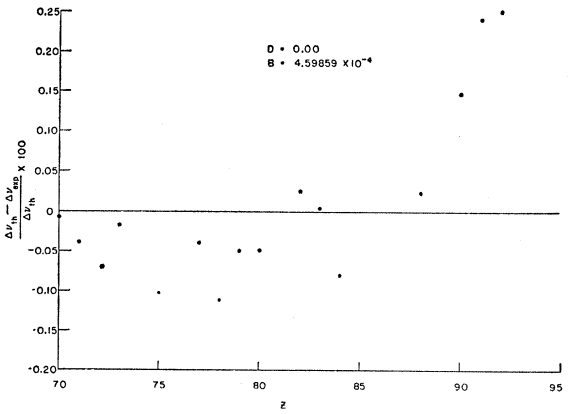

FIG. 4. Comparison of theory [expression (23)] with experimental measurements of $2p$ fine structure assuming a Coulomb field near the nucleus.

[11] H. A. Bethe and C. Longmire, Phys. Rev. **75**, 306 (1949).

[12] Y. Cauchois and H. Hulubei, Table de constantes et donnees numeriques. I. Longueurs d'onde des emissions X et des discontinuities d'absorption X, Paris, 1947).

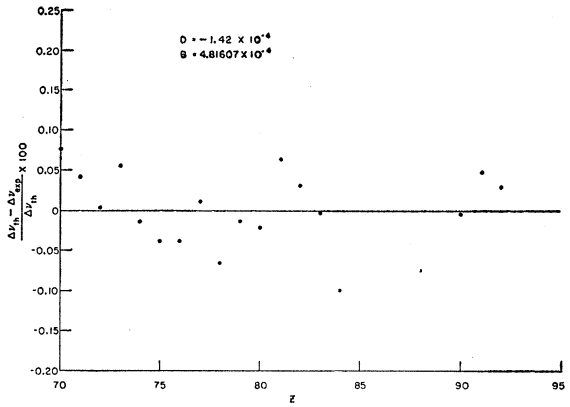

FIG. 5. Comparison of theory [expression (24)] with experimental measurements of $2p$ fine structure allowing for deviations from Coulomb field at the nucleus.

results indicated in Figs. 4 and 5. To a first approximation, $\Delta\nu$ increases as Z^4, so that a curve of $\Delta\nu$ itself is not convenient. The percentage difference between experimental and theoretical values of $\Delta\nu$ is hence shown in Figs. 4 and 5 as a function of Z.

Figure 4 compares experimental results with Christy and Keller's expression (23) after B has been evaluated to give the least (weighted) mean square error. It is clear that there is some systematic deviation from expression (23) which produces a small decrease in $\Delta\nu$. The deviation increases in magnitude very rapidly with Z, so that even with a choice of B to give a (weighted) mean square fit, points for $Z\geq 90$ are in considerable error. It is clear, too, that a change in the value of the fine structure constant will not appreciably improve agreement.

At this point we pause to discuss the type of weight function used in minimizing the weighted mean square error. A given fractional error plotted on Fig. 4 represents an error in frequency or energy which increases as Z^4, since $\Delta\nu$ itself is approximately proportional to Z^4. Hence if there were a fixed error in frequency in all the x-ray measurements, a weight function of Z^4 should be used. On the other hand, a constant fractional error in measurement of $\Delta\nu$ calls for a constant weight function. The actual weight function used was Z^2, so that a given fractional error for $Z=92$ is weighted approximately 1.7 times the same error at $Z=70$. Since the weight function is not greatly different for these two extreme values of Z, details of the choice of a weight function are not critical. Hereafter wherever mean square errors are discussed, it is to be understood that the weight function Z^2 has been used.

If the small term (21) due to finite nuclear size is added to (23), one has

$$\frac{\Delta\nu}{R}=\left[(2/\alpha^2)S(\alpha Z)-2\alpha^2Z^3f(\alpha Z)+BZ^2\right]$$
$$\times[1+De^{0.0837(Z-60)}]. \qquad (24)$$

Here the value $b=0.0837$ has been chosen. This is an average between the two rather similar values for a nucleus of uniform charge, and one with a surface charge only.

Figure 5 shows that (24) gives good agreement with experimental values of $\Delta\nu$, with no noticeable systematic errors. For this figure, the known value $1/\alpha=137.038$ is used while D and B are chosen to minimize the mean square error. If $1/\alpha$ is also regarded as an unknown constant and also chosen to minimize the mean square error the value obtained is 137.04, in remarkable agreement with the accepted value. The constant D obtained in minimizing the errors is -1.42×10^{-4}.

The sign of D is expected to be negative, corresponding to a decrease in the fine structure splitting. For normal ^2p levels, the $^2p_{\frac{1}{2}}$ is lowest in energy. Because a nucleus of finite size does not give as large an attractive potential as a point nucleus, the difference in potential may be regarded as repulsive, which should increase the $^2p_{\frac{1}{2}}$ level for the normal case and bring it closer to the $^2p_{\frac{3}{2}}$. For the x-ray levels involving a missing electron from the $2p$ orbit, the fine structure should also be decreased by this effect.

DISCUSSION OF ERRORS

We shall consider here errors in the determination of D due to random experimental errors, to omission of terms in the Christy and Keller formulation which are dependent on high powers of Z, and to extrapolation or interpolation of Christy and Keller's values for the function $f(\alpha Z)$.

Deviations of the points from a straight line in Fig. 5 are approximately what may be expected from the accuracy to which the experimental points have been determined. It is evident from this figure that the accuracy of individual measured values of D is about 0.05%. From Fig. 4, it may be seen that the systematic deviation in D due to non-Coulomb fields near the nucleus is approximately six times the random errors of points, or 0.3%. Hence the accuracy to which the constant D is known from the present experimental data is about 15%, or possibly better if a probable error from statistical considerations is quoted. In terms of an effective nuclear radius, the 15% error in D corresponds to 10% uncertainty in the radius.

Although the data came from x-ray work of high precision, the measurements were not made with the application of this paper in mind, and hence it is hoped that more precise x-ray measurements for the determination of deviations from a Coulomb field can be made. Extremely careful measurements might even show up variations in effective radii due to shell structure.

There are two evident possible sources of error which come from using the calculations of Christy and Keller as is done above. One is in interpolation or extrapolation of the values in their Table II from which the $f(\alpha Z)$ of expression (24) is obtained. The function $f(\alpha Z)$ was obtained from this table by assuming it to be a power series in αZ or a power series in $1-(1-\alpha^2Z^2)^{\frac{1}{2}}$ using various numbers of terms in the series. The agreement among the different types of expansions indicates rather clearly that errors in interpolation or extrapolation of $f(\alpha Z)$ do not produce errors in D greater than 2 or 3%. The expansion in $1-(1-\alpha^2Z^2)^{\frac{1}{2}}$ appears best, since it converges most rapidly.

Another source of error might be neglected small terms in the Christy and Keller formulation which depend on high powers of Z. The largest of these appear to be involved in the assumption that the function $[S(\alpha Z)/(\alpha Z)^2]f_2(\alpha Z)$ is BZ^2, where B is a constant. The actual function is unknown, but may be expected to be something like the term in s^2 obtained from the expansion of $S(\alpha(Z-S))$, or

$$BZ^2\approx\left(\frac{s}{Z}\right)^2\times\left[\frac{6}{32}(\alpha Z)^4+\frac{75}{256}(\alpha Z)^6+\frac{371}{1024}(\alpha Z)^8+\cdots\right] \quad (25)$$

Here s is the screening constant. Coefficients of powers of αZ in this expression are rather large, being an order of magnitude larger than similar coefficients for the fine structure without screening. The term $(\alpha Z)^6$ in the brackets of (25) actually is proportional to Z^4 because of the $1/Z^2$ in front of the brackets. Hence it should add to the fine structure, which is also approximately proportional to Z^4. It results in a change in the fine structure of about $\Delta\nu/160$, and hence in the fine structure constant α of about $\alpha/640$. This would change $1/\alpha$ from 137.04 in excellent agreement with other values to 136.83 in rather poor agreement. Hence the actual coefficient of the term $(s/Z)^2(\alpha Z)^6$ in BZ^2 must be somewhat less than that indicated by (25). Since the term $(\alpha Z)^8$ in the brackets of (25) would produce some deviation at large Z from the fine structure expression of Christy and Keller, it may be expected to contribute to the constant D. Calculation shows that its contribution to D is 14% of the experimentally observed value. Furthermore, since the value of α indicates that the coefficient of the preceding term in $(\alpha Z)^6$ is considerably less than that given by (25), it may be expected that the coefficient of the term in $(\alpha Z)^8$ and its contribution to D is also less than is given here. Additional higher powers of αZ make contributions to D which are still smaller, and hence may be neglected.

DISCUSSION OF THE MAGNITUDE OF D

The value found for D, -1.42×10^{-4}, is of the correct sign and of approximately the correct magnitude to be expected from the nuclear effect described above. If D is attributed entirely to the finite nuclear size, the result may be expressed in terms of the radius of a fictitious

uniformly charged nucleus as

$$r=(2.1\pm0.2)\times10^{-13}A^{\frac{1}{3}}\text{ cm},\qquad(26)$$

where A is the atomic number. This uses the normalization mentioned above of Wertheim and Igo.[8] More precisely, the result gives $(r^{1.5})_{Av}$ for the nuclear charge distribution as pointed out above. This value of the radius is much too large to be consistent with recent experiments on nuclear radii. These indicate an effective radius near $1.2\times10^{-13}A^{\frac{1}{3}}$ cm, which would give a value of D of only about -0.60×10^{-4}. The discrepancy between -0.60×10^{-4} and the observed value of -1.42×10^{-4} requires an examination and comparison of the various methods for determining nuclear "size." A comparison of this general type has already been given by Hill and Ford.[13] However, certain points are worth restatement and further elaboration here.

Recent experiments on electron scattering, on x-rays from μ mesonium, and the present fine structure anomaly all depend on observing variations from a pure Coulomb field and attributing these variations to the finite size of the nucleus. It must be realized, however, that these three methods of measurement determine different functions of the deviations from a Coulomb field, and hence can be expected to yield different effective radii. This difference is primarily because of the varying wavelengths of the particles involved. The μ meson and the electrons which are scattered have wavelengths of the order of 10^{-12} cm, whereas the wavelength of the $2p$ electron in heavy elements is about 1000 times larger.

As shown above, the present method yields a quantity of the approximate form $(Ze)^{-1}\int\rho_n r^2\rho_e dv$, which is very nearly $(r^{1.5})_{Av}$. The μ-mesonic x-ray measurements determine a quantity which is rather similar but which involves the probability density for the μ meson rather than that for the electron. This assumes that there is no specific interaction between the μ meson and nuclear matter large enough to affect the results. For the $1s$ state, the probability density of the μ meson is greatest at the center of the nucleus, dropping to about $\frac{1}{3}$ of this value at the edge of the nucleus in the case of Pb, and to 1/1.7 for the case of Cu. Thus the μ-mesonium experiment gives a weighted mean square radius with the weight function increasing rather rapidly towards the center of the nucleus. Hence if the charge density is greatest in the center of the nucleus, the μ-meson experiment can be expected to yield effective radii which are somewhat less than those from x-ray fine structure.

Electron scattering can in principle give rather complete information about the nuclear charge distribution. Howerer, the grosser effects and results which are presently available depend most strongly on regions where the potential changes very rapidly. Thus electron scattering determines most readily charge distribution in the central dense region of the nucleus and is relatively insensitive to the region outside this where the charge density may be smaller and decreasing slowly One may hence expect that electron scattering will also give effective radii somewhat less than those obtained from x-ray fine structure.

These considerations strongly suggest that there is some non-Coulomb field extending over a distance considerably larger than the usual nuclear radius which would contribute importantly to the fine structure separation, but not to the other two types of measurement. One such possibility is a long tail on the nuclear charge distribution. Although it appears possible to fit the results of all three types of measurements with such a nuclear model, the "tail" must be so long as to be quite unreasonable. A second and more attractive explanation involves quantum electrodynamic effects which produce deviations from a Coulomb field over a distance considerably larger than the nuclear radius. In fact, the present results seem to give evidence for the existence of a Lamb shift for these levels, and provide a valuable method of measuring such effects when αZ is not small compared with unity.

ELECTRODYNAMIC EFFECTS

Quantum electrodynamic effects may be of some importance in each of the three types of experiments mentioned above. Cooper and Henley show,[3] for example, that vacuum polarization due to the creation of electron pairs affects the nuclear radius determined from the μ-mesonium experiment by about 1%. Christy and Keller[8] give the correction to x-ray fine structure due to vacuum polarization as approximately $(mc^2/160\pi Z)(\alpha Z)^7$. This has the largest fractional effect on the fine structure for large Z, where it is close to 1/5000 of the fine structure splitting. It is not only too small, but also varies too slowly with Z to produce much error in the nuclear size effect discussed above. This expression given by Christy and Keller is accurate, however, only for small values of Z, and examination shows that it is quite misleading for Z in the range 70 to 92.

When αZ is not much smaller than unity, it is very difficult to calculate the effects of vacuum polarization and the Lamb shift. We can only show here that quantum electrodynamic effects are probably large enough to produce a major part of the fine structure anomaly.

If $\alpha Z\ll1$, electrons about the nucleus experience an attractive potential due to vacuum polarization of the form.

$$V_p=\frac{\alpha}{2\pi}\frac{Ze}{r}f(\rho),\qquad(27)$$

where $\rho=2\pi r/\lambda$, and λ is the Compton wavelength $\approx2.4\times10^{-10}$ cm.

$$f(\rho)=\int_0^1\frac{2v^2(1-1/3v^2)}{(1-v^2)}\exp\left[\frac{-2\rho}{(1-v^2)^{\frac{1}{2}}}\right]dv.$$

[13] D. L. Hill and K. W. Ford, Phys. Rev. 94, 1617 (1954).

It may be noted from that the vacuum polarization may be considered to have a range near $\lambda/2\pi$, or 4×10^{-11} cm. This is much larger than the nuclear radius, but considerably smaller than the average radius of a $2p$ electron orbit.

Although the expression (27) for V_p is accurate only for $\alpha Z\ll1$, it was used to obtain a rough magnitude for the effect of vacuum polarization on the $2p_{\frac{1}{2}}$–$2p_{\frac{3}{2}}$ fine structure. The results showed that vacuum polarization gives an effect on the fine structure which varies with Z much like the effect of the finite nuclear radius, and about 40% as large as that expected from a nuclear radius of $1.5\times10^{-13}A^{\frac{1}{3}}$. Since the potential is attractive, however, it is in the wrong direction to account for the anomalously large effect of the "finite nuclear radius" observed above. Wichmann and Kroll[14] have recently succeeded in making a much more complete calculation of vacuum polarization without requiring that $\alpha Z\ll1$. They find an effect on the x-ray fine structure which is not very different from that given here.

Since vacuum polarization gives the equivalent of an attractive potential, its effect is to give too small an effective radius for the nucleus rather than the observed large radius. The Lamb shift, on the other hand, does correspond to a repulsion and hence gives an effect of the correct sign. Its calculation for the case when αZ is not very small is difficult and has not yet been done. When it is accomplished, the observed fine structure anomaly may afford a good method of testing these electrodynamic corrections for large Z.

[14] E. Wichmann and N. M. Kroll, Phys. Rev. (to be published).

PHYSICAL REVIEW VOLUME 100, NUMBER 5 DECEMBER 1, 1955

Radioactive Ca^{47}†

W. S. Lyon and T. H. Handley
Oak Ridge National Laboratory, Oak Ridge, Tennessee
(Received June 20, 1955)

Ca^{47} (4.5 day) has been investigated by using beta-gamma coincidence measurements and NaI gamma-ray spectrometry. Three gamma rays of energies 1.29 Mev, 0.812 Mev, and 0.500 Mev are present with absolute intensities of (71±6)%, (5±0.5)%, and (5±0.5)%, respectively. These gammas are in coincidence with a 0.70±0.02 Mev beta group. In addition, (24±6)% of the decay is through a 1.9±0.2 Mev beta group directly to the ground state.

INTRODUCTION

CALCIUM-47 has been the subject of several recent investigations; the results of these studies, however, indicate considerable uncertainty exists concerning the decay characteristics of this nuclide. Cork *et al.*[1] prepared Ca^{47} by an (n,γ) reaction on enriched (9.6%) Ca^{46}. Marquez[2] prepared Ca^{47} by irradiating $K_2Cr_2O_7$ in the 450-Mev synchrocyclotron at the University of Chicago.

Aten, Grevell, and Van Dijk[3] prepared Ca^{47} by the reaction $Ca^{48}(d,dn)Ca^{47}$ and $Ti^{50}(d,\alpha p)Ca^{47}$ using 26-Mev deuterons. Table I is a summary of data obtained by these workers.

From Table I, it is apparent that some serious discrepancies appear in the beta energies and that the gamma spectrum is in some doubt. Because of the possible medical and biological interest in Ca^{47}, it seemed worthwhile to attempt clarification of the decay scheme.

PREPARATION AND PURIFICATION OF THE SAMPLE

Calcium-47 was prepared in two different ways:

(a) In the 86-inch cyclotron at Oak Ridge National Laboratory (ORNL) by irradiation of CaO with ~14-Mev protons:

$Ca^{48}(p,2p)K^{47}$; $K^{47}\rightarrow Ca^{47}$ and $Ca^{48}(p,pn)Ca^{47}$.

(b) By irradiation with thermal neutrons on enriched (9.6%) Ca^{46} in the low-intensity test reactor at ORNL: $Ca^{46}(n,\gamma)Ca^{47}$.

Gamma ray and decay data obtained from Ca^{47} prepared in each manner were in agreement. Because of the larger amounts of calcium in the proton-irradiated material, 4π beta counting was not tried with this source. Chemical processing of the sample was the same for both methods of preparation and is described below.

The calcium oxide was dissolved in dilute nitric acid and the *p*H adjusted to ~2.0. The daughter Sc^{47} activity, together with other Sc activities was removed by successive extractions with 0.5*M* thenoyltrifluoroacetone (TTA) in xylene.[4]

To the aqueous solution, iron (III) carrier was added

† Work performed under contract to the U. S. Atomic Energy Commission.
[1] Cork, LeBlanc, Brice, and Nester, Phys. Rev. **92**, 367 (1953).
[2] Luis Marquez, Phys. Rev. **92**, 1511 (1953).
[3] Aten, Grevell, and Van Dijk, Physica **19**, 1049 (1953).
[4] W. S. Lyon and B. Kahn, Phys. Rev. **99**, 728 (1955).