

These results are in marked disagreement with those obtained by Krone and Seagondollar,<sup>3</sup> who report a resonance in  $B^{10}(p,\gamma)$  at 0.78 Mev and possible resonances at 0.95 Mev and 1.33 Mev. The procedure by which we obtained the excitation curve (Fig. 5) involved some uncertainties since insufficient data were taken to do it exactly. Therefore, it is possible that we would have overlooked a weak resonance at 0.78 Mev; however, it would certainly be much less intense than our observed resonance at 1.2 Mev. The latter should then have been seen easily by Krone and Seagondollar. It would appear that further work

should be done to resolve the discrepancy between the two sets of data.

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## Coulomb Radius Constant from Nuclear Masses

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On the assumption of a uniform charge distribution, the Coulomb energy constant obtained from a recent adjustment of the semiempirical formula to the mass data corresponds to the Coulomb radius constant 1.237 (in  $10^{-13}$  cm). This result is in good agreement with the radius constant obtained from recent studies of  $\mu$ -mesonic x-rays, and can be readily reconciled with the radii obtained from electron scattering, isotope shift measurements, and mirror nuclide mass differences. In view of the importance of the question of the Coulomb radius, an attempt is made to refine the determination of the Coulomb radius constant based upon nuclear masses by using an objective criterion for best fit. A least-squares analysis involving a new adjustment procedure yields the radius constant 1.216. An investigation of the precision of this determination leads to the assignment of a probable error of 1 percent. This new radius constant agrees with the average radius constant obtained from  $\mu$ -mesonic x-rays within the small probable error assigned to each.

### 1. INTRODUCTION

IN a previous study Green and Engler<sup>1</sup> found an adjustment of constants which brought the Weiszäcker semiempirical equation into good agreement with experimental nuclear masses. It was noted that the energy constants which accomplish this were substantially larger than those appearing earlier in the literature. In view of the lack of understanding of nuclear forces, the significance of the increased nuclear energy constants is obscure. However, the increased Coulomb energy constant ( $a_3$ ) has a simple interpretation if one assumes that the charge in a nucleus may be characterized by a simple charge distribution. In particular, if a uniform charge distribution and the radius formula  $R=r_0A^{\frac{1}{3}}$  are accepted,

$$a_3 = 3e^2/5r_0. \quad (1)$$

Accordingly, the larger Coulomb energy constant (0.750 mMu) corresponds to a smaller Coulomb radius constant<sup>2</sup> (1.2369 in units of  $10^{-13}$  cm). It has been pointed out by Bitter and Feshbach<sup>3</sup> that this new

constant is in good agreement with the radius constant obtained recently from  $\mu$ -mesonic x-ray studies,<sup>4,5</sup> electron scattering studies,<sup>6</sup> and isotope shift studies.<sup>7</sup>

Green and Engler halted their iterative adjustment process when they found a convenient rounded set of energy constants ( $a_1=16.720$ ,  $a_2=18.500$ ,  $a_3=0.750$ , and  $a_4=100$ , all in mMU) which reduced the discrepancy between the Weiszäcker formula and the mass data to the order of magnitude of uncertainty caused by shell effects. In view of the importance of the question of the Coulomb radius, it seems worthwhile to attempt to refine the determination of the Coulomb radius constant based upon nuclear masses by the use of an objective criterion for best fit and to establish the precision of such a determination.

### 2. METHOD OF ADJUSTMENT

The basic assumptions made are that nuclear energies may be expressed in the form

$$E^w = -a_1A + a_2A^{\frac{2}{3}} + a_3(Z^2/A^{\frac{1}{3}}) + a_4(N-Z)^2/4A, \quad (2)$$

<sup>4</sup> V. L. Fitch and J. Rainwater, *Phys. Rev.* **92**, 789 (1953).

<sup>5</sup> A. N. Cooper and E. M. Henley, *Phys. Rev.* **92**, 801 (1953).

<sup>6</sup> Hofstadter, Fechter, and McIntyre, *Phys. Rev.* **92**, 978 (1953).

<sup>7</sup> P. Brix and H. Kopferman, *Festschr. Acad. Wiss. Göttingen* **17**, 49 (1951).

<sup>1</sup> A. Green and N. Engler, *Phys. Rev.* **91**, 40 (1953).

<sup>2</sup> A. Green, Gordon Conference on Nuclear Chemistry, June, 1953 (unpublished).

<sup>3</sup> F. Bitter and H. Feshbach, *Phys. Rev.* **92**, 837 (1953).

and that the constant  $a_3$  is related to the constant radius  $r_0$  by Eq. 1. The usual procedure for evaluating the semiempirical constants<sup>8</sup> is to evaluate  $a_3$  by fitting the mass differences of mirror nuclides, fix  $a_4$  by fitting the experimental line of beta stability, and fix  $a_1$  and  $a_2$  by fitting the masses of beta stable nuclides. It is clear now that the first step in this procedure is in error because of the correlation effects pointed out by Cooper and Henley<sup>5</sup> and because of the fact that the low end of the mass scale is a poor place to adjust a constant of a statistical theory. In essence the alternative method used here is to fix  $a_4$  to  $a_3$  by the constraint imposed by the experimental line of beta stability, and to adjust  $a_1$ ,  $a_2$ , and  $a_3$  by fitting the masses of beta-stable nuclides.

The numerical work was greatly expedited by using the results of Green and Engler as a starting point. Accordingly, the residual between the experimental mass defects,  $\Delta^x$ , for beta-stable nuclides and the best statistical expression  $\Delta_m^b(A)$  for the mass defects of such nuclides is expressed as<sup>9</sup>

$$R = [\Delta^x - \Delta_m^r(A)] - [\Delta_m^0(A) - \Delta_m^r(A)] - [\Delta_m^b(A) - \Delta_m^0(A)], \quad (3)$$

where  $\Delta_m^r(A)$  are the reference mass defects [Eq. (2), reference 1] and  $\Delta_m^0(A)$  are mass defects based upon the Green-Engler constants. The difference  $R^x = \Delta^x - \Delta_m^r(A)$  requires a study of the experimental data, whereas the residuals  $R^b = \Delta_m^b(A) - \Delta_m^0(A)$  and  $R^0 = \Delta_m^0 - \Delta_m^r(A)$  can be handled analytically.

If  $D_m^b(A)$  and  $D_m^0(A)$  are used to denote the functions which characterize the theoretical lines of beta stability, it is not difficult to show that, to a very good approximation,

$$D_m^b - D_m^0 = (\delta\rho/\rho^b)F + (\delta a_4/a_4^b)G, \quad (4)$$

where

$$F = \rho^0 A^{5/3} [1 - (D_m^0/A)] / (1 + \rho^0 A^{\frac{2}{3}}), \quad (5)$$

$$G = A(\Delta_n - \Delta_H) / a_4^0 (1 + \rho^0 A^{\frac{2}{3}}), \quad (6)$$

and  $\rho^0 = a_3^0/a_4^0$ . To relate  $\delta\rho/\rho^b$  and  $\delta a_4/a_4^b$  to  $\delta a_3$ , a particular value of  $A$  ( $A_s$ ) is chosen such that the difference  $D_m^b(A_s) - D_m^0(A_s)$  shall equal a designated quantity  $\gamma_s$ . By using straightforward but tedious algebraic transformations, it is then possible to express the residual  $R$  in the form

$$R = R^x - R^0 + \delta a_1 A - \delta a_2 A^{\frac{2}{3}} - \delta a_3 S(A) - \gamma_s T(A), \quad (7)$$

where

$$S(A) = [(A_s^{5/3} - A_s^{2/3} D_s^0) / 4 D_s^0] [D_m^0 + G - (G_s/F_s) F], \quad (8)$$

<sup>8</sup> J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952).

<sup>9</sup> Henceforth all quantities which refer to the Green-Engler set of constants will be designated by the superscript zero and quantities which relate to the "best" set of constants by the superscript  $b$ . Elsewhere the notation in reference 1 will be followed.

and

$$T(A) = (a_4^b + \Delta_n - \Delta_H) F / F_s - S[(A_s^{5/3} - A_s^{2/3} D_s^0) / (a_3^0 A_s^{2/3} + a_4^0)]. \quad (9)$$

It is convenient now to proceed by setting

$$A_s = 200, \quad \text{and} \quad \gamma_s = 0,$$

thus forcing  $D_m^b$  to intersect the  $D_m^0$  curve at  $A = 200$ . Such an intersection at high  $A$  insures the fact that the departures  $D_m^b - D_m^0$  will be very small ( $\lesssim 0.05$ ) for all values of  $A$ . The curve  $D_m^0$  matches closely the function determined by Fermi, and a number of studies<sup>10,11</sup> indicate that this smooth line of beta stability is quite satisfactory.

At this point the complex question arises as to how to handle the experimental residuals  $R^x$  which are known for about 241 beta-stable nuclides with  $A > 5$ . It is well known that Eq. (2) neglects a number of energy effects which might be characterized as (1) shell structure, (2) pairing effects and pairing anomalies, (3) Coulomb exchange, (4) nonuniform angular distribution of charge, (5) nonuniform radial distribution of charge, (6) compressibility. Also, experiment provides only  $\Delta$  values for integral  $N$  and  $Z$  values, whereas  $\Delta_m(A)$  refers to hypothetical mass values usually located at nonintegral  $N$  and  $Z$  values. Accordingly the departures of the experimental data from Eq. (2) are expected to reflect these neglected energy effects to a greater extent than they reflect the real experimental errors in masses. Nevertheless, for the purpose of the present investigation, it was deemed reasonable to regard the errors due to the inadequacies of Eq. (2) on an equal footing with the real experimental error, and to assume that these errors are normally distributed.<sup>12</sup>

To treat the large mass of experimental data, the mass scale was divided into twenty-four intervals, each spanning 10 units in  $A$  and centered at  $A = 10, 20, 30$ , etc. The centers of gravity of the mass residual values in each interval were then located at the centers of the interval. The twenty-four "normal places" so obtained were treated as basic experimental data with equal weight.<sup>13</sup>

Using these data in Eq. (7), the quantities  $\delta a_1$ ,  $\delta a_2$ , and  $\delta a_3$  were evaluated by least squares<sup>14</sup> under

<sup>10</sup> C. D. Coryell, *Ann. Rev. Nuclear Sci.* **2**, 305 (1953).

<sup>11</sup> A. Green and D. Edwards, *Phys. Rev.* **91**, 46 (1953).

<sup>12</sup> E. R. Cohen, *Revs. Modern Phys.* **25**, 709 (1953). The discussion in this article of the implications of the use of least squares for non-Gaussian distributions is relevant to this present study.

<sup>13</sup> R. A. Birge, *Revs. Modern Phys.* **19**, 298 (1947).

<sup>14</sup> Actually, considerable difficulty was encountered in the calculations because of the cancellations of almost all of the significant figures in the evaluation of various third degree determinants. These difficulties were overcome, however, by using  $A^{\frac{2}{3}} - 0.2A$  as the coefficient of  $\delta a_2$ , and  $S - 6.95A$  as the coefficient of  $\delta a_3$ , and by changing the coefficient of  $A$  accordingly. The advantages of these devices are probably analogous to the advantages associated with the use of orthogonal polynomials in the least-squares fitting of polynomials. (See reference 13.)

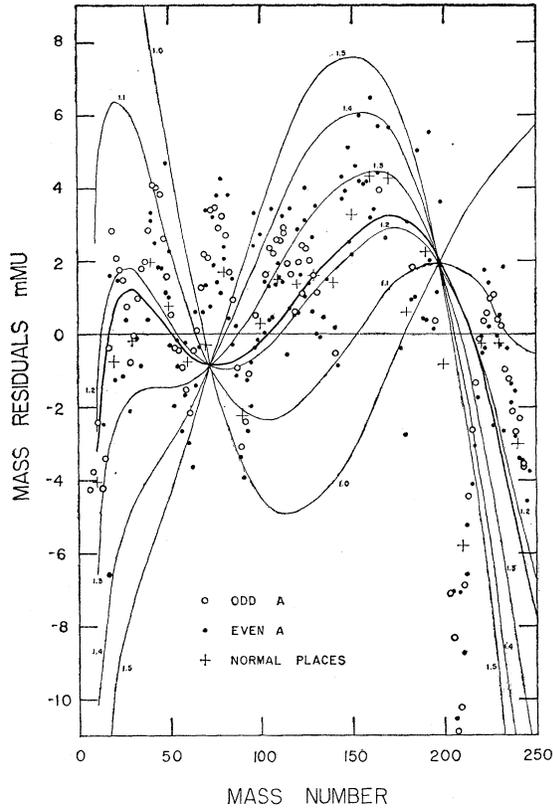


FIG. 1. The mass residuals of beta-stable nuclides in relation to the semiempirical functions obtained by least squares. The heavy curved line corresponds to the "best" fit to the normal places. The light curved lines represent the best fits attainable when the radius constant is fixed at the values indicated. All points are plotted as deviation from the reference values.

the assumption that  $\gamma_s=0$ . The new energy constants and the Coulomb radius obtained were

$$\begin{aligned} a_1 &= 16.9177, & a_2 &= 19.120, & a_4 &= 101.777, \\ a_3 &= 0.76278, & r_0 &= 1.2162. \end{aligned} \quad (10)$$

To determine the effect upon the radius constant of the assumption that  $\gamma_s=0$ , a least-squares calculation based upon Eq. (7) with nonvanishing values of  $\gamma_s$  was used. The Coulomb energy constant change and the corresponding fractional Coulomb radius change turned out to be

$$\delta a_3 = 0.00797\gamma_s, \text{ and } (\delta r_0/r_0) = -0.0103\gamma_s. \quad (11)$$

A detailed study of the line of beta stability indicates that only small changes in  $\gamma_s$  ( $\sim \pm 0.2$ ) can be tolerated if one retains the functional form obtained from the Weiszäcker mass formula. Even the extreme assumption  $\gamma_s = \pm 1$  leads to a shift in  $r_0$  of only 1 percent. Since the effect of possible departures of  $\gamma_s$  upon  $a_1$  and  $a_2$  is also small, for the purposes here  $\gamma_s$  may be set to zero.

By using the new set of constants here obtained, the new residuals  $\Delta^x - \Delta_m^b$  were computed for the twenty-

four normal places. The standard deviation of these points was found to be

$$\sigma = 1.98 \text{ mMU}. \quad (12)$$

In Fig. 1 we show the relationship of this new semiempirical curve (heavy line) to the experimental data,<sup>15</sup> as viewed from a reference line based upon the empirical equation,

$$\Delta_m^r(A) = 0.01(A - 100)^2 - 64. \quad (13)$$

Also shown are the twenty-four normal places.

For assigning an uncertainty to  $r_0$ , the usual procedure used for evaluating the standard deviation of a constant which has been fixed by least squares was employed. On the basis of the assumption that all the input data have the same absolute error ( $e$ ) it was found that the error in  $a_3$  is

$$\epsilon(a_3) = 0.0295e/n^{1/2}, \quad (14)$$

where  $n$  is the number of observations. Assigning to  $e$  the standard deviation in mass defects for  $r_0 = 1.216$ , the standard deviation in  $a_3$  is found to be  $\sigma(a_3) = 0.0119$  mMU. With  $a_3 = 0.7628$  and  $p = 0.674\sigma$ , the percentage probable error in  $r_0$  is  $p = (100)(0.674)(0.0119)/0.7628 = 1.03$ .

To obtain a more intuitive picture of the sharpness of a Coulomb radius determination based upon nuclear masses the following calculations were carried out. The Coulomb radius constant  $r_0$  was fixed at 1.0, 1.1, 1.2, 1.3, 1.4, and 1.5. The corresponding values of  $\delta a_3$  in Eq. (7) were used with  $\gamma_s = 0$ , and  $\delta a_1$  and  $\delta a_2$  were determined by least squares. The values of the corresponding energy constants are given in Table I. The mass defect functions,  $\Delta_m^{r_0}(A)$  for the various values of  $r_0$  may be placed in the form

$$\Delta_m^{r_0} = \Delta_m^b(A) - \delta' a_3 U(A), \quad (15)$$

TABLE I. Sets of semiempirical constants.

$r_0$	$a_1$	$a_2$	$a_3$	$a_4$	$\sigma$
1.0	19.2675	26.259	0.9277	124.70	6.50
1.1	18.0659	22.607	0.8433	112.97	3.59
1.2	17.0647	19.565	0.7731	103.21	2.01
1.3	16.2174	16.990	0.7136	94.94	2.71
1.4	15.4912	14.784	0.6626	87.86	4.24
1.5	14.8617	12.871	0.6184	81.71	5.74
1.216 <sup>a</sup>	16.9177	19.120	0.7628	101.78	1.98
1.237 <sup>b</sup>	16.7200	18.500	0.7500	100	2.12
1.480 <sup>c</sup>	15.04	14.0	0.627	83	8.11

<sup>a</sup> Best fit.

<sup>b</sup> Green-Engler constants.

<sup>c</sup> Fermi constants with  $\Delta_n = 8.930$ ,  $\Delta_H = 8.123$ .

<sup>15</sup> In addition to the mass data used in reference 1, recently determined masses by Collins, Johnson, and Nier and by Hogg and Duckworth have been included in this study. The writer wishes to express his appreciation to Professor A. O. Nier and Professor H. E. Duckworth for transmitting these data before publication.

where  $\delta'a_3$  is the deviation of  $a_3$  from the best value and

$$U(A) = S(A) + 43.294A^{\frac{1}{2}} - 14.2477A. \quad (16)$$

The function  $U(A)$  vanishes at  $A = 72.6$  and  $A = 197.0$ , and hence the  $\Delta_m^{r_0}(A)$  curves all intersect  $\Delta_m^b(A)$  at these two points. The light curves in Fig. 1 represent these functions. It is clear that  $r_0 = 1.20$  and  $r_0 = 1.216$  are far superior to the remaining curves. It may also be apparent that  $r_0 = 1.216$  is somewhat superior to  $r_0 = 1.200$  when viewed in relation to the normal places. This is an indication of the sensitivity of this method of determining the radius constant. One might use Fig. 1 to estimate variations in the nuclear radius constant. However, such a refinement in itself would appear unwarranted unless consideration were given to the other small effects which have been neglected. Such a study is underway.

As a further indication of the sensitivity of nuclear masses to the assumed radius constant, the standard deviations of the normal places from the predicted masses for the different cases are given in column 6 of Table I.

### 3. CONCLUSION

Since this analysis has been carried through exclusively with small residuals, it is important to point out that the fundamental physical quantity of interest here, the nuclear energy, ranges from 0 mMU to about -2000 mMU. The Coulomb energy itself ranges from

0 to about 1200 mMU. The fact that Eq. (2) with the four adjusted constants obtained here (one of which should be credited with the prediction of the line of beta stability) is capable of predicting these energies or the corresponding mass defects with a standard deviation of only 2 mMU,<sup>16</sup> would appear to indicate that the equation has considerable validity. It is satisfactory that the radius constant associated with this "best" fit agrees with the average radius constant<sup>17</sup> obtained from  $\mu$ -mesonic x-ray studies to within the limits of the small errors ( $\sim 1$  percent) assigned to each. It is also satisfactory that this small Coulomb radius constant may be reconciled<sup>3</sup> with the results of electron scattering and isotope shift studies, as well as the mass differences of mirror nuclides.<sup>5</sup>

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<sup>16</sup> This deviation may be reduced by more than a factor of two by introducing a shell correction of the type proposed in reference 11.

<sup>17</sup> V. L. Fitch and J. Rainwater (reference 4) quote the radius constants 1.17, 1.21, 1.22, 1.17 for  $Z = 22, 29, 51,$  and  $82$ . The average of these is 1.198. The accuracy of their energy measurements is about 1 percent.

## Relative Photofission Yields of Several Fissionable Materials\*

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Relative photofission yields of  $U^{238}$ ,  $U^{236}$ ,  $U^{235}$ ,  $U^{234}$ ,  $U^{233}$ ,  $Th^{232}$ ,  $Np^{237}$ , and  $Pu^{239}$  were measured at three betatron energies with a photomultiplier fission fragment detector. Results at betatron energies of 17 and 20 Mev are in agreement and give the following relative fission values:  $Th^{232}$ , 0.31;  $U^{238}$ , 1.00;  $U^{236}$ , 1.43;  $U^{235}$ , 2.40;  $U^{234}$ , 1.82;  $U^{233}$ , 2.54;  $Np^{237}$ , 2.40;  $Pu^{239}$ , 3.17. The relative  $(\gamma, f)$  yields are empirically correlated with the nuclear parameter  $Z^2/A$ . Some comments are made on the competition between fission and neutron emission.

### INTRODUCTION

McELHINNEY and Ogle<sup>1</sup> measured the relative photofission yields of several fissionable materials with respect to  $U^{238}$  by a "catcher" method. They obtained the following results:  $U^{238}$ , 1.00;  $U^{236}$ , 2.49;

$U^{235}$ , 1.49;  $Pu^{239}$ , 2.51;  $Th^{232}$ , 0.257; and  $Th^{230}$ , 0.847. Owing to the importance of such measurements in the theoretical interpretation of the photofission process, we have remeasured the relative  $(\gamma, f)$  yields of the above materials by a different method and have extended the measurements to several other materials. The relative photofission yields were measured at three betatron energies with a photomultiplier fission fragment detector.<sup>2</sup>

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<sup>1</sup> J. McElhinney and W. E. Ogle, *Phys. Rev.* **81**, 342 (1951).

<sup>2</sup> James Gindler, University of Illinois thesis, 1954 (unpublished).