Theory of Nuclear Coulomb Energy

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The characteristic oscillatory behavior of the Coulomb energy differences for light nuclei is explained in terms of the different values of the average Coulomb interaction between two particles when the wave function is, respectively, symmetrical and antisymmetrical in the space coordinates of the particles. The possible effect of the low binding energy of the least strongly bound particle in 4n+1 nuclei is discussed. It is shown how the Coulomb energy differences obtained empirically from odd nuclei may be used to estimate Coulomb energy differences for various other cases.

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INTRODUCTION

`HE Coulomb energy of nuclear systems has been estimated by several distinct methods. These are described briefly in the following paragraphs.

(a) For light nuclei $(A \leq 16)$, single particle wave functions are combined to form Hartree type total wave functions with definite angular momentum and symmetry quantum numbers. These wave functions are used to calculate the expectation value of the electrostatic interaction. Although this procedure does not yield precise quantitative results, it reveals a characteristic oscillatory behavior of the Coulomb energy as a function of atomic number in qualitative agreement with the observations.¹ In the special case of three particle systems (He³ and H³), more refined calculations based on models of nuclear forces correlate the Coulomb energy with the assumed range of the force in a satisfactory manner.²

(b) The classical energy of a continuous charge distribution spread uniformly throughout a sphere of radius R has the value $3Z^2e^2/5R$, if Ze is the total charge. To obtain the nuclear Coulomb

energy, Z^2 is replaced by Z(Z-1) with the result

$$E_c = 3Z(Z-1)e^2/5R.$$
 (1)

The assumption of constant nuclear density with

$$R = 1.47 \times 10^{-13} A^{\frac{1}{3}} \text{ cm}$$
 (2)

$$E_c = 0.592Z(Z-1)/A^{\frac{1}{3}}$$
 Mev. (3)

For odd isobaric nuclei with $N-Z=\pm 1$ the Coulomb energy difference is simply

$$\Delta E_c = E_c(-1) - E_c(+1) = 0.592(A-1)/A^{\frac{1}{2}} \text{ Mev}$$
(4)

in excellent agreement with the experimental determinations for A greater than 15, excepting one outstanding discrepancy at A = 27. Below A = 16there are numerous discrepancies, principally because of the oscillatory behaviour of the experimental mass differences.

(c) Attempts have been made to correlate the Coulomb energy differences of odd nuclei with the binding energy of the least strongly bound particle.^{5,6} Consider, for example, the systems $_{4}Be^{9}$ and $_{5}B^{9}$. The odd particle (neutron or proton) is lightly bound and one might, therefore, expect the effective nuclear radius to exceed the value given by Eq. (2) and the Coulomb energy difference to fall below the value ΔE_c defined by Eq. (4). The qualitative correlation is in the right direction, but no quantitative formulation free from arbitrary assumptions has yet been devised. In view of the extreme nature of these assump-

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This work was carried out while one of us (G. G.) held a fellowship from the Radio Receptor Company, Inc., of

New York. ¹E. Feenberg and M. Phillips, Phys. Rev. **51**, 597 (1937). M. Phillips and E. Feenberg, Phys. Rev. **59**, 400 (1941).

²S. S. Share, Phys. Rev. 50, 488 (1936). William Rarita and R. D. Present, Phys. Rev. 51, 788 (1937). The application of the calculations in references 1 and 2 to the experimental material involves the assumption that the neutron-neutron and proton-proton interactions differ only in the electrostatic interaction between protons. The same assumption is made in the present analysis of the Coulomb energy problem.

 ³ E. Wigner, Phys. Rev. 51, 947 (1937).
 ⁴ W. Barkas, Phys. Rev. 55, 694 (1939).
 ⁵ D. R. Elliott and L. D. P. King, Phys. Rev. 60, 489 (1941). ⁶ H. A. Bethe, Phys. Rev. 54, 436 (1938).

tions, it seems reasonable to conclude that some other effect must be invoked to account for the major part of the experimental periodicity.

(d) The Coulomb energy is an important factor in studies of nuclear structure based on the alpha-particle model.⁷ However, it is not likely that estimates of the Coulomb energy can supply evidence for or against the spatial localization suggested by the model. It is satisfactory that a simple estimate agrees fairly well with the experimental determinations.

The present calculation avoids the approximations made in (a) and (b) and yields the characteristic oscillations found in (a) while going over into (b) for sufficiently large values of A. Because of the effect described in (c), certain relations must be interpreted as inequalities for Aless than 13. The method of calculation is adapted from that used by Wigner in his theory of nuclear mass defects.³

GENERAL FORMULATION

The operator⁸

$$X = \frac{1}{4} (1 - \tau_{\zeta 1}) (1 - \tau_{\zeta 2}) P_{12} \tag{5}$$

has the eigenvalues 1, -1, and 0, the first two occurring only when both particles are protons. From the expectation values of the operators

$$X_{s} = \frac{1}{2}(P_{12}+1)X,$$

$$X_{a} = \frac{1}{2}(P_{12}-1)X$$
(6)

one obtains the probabilities

$$p_{s} = (\psi, X_{s}\psi) = (X_{s}\psi, X_{s}\psi),$$

$$p_{a} = (\psi, X_{a}\psi) = (X_{a}\psi, X_{a}\psi)$$
(7)

for the eigenvalues 1 and -1, respectively, of X. Summing Eq. (7) over all pairs of particles yields the result

$$\frac{1}{2}A(A-1)(p_s+p_a) = \frac{1}{2}Z(Z-1).$$
 (8)

Thus, $\frac{1}{2}A(A-1)p_s$ may be interpreted loosely as

the number of proton pairs for which the wave function is symmetric in the space coordinates of the two protons.

The Coulomb energy, given by the equation

$$E_{c} = \frac{1}{8} \sum' \left(\psi, (1 - \tau_{\varsigma i})(1 - \tau_{\varsigma j}) \frac{e^{2}}{r_{ij}} \psi \right), \quad (9)$$

is readily expressed in the form

$$E_{c} = \frac{1}{2}A(A-1) \left[\left(X_{s}\psi, \frac{e^{2}}{r_{12}}X_{s}\psi \right) + \left(X_{a}\psi, \frac{e^{2}}{r_{12}}X_{a}\psi \right) \right]. \quad (10)$$

The quantities L_s and L_a , defined by the equations

$$p_{s}L_{s} = \left(X_{s}\psi, \frac{e^{2}}{r_{12}}X_{s}\psi\right),$$

$$p_{a}L_{a} = \left(X_{a}\psi, \frac{e^{2}}{r_{12}}X_{a}\psi\right),$$
(11)

are interpreted as average values of the Coulomb interaction between two particles when the wave function is, respectively, symmetrical and antisymmetrical in the space coordinates of the particles. In the applications an approximation is introduced by the assumption that L_s and L_a are constant in an isobaric series and vary smoothly as functions of A. A possible limitation on the validity of this assumption is discussed in the concluding section.

It is convenient to replace L_s and L_a by the linear combinations

$$L_c = \frac{1}{4}(L_s + 3L_a), \quad L'_c = L_s - L_a.$$
 (12)

Then, using Eqs. (8), (11), and (12), Eq. (10) becomes

$$E_{c} = \frac{1}{2}Z(Z-1)L_{c} + \frac{1}{8}A(A-1)(3p_{s}-p_{a})L'_{c}.$$
 (13)

The assumption that the nuclear volume is proportional to the number of particles, as in Eq. (2), implies that $A^{\frac{1}{2}}L_{c}$ approaches a constant limiting value as A grows larger. For L'_{c} one expects a very rapid decrease with increasing A. In agreement with this expectation, the empirical data require that L_{s} and L_{a} approach equality beyond A = 20.

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⁷ L. R. Hafstad and E. Teller, Phys. Rev. **54**, 681 (1938). Harrison Brown and D. R. Inglis, Phys. Rev. **55**, 1182 (1939).

⁽¹⁾ ${}^{8}\tau_{1}$ has the value 1 if the particle is a neutron and the value -1 for a proton. The operator P_{12} interchanges the space coordinates of particles 1 and 2. We use a completely antisymmetric wave function (in the space, spin, and charge coordinates of the particles) to obviate the need to distinguish between matrix elements of operators with subscripts 1 and 2 and any other pair of subscripts.

To determine p_s and p_a , some specification of the symmetry properties of the wave function is required. Wigner's first approximation,³ which yields wave functions belonging to irreducible representations of the symmetric group, is assumed in this discussion.

EVALUATION OF p. AND p.

Two procedures will be described. The first may be called intuitive, since it interprets the symmetry properties of the wave function in terms of an alpha-particle structure without, however, specifying the spatial location of the alpha-particles (more properly the "four" groups). The second is a formal and rigorous operator calculation.

1. For a single alpha-particle, there is only one interaction term and the Coulomb energy is represented by $E_c = L_s$. The same formal result is obtained for the systems He⁵ and He⁶ in their normal states. If a proton is added to the alpha-particle, the spin wave function of the three protons has the form

$$2^{-\frac{1}{2}} \{S_{+}(1)S_{-}(2) - S_{+}(2)S_{-}(1)\} S_{+}(3). \quad (14)$$

It is easily seen that the spin state associated with particles 2 and 3 is a linear combination of singlet and triplet with statistical weights one and three, respectively. Consequently, the addition of the proton increases the Coulomb energy by the amount

$$\frac{1}{2}(L_s+3L_a).$$
 (15)

The Coulomb interaction energy between two "four" groups is then evidently L_s+3L_a .

In the A = 4k series with A = 2Z there are Z/2"four" groups and Z(Z-2)/8 pairs of "four" groups. The Coulomb energy has the value

$$E_{c} = \frac{1}{2}ZL_{s} + Z(Z-2)(L_{s}+3L_{a})/8$$

= $\frac{1}{2}Z(Z-1)L_{c} + 3ZL'_{c}/8.$ (16)

The same result holds for all nuclei containing an even number of protons. In the A = 4k+1 series with A = 2Z-1 there are $\frac{1}{2}(Z-1)$ "four" groups, (Z-1)(Z-3)/8 pairs of "four" groups, and one additional proton. One obtains

$$E_{c} = \frac{1}{2}(Z-1)L_{s} + (Z-1)(Z-3)(L_{s}+3L_{a})/8$$

+ $\frac{1}{4}(Z-1)(L_{s}+3L_{a})$ (17)
= $\frac{1}{2}Z(Z-1)L_{c} + 3(Z-1)L'_{c}/8$

for these nuclei and for all others containing an odd number of protons.

This simple procedure yields the Coulomb energy without explicit reference to p_s and p_a . These quantities may be inferred from Eqs. (8), (13), (16), and (17). For systems containing an even number of protons,

$$A(A-1)p_s = \frac{1}{4}Z(Z+2);$$

$$A(A-1)p_a = \frac{3}{4}Z(Z-2).$$
(18)

For an odd number of protons

$$A(A-1)p_s = \frac{1}{4}(Z-1)(Z+3),$$

$$A(A-1)p_a = \frac{3}{4}(Z-1)^2.$$
(19)

Equations (16), (17), (18), and (19) are valid only for states with the same symmetry character (the same (P P' P'')) as the normal state.

2. The formal and rigorous derivation starts from the relation

$$P_{ij} = -\frac{1}{4} (1 + \boldsymbol{\sigma}_i \cdot \boldsymbol{\sigma}_j) (1 + \boldsymbol{\tau}_i \cdot \boldsymbol{\tau}_j)$$
(20)

which equates the space exchange operator to the negative product of spin and charge spin exchange operators. We wish to evaluate the diagonal matrix elements of the operator

$$W = \frac{1}{2} \sum_{ij'} \frac{1}{4} (1 - \tau_{\zeta i}) (1 - \tau_{\zeta j}) P_{ij}$$

$$= \frac{1}{32} \sum_{ij'} (1 + \boldsymbol{\sigma}_i \cdot \boldsymbol{\sigma}_j) (1 + \boldsymbol{\tau}_i \cdot \boldsymbol{\tau}_j)$$

$$\times (1 - \tau_{\zeta i}) (1 - \tau_{\zeta j}).$$
(21)

It is evident from Eqs. (5)-(7) that

$$\frac{1}{2}A(A-1)(p_s - p_a) = (\psi, W\psi).$$
(22)

With the help of Eq. (8), p_s and p_a can be expressed in terms of the diagonal matrix elements of W.

The triad of relations

$$\tau_{\xi}\tau_{\eta} = -i\tau_{\zeta}; \quad \tau_{\eta}\tau_{\zeta} = -i\tau_{\xi}; \quad \tau_{\zeta}\tau_{\xi} = -i\tau_{\eta}$$

permits the reduction of an arbitrary power series in the variables τ_{ξ} , τ_{η} , τ_{ζ} to a linear function of the same variables. In particular

$$(1 + \tau_i \cdot \tau_j)(1 - \tau_{\xi i})(1 - \tau_{\xi j})$$

= 2(1 - \tau_{\xi i})(1 - \tau_{\xi j}) (23)

and Eq. (21), defining W, may be replaced by

$$-W = \frac{1}{16} \sum_{ij} (1 + \boldsymbol{\sigma}_i \cdot \boldsymbol{\sigma}_j) (1 - \tau_{\zeta i}) (1 - \tau_{\zeta j}). \quad (24)$$

The right-hand member of Eq. (24) can be expressed in terms of known quantities by the expedient of adding and subtracting a diagonal term to the sum over i and j:

$$-W = \frac{1}{16} \left\{ \sum_{i} (1 - \tau_{\xi i}) \right\}^{2} + \frac{1}{16} \left\{ \sum_{i} \sigma_{i} (1 - \tau_{\xi i}) \right\}^{2} - \frac{1}{2} \sum_{i} (1 - \tau_{\xi i}) \quad (25) = \frac{1}{4} Z^{2} - Z + S_{\pi}^{2}.$$

Here, S_{π}^{2} is the total proton spin operator. We are concerned with states which are eigenstates of $S_{\pi}^{2} = S'(S'+1)$ with the eigenvalues 0 (Z even) and $\frac{1}{2}(1+\frac{1}{2})$ (Z odd). These are the states with maximum symmetry in the space coordinates of the particles. Equations (18) and (19) are now immediate consequences of Eqs. (8), (22), and (25) combined with the above eigenvalues of S_{π}^{2} . In reality the total proton spin does not commute with the Hamiltonian operator and the reference to eigenvalues is inappropriate. However the diagonal matrix elements of S_{π}^{2} cannot differ appreciably from the quoted eigenvalues.

COULOMB ENERGY DIFFERENCES

The formulae for E_c can be put in the following form :

$$E_{c} = \frac{1}{2}Z(Z-1)L_{c} + \frac{3}{8}[Z-\frac{1}{2}+\frac{1}{2}(-)^{Z}]L'_{c}.$$
 (26)

Now, setting $Z = \frac{1}{2}(A \pm u)$,

$$E_{c}(A, \pm u) = \frac{1}{8}(A \pm u)(A \pm u - 2)L_{c} + \frac{3}{16}[A \pm u - 1 + (-1)^{(A \pm u)/2}]L'_{c}.$$
 (27)

Then,

$$\Delta E_{c}(A, u) = E_{c}(A, u) - E_{c}(A, -u)$$

$$= \frac{1}{2}u(A-1)L_{c} \qquad (28)$$

$$+ \frac{3}{8} \left[u + \frac{1}{2}(-1)^{(A+u)/2} \{1 - (-1)^{u} \} \right] L'_{c}.$$

Using Eq. (28), Table I is easily constructed:

A	N-Z	$\Delta E_{\bullet}(A, N-Z)$			
$ \frac{4k+1}{4k+3} \\ 2k \\ 4k+1 \\ 4k+3 \\ 4k+3 $	1 1 2n 3 3	$\frac{\frac{1}{2}(A-1)L_{e}=F}{\frac{1}{2}(A-1)L_{e}+\frac{3}{4}L'_{e}=G}$ n(F+G); n=1, 2, 3, F+2G 2F+G			





FIG. 1. Plot of experimental Coulomb energy differences against mass number. Ordinates are in Mev and have been multiplied by the factor 2/(A-1).

We also include the relations between F, G and L_s , L_a :

$$L_s = G - \frac{A-3}{A-1}F, \quad L_a = -\frac{1}{3}G + \frac{1}{3}\frac{A+5}{A-1}F. \quad (29)$$

Evidently, for |N-Z| = 1, the alternation of the coefficient of L'_{e} as one proceeds along the series of odd nuclei causes an oscillatory behavior of the Coulomb energy differences. That is, one expects the data for odd nuclei with |N-Z| = 1to fall on two separate curves, marked F and G in the table, rather than on a single curve.

Now, if F and G were smoothly varying functions of the mass number, it would be possible to use the experimental data for the 4n+3 nuclei to determine G for all A and similarly to use the data for the 4n+1 nuclei to determine F. With these results and also Table I, Coulomb energy differences could be predicted for many nuclei.

However, F and G may possibly change irregularly with A for the following reason, already mentioned in the introduction under (b). It is likely that the nuclear radius is not a smoothly varying function of mass number below A = 12, since the binding energy of the least strongly bound particle in this range does not vary smoothly with A. The nuclei of the type 4k+1might be expected to have larger radii than would be predicted from the radii of nuclei with neighboring mass numbers. Now it seems likely



FIG. 2. Plot of L_s and L_a against mass number. Ordinates are in Mev. The dotted curves correspond to the following change in Fig. 1: The A = 4n+1 curve on Fig. 1 is held constant and equal to 0.497 for A less than 13.

that $L_{s}R$ and $L_{a}R$, where R is the effective nuclear radius, change in a fairly regular manner with mass number, even though possibly L_s and L_a do not. It follows that one expects FR and GR to vary more smoothly with mass number than Fand G. This means that the experimentally determined F curve, for A less than 13, applies rigorously only to the 4k+1 nuclei, while the interpolated points for other nuclei may be too low. Similarly, the G curve is correct for all but the 4k+1 nuclei, for which it may be too high.

The experimentally determined curves of 2F/(A-1) and 2G/(A-1) (not corrected for the above possibility) are given in Fig. 1. The experimental points are also indicated there. The derived curves, L_s and L_a , are plotted in Fig. 2. Table II lists the experimental data and the ordinates of the various curves. The Coulomb energy difference computed from Eq. (4) is also included.

Summarizing the preceding discussion, the Fcurve of Fig. 1 is to be interpreted, for all but the 4k+1 nuclei, as a lower limit to the correct value to be used in Table I. Similarly, the G curve is, for the 4k+1 nuclei, an upper limit. The curves for L_a and L_s are lower and upper limits to the correct values for all mass numbers. Above mass 12, all statements concerning upper and lower

limits may be replaced by statements concerning equalities.

In order to show the effect of a change in F on the derived curves of Fig. 2, the dotted curves of 2 were plotted. These correspond to the assumption that 2F/(A-1) is constant and equal to 0.497 for A less than 13.

It will be possible to determine more accurately the values of F for other than 4k+1 nuclei when sufficient data are obtained for the Coulomb energy differences of even nuclei. Or, if some estimate is obtained of the variation of nuclear radius with mass number which is sufficiently accurate in this region, the experimental data can be used to determine FR and GR for all A. One then will be in a position to determine more nearly correct values of F and G for all A.

A few words should be said about the experimental values as plotted. Because of the uncertainties of the data, the curves of Fig. 1 are somewhat arbitrary. However, it seems quite clear that the 4k+1 nuclei fall on a separate curve from the 4k+3 nuclei. The apparently

TABLE II. Experimental Coulomb energy differences and ordinates of curves plotted in Figs. 1 and 2.

American do T	ΔE_{e} F	Refer-	$2\Delta E_c$	2G	2 <i>F</i>	,		
A	(Mev)	ences	A - 1	A-1	A -1	L.	La	1.184 <i>A</i> -
3	0.74	a	0.74	0.74	0.37	0.74		0.821
5	0.80	b	0.40	0.67	0.40	0.94	0.22	0.692
7	1.62	с	0.54	0.62	0.43	0.98	0.25	0.619
9	1.84	с	0.46	0.58	0.46	0.93	0.30	0.569
11	2.73	с	0.546	0.55	0.48	0.80	0.38	0.532
13	2.98	с	0.497	0.52	0.50	0.65	0.45	0.504
15	3.5	d	0.50	0.50	0.49	0.55	0.47	0.480
17	3.88	е	0.485	0.48	0.47	0.49	0.47	0.461
19	3.98	f	0.442	0.45	0.45	0.45	0.45	0.444
21	4.32	g	0.432	0.43	0.43	0.43	0.43	0.429
23	4.60	f	0.418	0.42	0.42	0.42	0.43	0.416
25	4.77	f	0.398	0.41	0.41	0.41	0.41	0.405
27	5.32	h	0.408	0.40	0.40	0.40	0.40	0.395
29	5.41	g	0.386	0.39	0.39	0.39	0.39	0.385
31	5.65	i, g	0.377	0.38	0.38	0.38	0.38	0.377

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ⁱ See reference 5.

anomalous experimental value for A = 7 is not yet sufficiently well established to justify an attempt at explanation or interpretation.

It is, of course, conceivable that the entire experimental difference G-F is attributable to the variations of R alone, rather than to a

combination of causes, including the difference in L_{s} and L_{a} . However, there seems to be no plausible excuse for assuming L_s and L_a to be equal. What is surprising is that the experimental evidence requires them to approach equality for relatively low mass number $(A \sim 20)$.

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Elastic Backscattering of d-d Neutrons

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The backscattering of d-d neutrons was investigated for several materials. A directional thick paraffin detector was used. The detector was sensitive primarily to neutrons which had been scattered elastically or with little energy loss.

INTRODUCTION

*****HE purpose of the present experiments was to measure the cross sections of various materials for the backward scattering of d-dneutrons which had suffered elastic collisions or inelastic collisions with small energy loss. For this purpose a detector was used, the sensitivity of which is a rapidly increasing function of neutron energy. The detector used the recoils originating from a thick layer of paraffin. Such a detector has a high efficiency, can be made directional, and is most sensitive to the neutrons of highest energy. The directional property of recoil protons eliminates the necessity for a shadow cone usually required to keep the large direct beam from completely masking the small fraction of reflected neutrons.

SOURCE

The neutrons were obtained from the d-d reaction by bombarding a thick D₂O ice target with 50-100 µa of 200-kev unanalyzed ions of deuterium accelerated by means of a Cockcroft-Walton set. The target was cooled with a liquid oxygen refluxing system which required only a small amount of material near the target.

The neutron flux was monitored by counting the protons from the companion $D(d, p)H^3$ reaction.

The deuteron beam was collimated by two tungsten diaphragms with $\frac{3}{8}$ diameter apertures to define the source on the target. For the conditions of the experiment, the neutron spectrum extends from 2.5 to 3.1 Mev with an energy spread of 0.3 Mev at half-maximum.

The angular distribution of the neutrons from this source for the conditions of the experiment



FIG. 1. Spherical ionization chamber. Recoil protons from a thick layer of paraffin are detected.

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