PHYSICAL REVIEW · VOLUME 109, NUMBER 1 JANUARY 1, 1958

Superheavy Nuclei

FREDERICK G. WERNER AND JOHN A. WHEELER
Palmer Physical Laboratory, Princeton University, Princeton, New Jersey
(Received August 19, 1957)

The semiempirical mass formula and other data previously extrapolated by one of us indicated possible existence of nuclei with mass values up to twice the largest now known. Here this mass region is further explored. Extrapolations are revised and the properties of such superheavy nuclei are estimated in more detail. Despite Z values substantially higher than 137, the K electrons behave perfectly normally because of the finite extension of the nucleus. Vacuum polarization and vacuum fluctuations are roughly estimated to make relatively minor alterations in the K electron binding—which exceeds mc^2. The effect of nuclear attraction in speeding up beta decay is calculated approximately. Calculated beta lives are never much less than 10^{-4} sec. Beta decay energies and neutron binding energies are calculated from the semiempirical mass formula. Fission barriers and cross sections for the (n,γ) process are estimated. Branching ratios in beta decay are calculated for the processes of simple beta decay and for "delayed" neutron emission and "delayed" fission. The latter quantity sets an irreducible minimum to the losses that occur in the process of buildup under even the heaviest neutron flux. The calculated fractional yield of nuclei which reach $Z=147$, $A=500$ is >0.05. Under a lower flux the losses are greater. *All stability calculations in this paper depend upon substantial extrapolations, with complete disregard of shell effects and other particularities that may be important, and therefore can be completely in error.* Conversely, observation of existence or absence of superheavy nuclei with lives less than a second should test stringently the semiempirical mass formula and the semiempirical estimates of spontaneous fission barriers.

I. INTRODUCTION

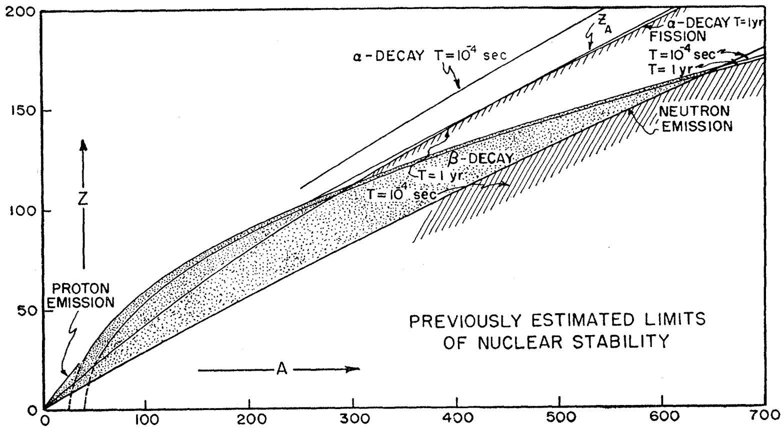
As of January 1953 there were known 383 beta-stable—or nearly beta-stable—nuclei; but stable and beta-active nuclei together totaled 1134. In the meantime additional nuclei have been found[1] with charges up to $Z=101$ and mass numbers up to $A=256$. How much farther can one go? Recently one of us analyzed the stability of still heavier nuclei with respect to spontaneous fission, alpha and beta stability, and neutron escape.[2] The analysis was based on three assumptions, any one of which may be wrong: (1) The semiempirical mass formula can be extrapolated [Fig. 1(a)]. (2) Shell effects and other particularities do not dominate the stability relations of superheavy nuclei. (3) Available information about fission barriers and spontaneous fission half-lives and other transformation processes allows of simple extrapolation. It was concluded that if these assumptions make sense then nuclei very much heavier than those now known [shaded wedge-shaped region in Fig. 1(a)] will live long enough ($\sim 10^{-4}$ sec) to be subject to experimental observation. The exact location of the wedge-shaped region is sensitive to the constants in the semiempirical mass formula. The constants in the earlier formula used in reference 2 were based upon a nuclear radius of $1.48\times 10^{-13}A^{\frac{1}{3}}$ cm and put the tip of the wedge near $A\sim 650$, $Z\sim 170$. With the present constants of Green the radius is $1.2162\times 10^{-13}A^{\frac{1}{3}}$ and the region of appreciable lifetimes is now calculated to end near $A\sim 500$, $Z\sim 147$ [Fig. 1(b)]. Estimated contour lines for neutron binding energy B_n and fission barrier height E_f are shown in Fig. 2 and beta decay energy Δ in Fig. 3.

Spontaneous fission lifetimes and fission barriers are by far the most uncertain features in the extrapolation. Swiatecki[3] reasons that the nucleus at the top of the barrier is so highly deformed that nearly all shell structure can be considered to be broken up. Therefore the absolute energy of the top of the barrier should depend smoothly upon Z and A apart from even-odd effects, as confirmed (Swiatecki) by the available evidence. However, the energy of the starting point and, even more, the equilibrium deformation of the nucleus, are dependent upon shell effects. Swiatecki's analysis of the observational evidence suggests therefore that the major uncertainty in fission barrier heights for superheavy nuclei will arise from inability to predict equilibrium deformations in this so far unexplored region. In lieu of anything better we have extrapolated barrier heights via the liquid drop theory of fission of nuclei that are *spherical* at equilibrium. It is conceivable among other possibilities that the equilibrium shapes change drastically from one part of the superheavy region to another. In this event the nuclei in one region may be much more stable than the present predictions, and may not exist at all in the other region. Only observation can reveal the true stability relations.

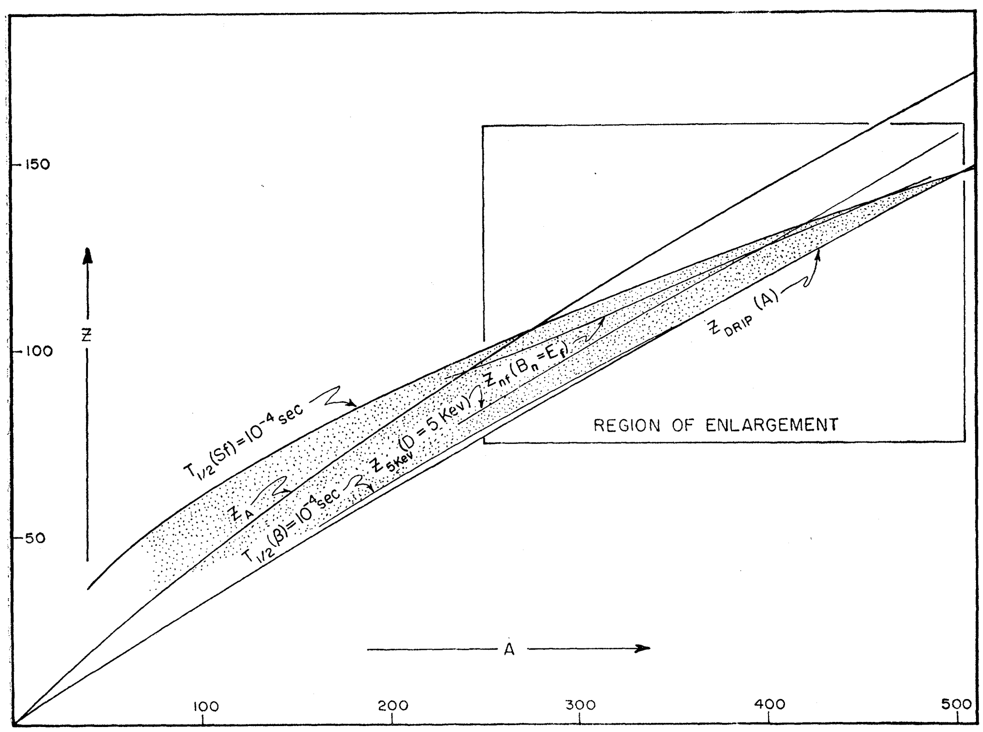
[1] Ghiorso, Harvey, Choppin, Thompson, and Seaborg, Phys. Rev. **98**, 1518 (1955). We made the January, 1953 count from the table of nuclei presented by Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953).

[2] J. A. Wheeler, in *Niels Bohr and the Development of Physics* (Pergamon Press, London, 1955), Chap. 9. A report of this work was also given at Geneva: J. A. Wheeler, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, August, 1955* (United Nations, New York, 1956), Vol. 2, p. 155. In the discussion at that meeting the question was raised by D. I. Blokhintsev (Vol. 2, p. 224) as to the stability of the K electron in the field of a nucleus of charge greater than 137. The reply (Vol. 2, p. 224–5) gave a brief report of the present considerations, including the binding energy of about 1.85 mc^2 for $Z=170$. A recent paper of Blokhintsev makes brief reference to the same question: D. I. Blokhintsev, Uspekhi Fiz. Nauk **61**, No. 2, 137 (1957).

[3] W. J. Swiatecki, Phys. Rev. **101**, 97 (1956).



(a)



(b)

FIG. 1. (a) Previously estimated limits of nuclear stability (taken from reference 2, where the Fermi semi-empirical mass formula constants were used). The dotted zone indicated the general region occupied by "normal" nuclei that live longer than 10^{-4} sec. Alpha-decay energies were estimated from the semiempirical mass formula; and alpha-decay rates, from the usual barrier penetration formula. The limit for stability against neutron loss was also calculated from the semiempirical mass formula without allowance for odd-even differences and may be significantly in error owing to the long extrapolation from the region of normal neutron-proton ratios. Beta-decay rates were estimated on the assumption of allowed transitions, and beta energies were estimated as if only odd-A values were relevant; both estimates were very crude, as the cross-hatching was meant to suggest. Spontaneous fission rates were estimated as if all nuclei had even-even character, whereas spontaneous fission will be hindered by a factor of the order of $10^{3\pm1}$ for odd A. No account was taken of occasional nuclei endowed with exceedingly high spin and with low decay rates which can stand outside the dotted region and still have lives greater than 10^{-4} sec. Insofar as nuclei with lives of 10^{-4} sec or more are subject to experimentation, the dotted region attributed a testable reality to nuclei twice as heavy as known nuclei. (b) Revised estimated limits of nuclear stability using the Green semiempirical mass formula constants of reference 15. In comparison with Fig. 1(a) the line of stability against neutron loss now rises to cross the line of 10^{-4}-sec spontaneous fission lifetime at $A\sim500$, $Z\sim147$. The line of 10^{-4}-sec beta lifetime based upon Eq. (41) of Sec. III now goes just a little above the neutron loss line.

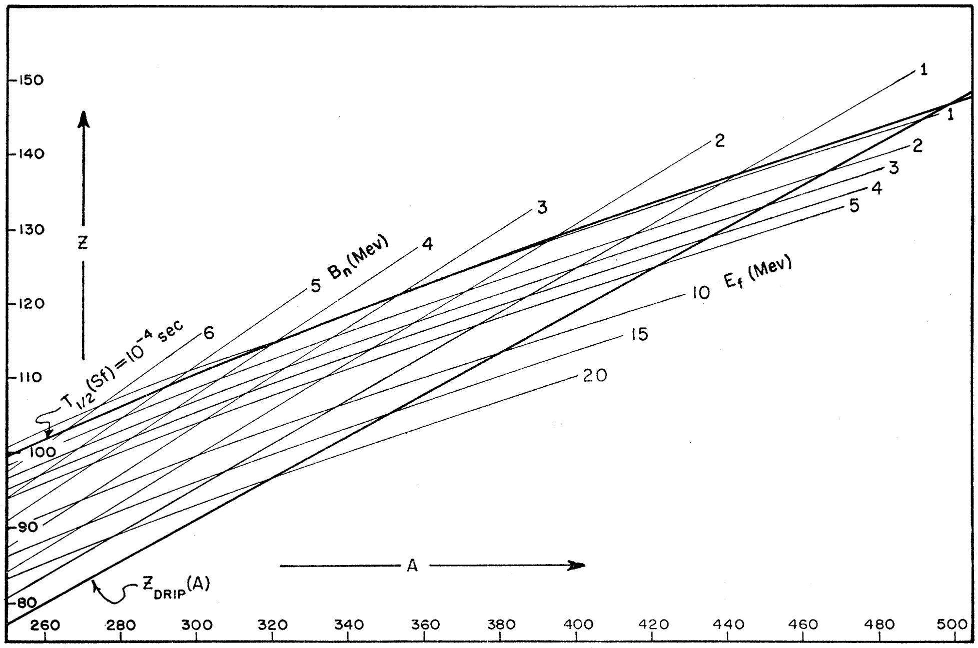


FIG. 2. Enlargement of outlined region of Fig. 1(b) showing estimated contour lines of specified neutron binding B_n(Mev) and fission barrier E_f(Mev).

If superheavy nuclei exist with properties anything like those predicted by assumptions (1)–(3), what will happen to their K electrons? What will be their beta-decay rates? What will be the prospects for building them up in a heavy neutron flux, either in the neutron core of a star or in a thermonuclear test like that of November 1, 1952? This paper deals with these three topics.

(1) The Dirac equation predicts for the lowest bound electron state in a pure Coulomb field the energy

$$\text{rest energy}+\text{binding energy}=mc^2[1-(Z/137)^2]^{\frac{1}{2}}. \quad (1)$$

This formula becomes unphysical for $Z>137$. This circumstance has been regarded sometimes as an argument against the stability of nuclei with charge above 137. However, in the work reported here (Sec. II) we have integrated the Dirac equation for Z up to 170, making allowance for the finite size of the nucleus. With this very essential refinement we find quite reasonable values for the binding energies of the innermost bound electrons, as illustrated in Fig. 4. We estimate that vacuum polarization and vacuum fluctuations alter the level position by only a few kev. Consequently we conclude that the behavior of the bound electrons around an extended nucleus interposes no limitations on nuclear stability above those indicated in Fig. 1(b).

(2) Electrons in the continuous spectrum carry away energy in beta-decay processes. For nuclei with $Z>137$ will not the wave amplitude for such electrons be exceptionally high near the nucleus? Will not the rate of beta decay therefore be speeded up anomalously? This speed-up is indeed significant, but in reference 2 it has already been taken into account in an approximate fashion in estimating the limit for 10^{-4} sec beta-decay half-life as indicated by the cross-hatched regions in Fig. 1(a). Section III reports a more detailed analysis of this speed-up effect based on a numerical integration of Dirac's equation for positive energies.

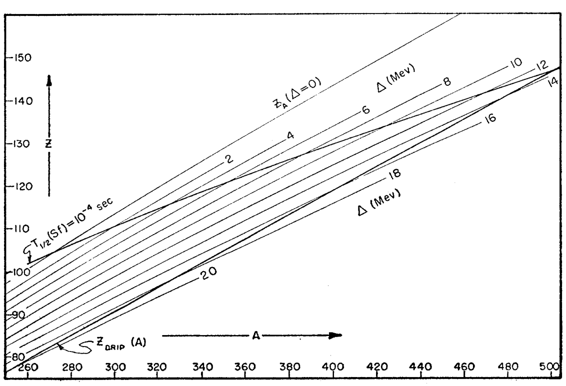


FIG. 3. Estimated contour lines of equal beta-decay energy, Δ(Mev).

We verified that (1) the energies of the tightest bound electron states and (2) beta-transition rates are perfectly normal for superheavy nuclei, not with the idea to make precision computations, but only to clarify the point of principle. We have not provided accurate tables of electronic binding energies for either $1s$ or higher states, such as would be needed to fix charge numbers from the usual measurements of internal conversion energies.

(3) Neutron capture and beta decay form the two obvious mechanisms for going to high A and Z (Sec. IV). However, an added neutron will have a high chance

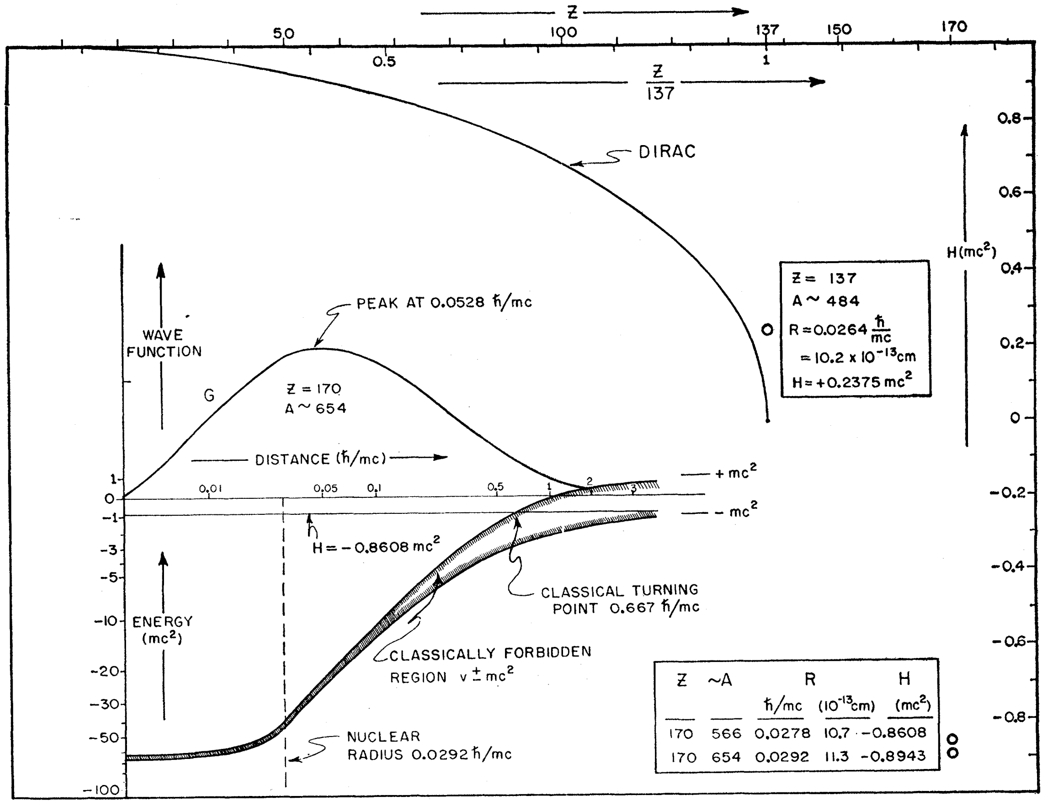


FIG. 4. Energy of the lowest bound electron level in the field of a nucleus of finite size. Smooth curve: Dirac value for point nucleus. Circles: Numerical calculation for nuclei of uniform charge density and of radius $A^{\frac{1}{3}}\times 1.3\times 10^{-13}$ cm. Lower insert: Sketch of potential and of large component of wave function, $G(r)$, for case $Z=170$, $A\sim 654$. The amplitude of the wave function is plotted on a linear scale, but distance and potential are plotted on hyperbolic sine scales to cover the wide range of these variables.

to destroy the nucleus unless the fission barrier is higher than the energy of condensation of the neutron. To keep this condensation energy as low as possible the excess of neutrons in the nucleus should stay as high as possible. The maximum excess corresponds to the line of "neutron drip" in Fig. 1(b). We calculate that to keep the path of buildup close to this line an exceedingly high neutron flux must be maintained. Presumably such a high neutron flux can be attained only at the fringe of a stellar neutron core. At the lesser flux obtainable in principle by sudden fission of a very large mass of uranium there are heavy losses by fission during the process of element building. Also losses by "delayed" fission are associated with the other essential process in buildup—beta decay—when the mass change exceeds the fission barrier, as is the case along the upper part of the wedge-shaped region in Fig. 1(b). Buildup of superheavy nuclei by collision of two lighter nuclei* is not treated here.

* *Note added in proof.*—Nobelium (102) has been produced by cyclotron bombardment of curium with high energy C^{13} ions, according to Fields, Friedman, Milsted, Atterling, Forsling, Holm, and Åström [Phys. Rev. **107**, 1460 (1957)].

II. SOLUTION OF DIRAC EQUATION FOR *K* LEVEL, AND CONTINUUM; *K*-LEVEL SHIFT BY POLARIZATION AND FLUCTUATIONS

We start with the idealization in which the electron is treated as a point particle, described by Dirac's Hamiltonian,

$$H=-(\boldsymbol{\alpha}\cdot\mathbf{p})-\beta+v(r). \tag{2}$$

Throughout Sec. II we express physical quantities in the following units: H, total energy, mc^2; v, potential energy, mc^2; p, momentum, mc; r, distance, $\hbar/mc$; angular momentum, $\hbar$. For central forces it is well known that an integral of the motion is the associated angular momentum

$$k=\beta[(\boldsymbol{\sigma}'\cdot\mathbf{r}\times\mathbf{p})+1], \tag{3}$$

where $\sigma_x'=\alpha_y\alpha_z/i$ and where the usual one-particle spectroscopic states belong, respectively, to the following k-values: $^2s_{\frac{1}{2}}$, $k=-1$; $^2p_{\frac{1}{2}}$, $k=1$; $^2p_{\frac{3}{2}}$, $k=-2$; $^2d_{\frac{3}{2}}$, $k=2$; $^2d_{\frac{5}{2}}$, $k=-3$; etc. Let a state be considered for which H and k take on proper values. Also let a factorization of the wave function into angular and radial parts be chosen such that (1) the radial part has

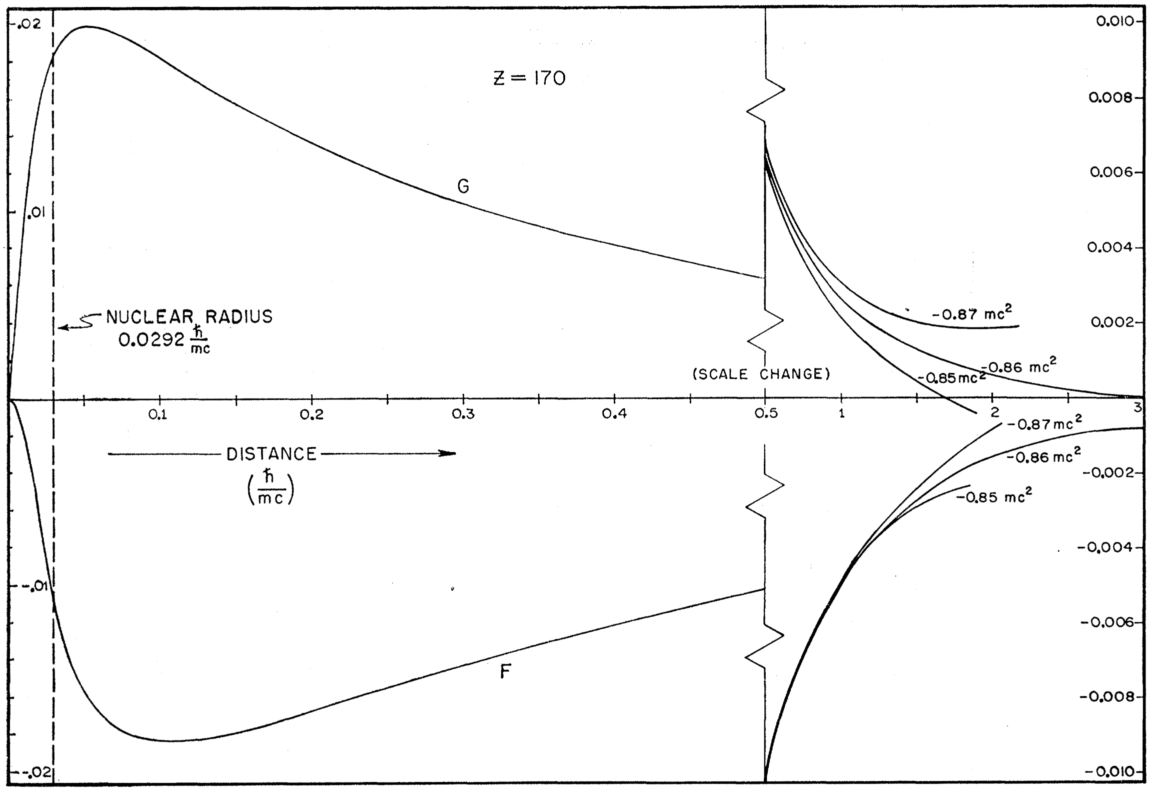


FIG. 5. Determination of K-level eigenvalue by trial and error. Case $Z=170$, with nuclear radius 0.0292 $(\hbar/mc)=11.3\times10^{-13}$ cm. The so-called large component of the radial part of the wave function is designated by G; the small one, by F. The individual curves are labeled according to the trial value of the energy (units mc^2).

two components,

$$\begin{pmatrix} r^{-1}F(r) \\ r^{-1}G(r) \end{pmatrix}, \tag{4}$$

and (2) the matrices relevant for the calculation of the radial part have the representation

$$\beta=\begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}; \quad r^{-1}(\boldsymbol{\alpha}\cdot\mathbf{r})=\begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}. \tag{5}$$

Then the radial part of the wave equation has the form[4]

$$\begin{aligned}(dG/dr)+(k/r)G-(H-v+1)F&=0,\\ (dF/dr)-(k/r)F+(H-v-1)G&=0.\end{aligned} \tag{6}$$

We adopt for the nucleus the model of a sphere of charge of uniform density and of radius

$$R(\mathrm{cm})=b\hbar/mc=r_0(\mathrm{cm})A^{\frac{1}{3}}. \tag{7}$$

Present evidence about the energy of the $2p\rightarrow1s$ transition of a μ meson in the field of force of a lead nucleus[5] and the scattering of high-energy electrons by heavy nuclei[6] argues for an effective value of r_0 in the neighborhood of 1.1 to 1.3×10^{-13} cm. Here in principle the quantity "effective radius" has to be taken to have slightly different values according as one deals with one physical property or another. Only if one treats the charge distribution as rounded off appropriately near the nuclear boundary can one account for both μ-meson levels and nuclear scattering by a single electrostatic potential.[7] We neglect this refinement in the present work and adopt the value

$$\begin{aligned} r_0&=1.3\times10^{-13}\text{ cm},\\ b&=3.37\times10^{-3}A^{\frac{1}{3}}.\end{aligned} \tag{8}$$

Thus we assume

$$\begin{aligned} v(r)&=-(Z\alpha/r) && \text{for } r>b,\\ v(r)&=(Z\alpha/b)[-1.5+0.5r^2/b^2] && \text{for } r<b,\end{aligned} \tag{9}$$

with $\alpha=1/137.04$; we consider r and a to be expressed in units $\hbar/mc=3.87\times10^{-11}$ cm, and take $k=-1$, corresponding to $^2s_{\frac{1}{2}}$ states.

For a point nucleus ($b=0$) with $Z<137$, Eq. (6) has the well-known ground-state solution

$$F=-N(Zr/137)^s\exp(-Zr/137), \tag{10}$$

$$G=N(137/Z)(1+s)(Zr/137)^s\exp(-Zr/137), \tag{11}$$

[4] See, for example, Leonard I. Schiff, *Quantum Mechanics* (McGraw-Hill Book Company, Inc., New York, 1949), p. 323.
[5] V. L. Fitch and J. Rainwater, Phys. Rev. **92**, 789 (1953).
[6] Hofstadter, Fechter, and McIntyre, Phys. Rev. **92**, 978 (1953).
[7] D. L. Hill and K. W. Ford, Phys. Rev. **94**, 1617 (1954).

TABLE I. Estimation of eigenvalues for K levels by the interpolation procedure described in the text. The first four lines test the linear variation with energy of the "discrepancy from exponential fall-off," D of Eq. (17). The next to the last column shows the values of D computed for comparision from the best fitting straight line, $D_{\text{linear}}=-32.310H-28.886$. The last column gives the energy eigenvalues, H, determined by linear interpolation in D. The quantity b measures the nuclear radius in units $\hbar/mc=3.87\times10^{-11}$ cm and r measures distance to the point of evaluation of the wave function in the same units.

Z	b	r	H_{trial}	Results of numerical integration 10^3G	$10^3dG/dr$	D	D_{linear}	$H_{\text{interpolated}}$
170	0.02778	1.20	−0.80	−3.032	−7.750	−2.563	−3.037	
			−0.87	+0.460	−3.973	−0.741	−0.776	−0.8940
			−0.89	1.505	−2.823	−0.129		
			−0.90	2.033	−2.233	+0.194		
170	0.02778	2.00	−0.89	0.090	−1.204	−0.852		−0.8944
			−0.90	1.275	−0.101	+1.095		
170	0.02920	1.75	−0.86	0.851	−1.250	−0.087		−0.8608
			−0.87	1.849	−0.277	+0.999		
137	0.02640	2.75	+0.235	6.543	−5.245	+0.153		+0.2375
			+0.250	1.123	−9.336	−0.781		

where $s^2=1-(Z/137)^2$ and where the normalization factor N is so adjusted that

$$\int_0^\infty (F^2+G^2)dr=1. \tag{12}$$

The solution is not normalizable in this sense when $Z\geqslant137$.

In the case of an extended nucleus the potential has a finite value and zero slope at the origin. The same is true of the kinetic energy. The solutions F and G therefore start off, respectively, proportional to r^2 and to r, as in the case of a free electron of higher kinetic energy. The coefficients in the power series expansions,

$$\begin{aligned} G&=r+g_3r^3+g_5r^5+\cdots,\\ F&=f_2r^2+f_4r^4+\cdots \end{aligned} \tag{13}$$

follow from simple recursion relations:

$$\begin{aligned} f_2&=-[H-1+(1.5Z\alpha/b)]/3,\\ g_3&=-[H+1+(1.5Z\alpha/b)][H-1+(1.5Z\alpha/b)]/6,\\ f_4&=(0.1Z\alpha/b^3)+[H+1+(1.5Z\alpha/b)]\\ &\qquad\times[H-1+(1.5Z\alpha/b)]^2/30. \end{aligned} \tag{14}$$

With starting values taken from the series (13), the differential Eqs. (6) were integrated numerically on the IBM card-programed electronic computer of Princeton's Forrestal Research Center by the method of Runge and Kutta[8] under the supervision of Robert Goerss and Mrs. Bernice Bender, to whom we express here our appreciation.

In the case of the K level one has of course to deal with an eigenvalue problem, to the solution of which the machine approximated by the method of trial and error, as seen in Fig. 5. The best two runs gave the final estimated energy value by an interpolation procedure based on the JWKB method. According to this method of approximation, the solution for G in the region of exponential fall or rise has—for arbitrary energy—the form

$$G(r)=[c(r)/\kappa^{\frac{1}{2}}(r)]\left[A\exp\left(-\int^r\kappa(r)dr\right)+B\exp\left(\int^r\kappa(r)dr\right)\right], \tag{15}$$

where $c(r)$ and $\kappa(r)$ are known functions of r and of energy, H. The departure of the energy from an eigenvalue is measured by the ratio B/A, or equivalently, by the quantity

$$D=(B/A)\exp\left(2\int^r\kappa(r)dr\right). \tag{16}$$

We assume that this quantity, calculated for a fixed r but for different energies, H, varies approximately linearly with H in the neighborhood of an eigenvalue. We checked this assumption in one case as shown by the first four entries in Table I. Here the value of D has been obtained from the computed $G(r)$ and dG/dr by way of a formula where all other quantities are easily found from the JWKB analysis:

$$D=\frac{G[\kappa-(d/dr)\ln(c/\kappa^{\frac{1}{2}})]+(dG/dr)}{G[\kappa+(d/dr)\ln(c/\kappa^{\frac{1}{2}})]-(dG/dr)}. \tag{17}$$

The assumption of approximate linearity, once having been checked, is used to estimate eigenvalues by interpolation, as listed in the other entries in Table I.

The K-level eigenvalues were computed for $Z=170$ for two values of the nuclear radius in order to check the sensitivity to this parameter. A decrease of the radius by $0.02920-0.02778=0.00142$ $\hbar/mc$ was found to increase the tightness of binding by

$$\begin{aligned}&[-0.8608\pm0.0002]-[-0.8943\pm0.0005]\\ &\qquad=[0.0335\pm0.0005]\ mc^2.\end{aligned}$$

[8] See for example William Edmund Milne, *Numerical Calculus* (Princeton University Press, Princeton, 1949), p. 135, paragraph 38, Eqs. (1) and (2).

An independent evaluation of the energy change by perturbation theory gives

$$\delta H = \delta b \langle \partial v(r,b)/\partial b \rangle$$
$$= (\delta b/b)(3Z\alpha/2b) \int_0^b (1-r^2/b^2) \times (F^2+G^2)dr \bigg/ \int_0^\infty (F^2+G^2)dr. \quad (18)$$

The product of the first two factors represents the change in the central value of the electrostatic potential energy, $67.008-63.744=3.264$ (in units mc^2); and the subsequent ratio of two integrals, for $b=0.0292$, represents the probability for the electron to be within the nucleus, multiplied by the effective average value of $(1-r^2/b^2)$. The perturbation value of $\delta H = 3.264(2.495 \times 10^{-6}/2.402\times 10^{-4}) = 0.03391$ agrees satisfactorily with the less precise difference obtained from the numerical integrations.

The great binding energy of the K electron, ~ 1.9 mc^2, evidently makes the K-absorption edge of a supernucleus overlap strongly the region of the pair creation threshold. Presumably also this photo cross section will show new and interesting features in its energy dependence, as compared with the conventional $1/\nu^3$ or $1/\nu^{3.5}$ variation of the cross section of light elements.

The Lamb shift and vacuum polarization corrections to $E(1s)$ that are so small for hydrogen are probably appreciable for a supernucleus. To estimate the magnitude of these corrections, it is simple but incorrect to multiply the hydrogenic terms in proportion to Z^4:

$$\Delta E_{\text{vac. pol.}} = -(4/15\pi) Z^4 \alpha^5 mc^2/n^3$$
$$= -27 \; Mc \text{ times } h \quad \text{or} \quad -1.12\times 10^{-7} \text{ ev} \quad \text{for } H(2s);$$
$$= -8(170)^4 1.12\times 10^{-7} \text{ ev} = -0.75 \text{ kev} \quad \text{for } Z=170,\ 1s.$$

$$\Delta E_{\text{fluct.}} = +1066 \; Mc \text{ times } h \quad \text{or} \quad +4.41\times 10^{-6} \text{ ev} \quad \text{for } H(2s);$$

$$8\times (170)^4 \times 4.41\times 10^{-6} \text{ ev} = +30 \text{ kev} \quad \text{for } Z=170,\ 1s.$$

The assumption of a point nucleus is wrong for both effects, and the assumption of Z^4 proportionality is wrong for the second effect. For this reason we calculated the first-order vacuum polarization for the extended nucleus, using the Dirac wave functions that we had found by numerical integration. We got the result

$$\Delta E_{\text{vac. pol.}} \text{ (first order)} = -9 \text{ kev} \quad (1s;\ Z=170,\ R=11\times 10^{-13} \text{ cm}). \quad (19)$$

The fluctuation part of the Lamb shift is a much more difficult problem. The usual helpful division[9] of this effect into relativistic and nonrelativistic parts is out of question here owing to the relativistic binding of the electron in its ground state, 1.86 mc^2. No available analysis of the Lamb shift appears to be suitable for the present case.[10] Consequently we found ourselves forced to a very crude order-of-magnitude treatment. We went back to Bethe's original simple way of estimating the Lamb shift,[11]

$$\Delta E = (2\alpha/3\pi) \sum_n |\mathbf{v}_{0n}/c|^2 (E_n - E_0) \ln\left(\frac{K}{E_n - E_0}\right),$$

with K a cutoff energy of the order of mc^2. We replaced the factor $\mathbf{v}/c$ by the Dirac matrix $\boldsymbol{\alpha}$. In this matrix element we made no correction for the retardation factor, e^{ikx}. This factor is unimportant for hydrogen but for a supernucleus ought to reduce the average value of the matrix element in a major way. A proper correction for this big effect would make the simple Bethe treatment completely inapplicable. We used it nevertheless in default of anything better. We assumed that most of the line strength is concentrated in transitions from $1s$ $[E(1s) = -0.86\ mc^2]$ to $2p$ $[E(2p_{\frac{1}{2}}) \doteqdot 0.8\ mc^2]$, $3p$, $\cdots$ and other levels near mc^2. Accordingly we inserted for $E_n - E_0$ an average value of 1.86 mc^2. The logarithm is of course quite uncertain. It seems not unreasonable to assume for it an effective value somewhere between -1 and $+1$. The expression $\sum_n |\boldsymbol{\alpha}_{0n}|^2$ according to the sum rule has the value $(\alpha_x^2)_{00} + (\alpha_y^2)_{00} + (\alpha_z^2)_{00} = 3$. Accordingly we got an estimate for the fluctuation part of the Lamb shift of the order

$$\Delta E_{\text{fluct.}} \sim \pm (2\alpha/\pi) 1.86\ mc^2 \sim \pm 4 \text{ kev}. \quad (20)$$

It is quite possible that the Lamb shift has several times the total value estimated here,

$$\Delta E_{\text{Lamb}} = \Delta E_{\text{vac. pol.}} + \Delta E_{\text{fluct.}} \sim -9 \text{ kev} \pm 4 \text{ kev}. \quad (21)$$

Even so, it seems unlikely to make really substantial changes either in the qualitative nature of the electronic wave function or in the binding energy, 1.86 $mc^2 = 950$ kev. Consequently we have neglected both parts of the Lamb shift in the further analysis of superheavy nuclei.

An appendix reports the details of the calculation of the $1s$ level shift due to vacuum polarization by a finite nucleus.

Charge numbers greater than $Z=137$ cause no trouble for bound electrons. What is the effect of high charge on the electrons emitted in beta decay, and on the rate of beta decay?

The solutions of the Dirac equation for $2s_{\frac{1}{2}}$ states in the continuum were obtained by the same method previously employed for bound states. The numerical integration was stopped at the second maximum of G.

[9] Baranger, Bethe, and Feynman, Phys. Rev. **92**, 482 (1953); see also the summary in J. M. Jauch and F. Rohrlich, *The Theory of Photons and Electrons* (Addison-Wesley, Cambridge, 1955), Chap. 15.

[10] We are indebted to Professor Freeman Dyson and Dr. Eyvind Wichmann for helpful discussions of this question.

[11] H. A. Bethe, Phys. Rev. **72**, 339 (1947).

The two components, F and G, were extrapolated from this region to infinity by way of the JWKB approximation, and normalized so that the asymptotic value of $(F^2+G^2)_{\text{average}}$ took on the usual standard magnitude of $\frac{1}{2}$. The results of the numerical integration, so normalized, are shown in Fig. 6. The portions of F and G relevant to beta decay lie of course inside the nuclear boundary, indicated by the dashed vertical strokes.

For the fast decaying nuclei in which we are interested, we shall limit attention to high electron energies,

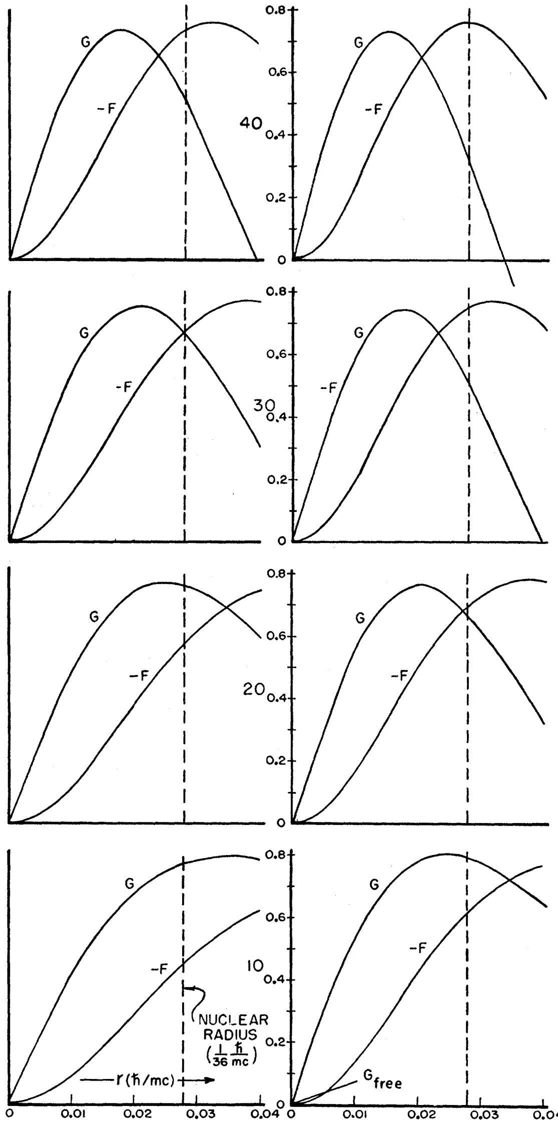


FIG. 6. Normalized solutions of the Dirac equation for energies in the continuum, $H=10$, 20, 30 and 40 (units mc^2). Left-hand column, $Z=137$; right-hand column, $Z=170$. The radius of the equivalent uniform nuclear charge distribution was taken in all eight cases to be $(1/36)(\hbar/mc)$ (indicated by dashed vertical lines). The normalization is such that at infinity $F_{\max}{}^2+G_{\max}{}^2=1$. The straight line segment in the lower right-hand case shows the initial behavior of the wave function near the origin as it would have been in the absence of nuclear attraction: $G=(121/220)^{\frac{1}{2}} \times \sin[(99)^{\frac{1}{2}}r]$.

of the order 10 mc^2 to 40 mc^2. For such energies we should now like to show that the JWKB approximation gives quite reasonable values, not only for the wave function near the second maximum, but also right down to the surface of the nucleus. This circumstance indicates that it is only necessary to resort to numerical integration to determine the wave function within the nucleus for states of other angular momentum; that the JWKB procedure is adequate outside.

We transform the coupled pair of first order differential equations (6) to the standard JWKB form of a single second order equation by way of the substitution given by Bartlett and Welton[12]:

$$G(r)\equiv(H-v+1)^{\frac{1}{2}}y(r). \tag{22}$$

Then the equation for $y(r)$ becomes

$$d^2y/dr^2=-K^2(r)y=\kappa^2(r)y. \tag{23}$$

Here one form or the other of the right-hand side of the equation is preferable according as one is dealing with the region of r where y is oscillatory or exponential. In either case the coefficients of y are abbreviations:

$$\begin{aligned}K^2(r)\equiv-\kappa^2(r)\equiv-(k+\tfrac{1}{2})^2/r^2+(H-v)^2-1\\+[H-v+1]^{-1}[(k/r)(dv/dr)-\tfrac{1}{2}d^2v/dr^2]\\-\tfrac{3}{4}[H-v+1]^{-2}(dv/dr)^2.\end{aligned} \tag{24}$$

Here a correct use of the transformation (22) would have given for the first term $-k(k+1)/r^2$, with all the other terms in (24) as listed. However, the change to $-(k+\frac{1}{2})^2/r^2$ is required[13] (1) to make the JWKB solution for $G(r)$ behave near the origin as $r^{|k+\frac{1}{2}|+\frac{1}{2}}$, as demanded by the power series expression for $G(r)$, (2) to give in similar well-studied problems the proper asymptotic phase shift, and (3) to make the JWKB approximation fit closer to the numerical results in cases previously investigated. This granted, the differential equation has a turning point, TP, for all k values, including $k=0$. Outside this turning point the normalized JWKB solution has the form

$$G(r)=2^{-\frac{1}{2}}(1-H^{-2})^{\frac{1}{4}}(H-v+1)^{\frac{1}{2}}K^{-\frac{1}{2}}(r)\times\sin\left[(\pi/4)+\int_{\mathrm{TP}}^{r}K(r)dr\right], \tag{25}$$

and between the turning point and the origin it has the form

$$G(r)=2^{-\frac{3}{2}}(1-H^{-2})^{\frac{1}{4}}(H-v+1)^{\frac{1}{2}}\kappa^{-\frac{1}{2}}(r)\times\exp\left[-\int_{r}^{\mathrm{TP}}\kappa(r)dr\right]. \tag{26}$$

We do not use (26). We use (25) to get G outside the

[12] J. H. Barlett, Jr., and T. A. Welton, Phys. Rev. **59**, 281 (1941).

[13] Yost, Wheeler, and Breit, Phys. Rev. **49**, 174 (1936).

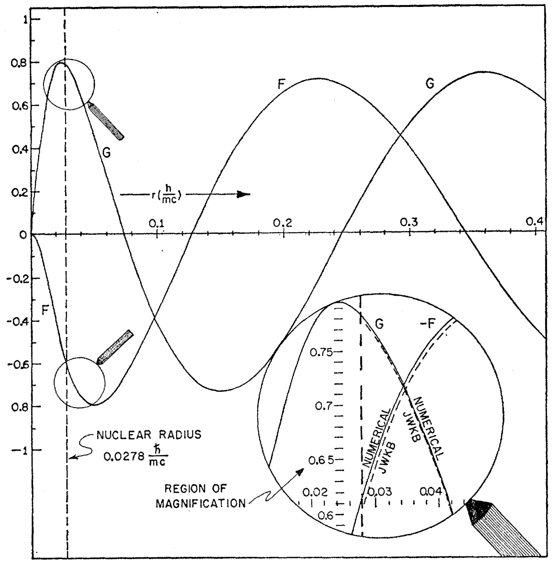


FIG. 7. Comparison of numerical and JWKB solutions of radial wave equation for case of a nuclear charge $Z=170$, radius $(1/36)(\hbar/mc)$, and rest plus kinetic energy of 10 mc^2 at infinity. The numerical solution was normalized to the JWKB formula at the point $r=0.248$ (units $\hbar/mc$). Particularly relevant is the close agreement of slopes of numerical and JWKB solutions at the nuclear surface.

nucleus, and compute F from the formula

$$F=(H-v+1)^{-1}[(dG/dr)+(k/r)G]. \tag{27}$$

Consider the cases of numerical integration summarized in Fig. 6. The JWKB fit to these results will be least accurate when (1) the charge is greatest ($Z=170$), and (2) the energy is least ($H=10$). For this case we have compared numerical and exterior JWKB results in Fig. 7. Evidently the fit is good down to the nuclear radius. Even the slopes at the nuclear surface agree much better than we have any right to expect:

$$\begin{aligned}(dG/dr)_{\text{numerical}}&=-7.92,\\ (dG/dr)_{\text{JWKB}}&=-7.66.\end{aligned} \tag{28}$$

In the absence of nuclear attraction the radial wave function, after normalization, has the form

$$\begin{aligned}G(r)&=2^{-\frac{1}{2}}(1-H^{-1})^{\frac{1}{2}}\sin[(H^2-1)^{\frac{1}{2}}r],\\ F(r)&=2^{-\frac{1}{2}}(1+H^{-1})^{\frac{1}{2}}\{\cos[(H^2-1)^{\frac{1}{2}}r]\\ &\quad-(H^2-1)^{-\frac{1}{2}}r^{-1}\sin[(H^2-1)^{\frac{1}{2}}r]\}.\end{aligned} \tag{29}$$

At the origin the component F is negligible compared to G. Consequently the probability, $\psi^*\psi$, to find an electron at the origin is proportional to G^2/r^2; that is, to $(dG/dr)_0{}^2$. This circumstance makes it reasonable to define a "central attraction factor," a_{central}, by the formula

$$a_{\text{central}}=\frac{[(dG/dr)_0{}^2]_{\text{actual field}}}{[(dG/dr)_0{}^2]_{\text{zero field}}}. \tag{30}$$

This factor is plotted as a function of energy in Fig. 8. The factor increases monotonically as the energy is decreased. There is no evident trace of anything like the Ramsauer effect found in the interaction between electrons and atoms. That effect occurs when the electron wavelength changes strongly over one electron wavelength. Then the ratio, (amplitude of the wave function inside)/(amplitude of the wave function outside), shows characteristic resonances. This effect does not occur in appreciable measure in the present problem because we are dealing with a smoothly varying potential and electron energies large compared to the depth of the potential.

Another measure of the attraction effect of the nucleus is the "average attraction factor," a_{average}, defined as the ratio of the values of the integral $\int_0^b(F^2+G^2)dr$, calculated (1) with the charge and (2) without charge,

$$a_{\text{average}}=\left(\int_0^b(F^2+G^2)dr\right)_{\text{actual field}}\bigg/\left(\int_0^b(F^2+G^2)dr\right)_{\text{zero field}}. \tag{31}$$

This average attraction factor is also plotted in Fig. 8.

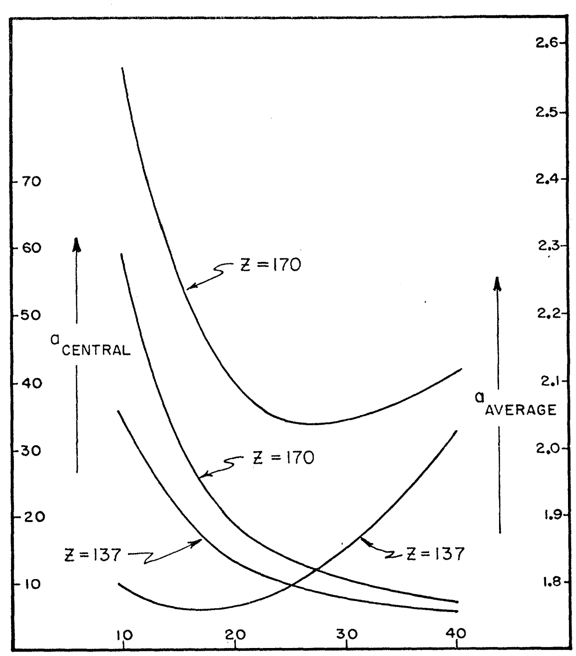


FIG. 8. "Central" and "average" attraction factors as a function of electron energy for two values of the nuclear charge. At the origin (where the component of the radial wave function F is negligible compared to G) the probability, $\psi^*\psi$, to find an electron is proportional to G^2/r^2; that is, to $(dG/dr)_0{}^2$. The ratio of this central probability for the actual field to its value for zero field is the central attraction factor, a_{central}. The average attraction factor, a_{average}, is the ratio of the values of the integral of the probability over the volume of the sphere with radius b, with and without uniform charge Z within the sphere.

This quantity is smaller than the "central attraction factor" for the following simple reason: (1) The electronic kinetic energy is roughly 40 mc^2 greater inside the nucleus than outside. (2) Consequently an electron with an energy of 10 mc^2 has its wave number increased by a factor of about 5. (3) Therefore the wave function varies more rapidly over the region $r=0$ to $r=b$ when the potential is present than when it is absent. (4) Hence an "interference effect" reduces the integral $\int_0^b(F^2+G^2)dr$ more, proportionately, for the case of Coulomb attraction. (5) This interference effect compensates in considerable measure the direct effect of the Coulomb field in increasing the probability amplitude at the center—the effect seen in a_{central}.

The "average attraction factor" shows a minimum for kinetic energies of 20 to 30 mc^2, depending upon charge. At lower energies the effect of the Coulomb attraction rapidly increases and becomes dominating, whereas the factor measuring the interference effect approaches a constant value. At higher energies interference begins to cut down the integral $\int_0^b(F^2+G^2)dr$ even for the free-particle case; and this diminution proceeds faster for the free-particle case than for the case of Coulomb attraction, leading to a rise in a_{average}.

III. BETA DECAY OF A SUPERNUCLEUS

The detail with which one analyzes the beta decay of a supernucleus will depend upon one's objectives. The most primitive arguments show that heavy nuclei will have beta half-lives longer than 10^{-5} sec. A more careful analysis allows for the reduction of the matrix elements through interference effects by way of one or another idealized model—a free nucleon picture, or a harmonic oscillator model. A still more precise treatment would decompose the contributions to the transition probability by the method of spherical harmonics. This treatment, though complicated to carry out, would be easy in principle to formulate, were the four particles in question described by scalar wave functions. Suppose one could write the electron and neutrino wave functions in the form

$$\psi_e=\exp(i\mathbf{k}_e\cdot\mathbf{x}), \tag{32}$$

$$\psi_\nu=\exp(i\mathbf{k}_\nu\cdot\mathbf{x}). \tag{33}$$

The transition probability, integrated over angles of emergence of electron and neutrino, would be proportional to

$$P=\int\int\left|\int\int\int\psi_p{}^*\psi_e{}^*\psi_\nu{}^*\psi_n dxdydz\right|^2 d\Omega_e d\Omega_\nu. \tag{34}$$

The method of analysis into spherical harmonics, Y_{lm}, reduces this expression to the form

$$P=\sum_{l,m,\lambda,\mu}|c_{lm\lambda\mu}|^2, \tag{35}$$

where

$$c_{lm\lambda\mu}=(4\pi)^2(-i)^{l+\lambda}\int\int\int\psi_p{}^*Y_{lm}(\theta,\varphi)[F_l(k_er)/k_er] \times Y_{\lambda\mu}{}^*(\theta,\varphi)[F_\lambda(k_\nu r)/k_\nu r]\psi_n r^2\sin\theta drd\theta d\varphi, \tag{36}$$

and where F_λ is that solution of

$$d^2F_\lambda/dr^2+[k_\nu{}^2-\lambda(\lambda+1)r^2]F_\lambda=0 \tag{37}$$

which is regular at the origin and is normalized to unit amplitude at infinity. To allow for Coulomb attraction it would only be necessary to replace the free-particle solution, F_l, of the given angular momentum by the solution of the wave equation with attractive potential. The analog of this procedure for spinor wave functions, with scalar and tensor beta-decay coupling, has been described by Takebe in three articles.[14] One can hope to go further with the program that Takebe has started when one knows nucleonic wave functions well enough to justify such precision. In the meantime it is reasonable to proceed with a less detailed analysis.

The most elementary analysis proceeds as follows. The semiempirical formula for nuclear binding energies[15] allows one to estimate for any given mass value the maximum number of neutrons which a nucleus can bind, as indicated by the appropriate line in Fig. 1(b). When one of these neutrons transforms to a proton, the beta-decay energy will have the maximum possible value consistent with the given choice of A, in so far as one looks apart from odd-even effects, When this correction is included, one can get still higher energies, as indicated in Table II. Consider a *single* nucleon bound in a potential whose value alters by 26 mc^2 when the particle changes its character from a neutron to a proton. Let the region of binding be so small that one can overlook any decrease of the beta-decay matrix element by reason of interference effects. Then the matrix element has the same value as one has for the decay of a free neutron. The lifetime will be smaller in

---

[14] H. Takebe, Progr. Theoret. Phys. Japan **12**, 561, 574, and 747 (1954). See also M. Yamada, Progr. Theoret. Phys. Japan **10**, 245 (1953) and R. Nataf, Compt. rend. **238**, 1012 (1954).

[15] See C. Coryell, in *Annual Review of Nuclear Science* (Annual Reviews, Inc., Stanford, 1953), Vol. 2 Chap. 13, for a review of different formulations of the semiempirical mass formula and agreement with experiment; also A. E. S. Green, *Nuclear Physics* (McGraw-Hill Book Company, 1956). We use the semiempirical formula without odd-even corrections in the form

$$M(Z,A)=A-C_1A+C_2A^{\frac{2}{3}}-C_3Z+C_4A(\tfrac{1}{2}-Z/A)^2+C_5Z^2/A^{\frac{1}{3}},$$

with Green's constants,

$$C_1=7.9357\text{ mMU},\quad C_2=19.120\text{ mMU},\quad C_3=0.84\text{ mMU},$$
$$C_4=101.78\text{ mMU},\quad C_5=0.07628\text{ mMU}.$$

$$M_{\text{neutron}}=1.008982\text{ amu}.\quad 10^3\text{ mMU}=1\text{ amu}=931.14\text{ Mev}.$$

H. B. Levy [Phys. Rev. **106**, 1265 (1957)], gives a new empirical equation for atomic masses in terms of first and second powers of A and Z with different coefficients for different nuclear shell regions. Separation into shell regions gives considerable improvement in reproduction of atomic masses. The form of the equation in each shell region is chosen for maximum algebraic simplicity and does not purport to have any theoretical justification. Therefore it is not suitable for the present substantial extrapolations beyond the known nuclei.

TABLE II. Beta-decay energies of nuclei at the limit of stability with respect to neutron emission, as estimated from the semiempirical mass formula.

A	Z_{drip}	$Z_{A-\frac{1}{2}}$	E_β (odd-A nucleus) Mev	E_β (odd-odd nucleus) Mev
500	147.0	171.2	13.2	13.8
400	119.2	143.4	15.8	16.5
350	105.2	128.6	17.0	17.7
300	91.2	113.2	18.1	18.7

inverse proportion to the decay factor, f, of beta-decay theory.[16] Consequently one estimates on this primitive basis a half-life

$$\begin{aligned} t_{\frac{1}{2}}(27\ mc^2) &= t_{\frac{1}{2}}(26\ mc^2+1mc^2) \\ &= [f(2.53)/f(27)]t_{\frac{1}{2}}(1.53+1=2.53\ mc^2) \\ &= 1.69\times 768/4.8\times 10^5 \\ &= 2.7\times 10^{-3}\ \text{sec.} \end{aligned} \tag{38}$$

Let the idealized bound nucleon be supposed to make its transition at the center of a charge cloud that (1) simulates the action of the actual nucleus on the electronic wave function but (2) does not have any direct effect on nucleonic processes. Then the rate factor of beta-decay theory is speeded up by the attraction effect (a_{central} of Fig. 8) to the value

$$f_{\text{modified central}}(H_0) = \int_1^{H_0} (H_0-H)^2(H^2-1)^{\frac{1}{2}}Ha_{\text{central}}(H)dH, \tag{39}$$

$$f_{\text{modified central}}(27) \doteq 4.8\times 10^5\times 39 = 1.9\times 10^7 \quad (\text{for } Z=147),$$

and the lifetime diminished to

$$t_{\frac{1}{2}} \doteq 1300\ \text{sec}/1.9\times 10^7 = 7\times 10^{-5}\ \text{sec.} \tag{40}$$

The primitive model of a single well-localized nucleon endowed with all the transition energy gives a much exaggerated idea of the speed-up of beta decay for a supernucleus: (1) in an actual nucleus the strength of the transition will be divided among many states; the residual nucleus will be left on the average with a number of Mev of excitation energy (ordinarily enough to drive off a "delayed" neutron); and the average energy of the transition will be less than has been supposed; (2) the Pauli principle will prevent transitions of the neutron to a state already occupied by a proton; and (3) the variation of the electron and neutrino wave functions over the nucleus will cut down the effective value of the attraction factor by interference (Fig. 8). Consequently it is easy to conclude that the mean life for beta decay will be longer than 10^{-4} sec for a supernucleus with $Z=147$, even with the maximum possible neutron-proton ratio.

It may become interesting to have a better estimate of mean life for beta decay than allowed by these primitive considerations. For this purpose two extreme and rather different models present themselves. In one the binding of the neutrons is idealized by a harmonic oscillator potential with one force constant; a different force constant is assumed for the protons; spin-orbit coupling is neglected; and the force constants are so chosen as to make the mean squared distance of the average nucleon from the center equal to the value estimated from μ-meson and electron scattering experiments by Ford and Hill.[7] The highest occupied levels of neutrons and protons are assigned an energy separation so as to agree with the mass difference $M(Z,A)$ and $M(Z+1,\ A)$ as determined from the semiempirical mass formula. The beta-decay rate is determined by matrix elements qualitatively of the form (36). Here the nucleonic wave functions are now harmonic oscillator functions. For the electron the radial function $F_l(kr)/kr$ in (36) is replaced by the proper spinor wave function, calculated as already described; similarly for the neutrino.

This is the plan of one method of analysis, but we have done nothing to carry it out, because there exists a still simpler method of analysis. We idealize the neutrons as one Fermi gas occupying the volume $(4\pi r_0^3/3)A$, the protons as a second gas filling the same volume in ordinary space. In momentum space the two kinds of particles occupy spheres as indicated in Table III. Beta decay is idealized as the transformation of a free neutron to a free proton, with different zeros of nucleon energy because of the different average binding forces experienced by the two kinds of particle. The beta decay of $Z=147$, $A=500$ releases at most 26 mc^2, according to Table II. The recoil of the decaying nucleon can at most be 26 mc. This amount is small compared to the difference of 136 mc in Fermi momenta of neutrons and protons. Moreover, the recoil is random in direction. A nucleon that starts as a neutron outside the Fermi sphere of the protons will be transformed—in the absence of the Pauli principle—to a proton inside that sphere with almost the same probability that a neutron inside will be transformed into a proton outside. Therefore it is reasonable to neglect altogether the recoil of the nucleon in the act of beta decay. This

TABLE III. Occupied spheres in momentum space as calculated for the example $A=500$, $Z=147$, $r_0=1.23\times 10^{-13}$ cm, from the formula $P_N=602\ mc(N/A)^{\frac{1}{3}}$.

	Fermi *kinetic* energy, F	Fermi momentum, P
Neutrons	78.3 mc^2	536 mc
Protons	43.6 mc^2	400 mc
Difference	34.7 mc^2	136 mc

[16] As tabulated for example by E. Feenberg and G. Trigg, Revs. Modern Phys. 22, 399 (1950), on the basis of the definition,

$$f(H_0) = \int_1^{H_0} (H_0-H)^2(H^2-1)^{\frac{1}{2}}HdH,$$

where H_0 is the upper limit of the kinetic energy of the electron, plus its rest energy, in units mc^2.

simplification leads to this conclusion: Every neutron outside the Fermi sphere of the protons decays quite uninhibited by the Pauli principle; every neutron inside is completely blocked from beta decay. This model leads to the following formula for the beta half-life of the nucleus:

$$0.693/\lambda_\beta = t_{\frac{1}{2}} = \frac{1300 \text{ sec}}{N_{\text{unhindered}} f(\Delta M)\langle a_{\text{average}}\rangle}. \quad (41)$$

Here $N_{\text{unhindered}} = N - Z$ is the number of neutrons with momentum greater than the Fermi momentum of the protons. The remaining factors in the denominator stand for

$$f(\Delta M)\langle a_{\text{average}}\rangle = \int_1^{H_0} (H_0 - H)^2 (H^2 - 1)^{\frac{1}{2}} H a_{\text{average}}(H) dH, \quad (42)$$

where $a_{\text{average}}(H)$ is the volume average attraction factor defined previously [Eq. (31), Fig. 8] and $(H_0 - 1)mc^2 = c^2 \Delta M$ is the energy difference between ground states of the initial and final nucleus. Illustrative numbers for two rather extreme cases appear in Table IV.

The Fermi gas model appears at first sight to be the very opposite of the harmonic oscillator model. The one picture treats the variation of electron and neutrino wave functions over the nucleus as sufficient to allow the leptons to feel out the momentum change of the decaying nucleon. The recoil momentum is regarded as concentrated in the individual heavy particle, not the nucleus as a whole. On the other hand, the harmonic oscillator model makes the source of the oscillator potential—the nucleus itself—the recipient of all recoil momentum. The contrast would appear still more clearly if the harmonic oscillator potential were replaced by an infinitely high square well potential, and if a detailed counting of particle states were replaced by a statistical analysis. Then the states of the one model would be identical with the states of the other model. But in the one case the particles are free, and take up momentum on an individual basis; in the other case the potential wall does all the taking up of momentum.

Different as these two models appear, we expect from them not very different results for the supernuclei; (1) The actual change of the lepton wave functions over the nucleus is intermediate between the rapid variation envisaged in the one model and the nearly constant behavior imagined in the other. (2) Detailed numerical application of the two quite different methods to another problem—charge exchange reaction between a μ meson in a K orbit and the central nucleus—led in the two cases to reasonably compatible results for the calculated reaction rate.[17] (3) That problem was similar to the present problem in the following respects: (a) the ratio, (lepton wavelength/nuclear dimensions), agrees at least as to order of magnitude in the two problems (80-Mev neutrino and the smaller nuclear radius of O^{16} in the charge exchange reaction; in the β-active supernuclei, a greater radius but a kinetic energy of the electron at the nucleus of the order of 25 Mev); (b) as an incidental and not very important point it happens that the variation of the wave function over nuclear dimensions is not very great for the other particle participating in the reaction (μ meson in the charge exchange reaction; ~10-Mev neutrino in the β decay); (c) the change in nucleon kinetic energy in both processes is of the order of 10 Mev. In view of the rough agreement of the two methods in the μ-meson case and the similarity of that problem to the present one, it appears reasonable to use the simple Fermi gas model and Eq. (41) to estimate the rate of beta decay of a supernucleus, as in the following section.

TABLE IV. Beta-decay speed-up factor after averaging over both volume and energy, Eq. (42).

Z	H_0 in mc^2	$f(\Delta M)\langle a_{\text{average}}\rangle$	$\langle a_{\text{average}}\rangle$
170	31	2.22×10^6	2.33
137	31	1.69×10^6	1.78

IV. BUILDUP OF SUPERHEAVY NUCLEI BY MASSIVE NEUTRON IRRADIATION

Neutron addition moves the representative point of the nucleus one unit to the right in Fig. 1(b) and beta decay moves it one unit upward. Both processes are required to carry a nucleus up into the wedge-shaped region near $A = 500$, $Z = 147$. Upward motion associated with beta decay occurs at its own natural rate, while the rate of motion to the right depends on the neutron flux. If this flux is too small, the representative point will move up on too steep a slope. It will intersect prematurely the line where the spontaneous fission rate is 10^{-4} sec or less, and the nucleus will be destroyed. Actually the danger point comes even sooner. The fission barrier E_f decreases as the nucleus moves up step by step via beta decay to higher Z values. The neutron binding B_n increases. Soon B_n exceeds E_f. Then the nucleus will be destroyed with very high probability by the first neutron it captures.

A rough semiquantitative estimate of the flux requirements for building superheavy nuclei is in order. For this purpose we shall make some drastic simplifications in the treatment of both essential processes, neutron addition and beta decay. (1) We neglect (n,α) processes altogether. The rate of spontaneous alpha emission for these very heavy nuclei is calculated to be much less than the rate of spontaneous fission, and to rise more slowly with increasing excitation. (2) When B_n exceeds E_f, we assume that neutron uptake leads to fission with 100% probability. (3) Conversely, when

[17] J. Tiomno and J. A. Wheeler, Revs. Modern Phys. 21, 153 (1949).

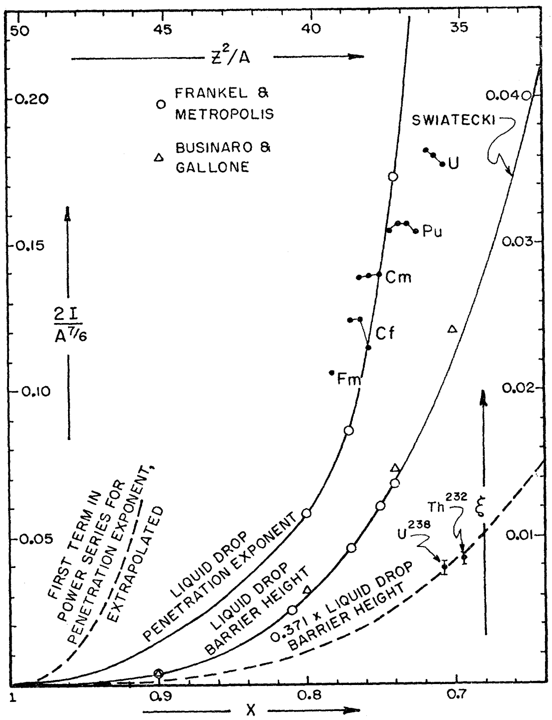


FIG. 9. Liquid-drop fission barrier height and reduced penetration exponent as functions of $x=(Z^2/A)/(Z^2/A)_{\text{crit}}$. The value $(Z^2/A)_{\text{crit}}=50.37$ used here is calculated from Green's semiempirical mass formula constants (reference 15). Points in circles are calculated from the work of Frankel and Metropolis (reference 18), those in triangles from Businaro and Gallone (reference 18). The reduced penetration exponents $2I/A^{7/6}$ for even-even nuclei from observed spontaneous fission rates are from Wheeler (reference 2). The smooth line of liquid-drop fission barrier height ξ in terms of undistorted surface energy is from the formula of Swiatecki (reference 18). The dashed line is Swiatecki's liquid drop barrier height multiplied by the constant factor 0.371 to make it pass between the points for Th^{232} and U^{238} calculated from known barrier heights from Wheeler (reference 19). There is no *theoretical* basis for assuming this constant correction factor nor for using it to extrapolate barrier heights to very heavy nuclei.

E_f exceeds B_n, we assume that radiative capture is the only neutron uptake process.

The line in Fig. 1(b) where $E_f=B_n$ is calculated with B_n values from the semiempirical mass formula and with fission thresholds E_f estimated in the following very crude manner, neglecting all variations from nucleus to nucleus due to shell effects and other particularities. (1) We take the detailed liquid drop calculations of Frankel and Metropolis, Businaro and Gallone, and Swiatecki[18] for $\xi=E_f/[4\pi r_0^2\times(\text{surface tension})\times A^{\frac{2}{3}}]$ as a function of $x=(Z^2/A)/(Z^2/A)_{\text{critical}}$. (2) We adopt for $(Z^2/A)_{\text{critical}}$ the value 50.37 based on Green's evaluation of the constants in the semiempirical mass formula.[15] (3) We do *not* evaluate the factor of proportionality between E_f and $\xi A^{\frac{2}{3}}$ from the mass formula, but (Fig. 9) from the empirical values[19] of E_f for Th^{232} and U^{238}. These data lead to a proportionality constant of 6.64 Mev, only 37.1% of the liquid drop value of $4\pi r_0^2\times(\text{surface tension})=17.89$ Mev (Green's constants). The liquid drop value refers of course to a spherical nucleus, whereas the observed fissionable nuclei are strongly deformed in the ground state. There is absolutely no warrant for assuming deformations sufficiently comparable for all the very heavy nuclei to justify a constant proportionality factor in the formula

$$E_f=6.64 \text{ Mev } \xi(x)A^{\frac{2}{3}}. \tag{43}$$

This formula is only an extrapolation formula relative to which nature may have surprises to offer.

When $E_f>B_n$ we have only the (n,γ) process to consider. The neutrons will be supposed to have energies of the order of 5 kev, corresponding to typical stellar temperatures, whether created naturally or artifically. When the binding energy set free on addition of a neutron is very low, as it is near the line of "neutron drip" in Fig. 1(b), then the capture probability will also be low. Moreover, the spacing of neutron resonances will be so wide that it will be entirely a matter of chance whether one resonance does or does not lie in the 5-kev interval of thermal energies. Odd-even differences between nuclei will also be important. Nuclei of such low neutron binding energies are here said to lie in "the region of particularities" in Fig. 1(b). We shall not analyze neutron capture quantitatively in this region. It does not contribute much to the build up of superheavy nuclei at the smaller and more critical neutron fluxes.

At the boundary of the region of particuliarities the neutron binding rises to a value $E=B_{n\,\text{crit.}}$ such that neutron resonances begin to come as close as $D\sim5$ kev. We calculate E from the formula given by Blatt and Weisskopf,[20]

$$D=G\exp[-2(aE)^{\frac{1}{2}}]. \tag{44}$$

The numerical values given for G and a by Blatt and Weisskopf have been fitted by us with the formulas

$$\begin{aligned} G&=(0.0652A-1.36)^2 \text{ Mev},\\ a&=(0.0512A+0.45)(\text{Mev})^{-1}. \end{aligned} \tag{45}$$

We found in this way from Eq. (44) the values of $B_{n\,\text{crit}}$ at the boundary of the zone of particularities as listed in Table V.

[18] S. Frankel and N. Metropolis, Phys. Rev. **72**, 914 (1947); U. L. Businaro and S. Gallone, Nuovo cimento **1**, 629 and 1277 (1955); W. J. Swiatecki, Phys. Rev. **104**, 993 (1956). We are indebted to Dr. Gallone and Dr. Swiatecki for kind communication of their results, also to Dr. Swiatecki for the formula used in constructing Fig. 9. See also U. L. Businaro and S. Gallone, Nuovo cimento **5**, 315 (1957) and V. G. Nossoff, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, August, 1955* (United Nations, New York, 1956), Vol. 2, p. 205.

[19] Summarized for example by J. A. Wheeler, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, August, 1955* (United Nations, New York, 1956), Vol. 2, p. 155.

[20] J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952), pp. 371–372.

Above the zone of particularities neutron capture will typically be due to more than one resonance. The cross section can be estimated from the familiar formula

$$\sigma(n,\gamma)\sim(\pi\lambda\!\!\!^{-2}/D)2\pi\Gamma_\gamma\Gamma_n/(\Gamma_\gamma+\Gamma_n). \tag{46}$$

The neutron width may be evaluated in order of magnitude from the expression[21]

$$\Gamma_n\sim 9\times 10^{-5}D\epsilon_{\text{ev}}^{\frac{1}{2}}, \tag{47}$$

where D is the level spacing in the compound nucleus and ϵ is the neutron energy in ev.

About Γ_γ we cite Blatt and Weisskopf,[20] " . . . it is necessary to admit that the theory does not yet make any significant predictions about the absolute magnitude of neutron capture widths." We therefore make the crudest kind of estimate,

$$\Gamma_\gamma\sim\text{constant (excitation)}^p, \tag{48}$$

where the constant and the power p are to be found from observation. If gamma emission were a single-channel process like slow neutron emission, we would think of Γ/D as the quantity with the simple dependence upon energy. However, the given level of the compound nucleus can pass by gamma-ray emission to a great number of lower levels. Consequently the level spacing cancels out of the total radiation width, as it also cancels out of typical sum rules—hence the absence of D from Eq. (48). To evaluate the two constants in (48), we use two observations: (1) Slow neutron resonances have widths typically of the order of 0.1 ev. Consequently (48) becomes

$$\Gamma_\gamma\sim 0.1\text{ ev }(E/7\text{ Mev})^p. \tag{49}$$

(2) At lesser excitations, as at the excitation $B_n\sim 7$ Mev, those transitions contribute the most to Γ_γ which are the strongest and which are therefore in a certain sense *atypical.* Consequently the width Γ_γ for a low-energy transition should be taken for the strongest known transitions, the $E2$ transitions due to collective nuclear vibrations. From Sunyar's analysis of such fast transitions[22] we estimate for the 43.6 kev $E2$ transition in U^{234} a mean life against radiation of $\tau\sim 4.3\times 10^{-7}$ sec and a width $\Gamma_\gamma\sim 1.5\times 10^{-8}$ ev. Inserting these data in Eq. (49), we find for the unknown exponent the value $p\sim 3.2$. We shall adopt† the simpler value $p=3$,

$$\Gamma_\gamma\sim 0.1\text{ ev }(E/7\text{ Mev})^3. \tag{50}$$

When capture of thermal (5 kev) neutrons produces an excitation of $B_n=2$ Mev, for example, Eq. (50) predicts a γ-ray width of 2.3×10^{-3} ev, whereas the power $p=3.2$ would give $\Gamma_\gamma\sim 1.8\times 10^{-3}$ ev. This difference is unimportant compared to the uncertainty of perhaps as much as a factor 10 in Γ_γ. In summary, we arrive at the formula

$$\sigma(5\text{-kev }n,\gamma)\sim 5\times 10^{-24}\text{ cm}^2/[1+(D/16\text{ ev})(7\text{ Mev}/B_n)^3]. \tag{51}$$

For the nucleus $Z=129$, $A=400$ with a calculated neutron binding of 1.64 Mev and level spacing of 5 kev, for example, we estimate $\sigma(5\text{-kev }n,\gamma)\sim 2\times 10^{-28}$ cm². Uptake of a neutron on the average within a time of 10^{-4} sec therefore requires a flux of $\sim 5\times 10^{31}$ neutrons/cm² sec.

In analyzing more fully the competition of beta decay with neutron uptake, we distinguish three outcomes for beta decay of the nucleus (Z,A) (Fig. 10): (1) The residual nucleus is left with an energy less than B_n (which we suppose less than E_f). Then it de-excites radiatively to yield the nucleus $(Z+1, A)$ in its ground state. (2) The residual excitation lies between B_n and E_f. Then neutron emission occurs so much more rapidly than radiation that we assume the nucleus is always transformed to $(Z+1, A-1)$ by "delayed" neutron emission. (3) The residual excitation exceeds E_f. Radiation is still more negligible compared to heavy particle processes. We estimate the relative probability of neutron emission and fission from the crudest kind of activation formula,

$$\lambda_f/\lambda_n\sim\exp[-(E_f-B_n)/(\tfrac{1}{2}\text{ Mev})], \tag{52}$$

TABLE V. Critical Z values as calculated from the semiempirical mass formula. Z_A, charge of β-stable nucleus (so high that the nucleus undergoes spontaneous fission almost at once). Z_{nf}, charge above which B_n exceeds E_f, so that neutron addition causes fission. $Z_{5\text{ kev}}$, charge above which the calculated spacing of neutron resonances is 5 kev or less. Z_{drip}, charge so low, and neutron number so high, that neutrons are no longer bound. The 1.49-Mev fission barrier for $Z=128$, $A=400$ appears at first sight too small to inhibit spontaneous fission, when compared to the 5.4-Mev barrier for $Z=92$, $A=238$. However, the U^{238} nucleus makes about 8×10^{15} yr$\times 3.1\times 10^7$ sec/yr/10^{-22} sec$=10^{45.4}$ collisions with the barrier before penetrating. The extrapolation arguments of reference 2 lead to a penetration exponent proportional to $A^{11/18}E_f^{5/6}$, or $45.4(400/238)^{11/18}(1.49/5.4)^{5/6}=21.3$, corresponding to a half-life not many powers of 10 different from $10^{21.3-22}$ sec$=0.2$ sec. Spontaneous fission of such a nucleus is so slow that it need not be taken into account in the present considerations of element building.

A	300	350	400
Z_A(β-stability)	113.2	128.6	143.4
$Z_{nf}(B_n=E_f)$	106.1	117.2	128.0
$Z_{5\text{ kev}}$ ($D=5$ kev)	97.9	113.6	128.9
Z_{drip} ($B_n=0$)	91.2	105.2	119.2
$B_n=E_f$ at Z_{nf}	3.52 Mev	2.39 Mev	1.49 Mev
B_n at $Z_{5\text{ kev}}$	1.95 Mev	1.78 Mev	1.64 Mev

[21] See for example the discussion by E. P. Wigner, Am. J. Phys. **17**, 99 (1949), leading to the formula $\Gamma_n\sim 4.4\times 10^{-4}fD\epsilon^{\frac{1}{2}}$, and the analysis of E. Vogt, Ph.D. thesis, Princeton University, 1955 (unpublished) and "The widths and spacings of nuclear resonance lines," Nuclear Development Associates Report NDA-14, April, 1955 (unpublished), where $f\sim 0.2$ is concluded to represent a reasonable average value.

[22] A. W. Sunyar, Phys. Rev. **98**, 653 (1955). The importance of nuclear spins, odd-even differences, and shell effects in a more detailed treatment of total nuclear radiation widths is stressed by A. G. W. Cameron [Can. J. Phys. **35**, 666 (1957)].

† *Note added in proof.*—See A. Stolovy and J. A. Harvey, [Phys. Rev. **108**, 353 (1957)], and references cited therein for alternative empirical formulas for the total gamma-ray width, designed to cover a much smaller range of nuclear excitation but to allow for shell effects.

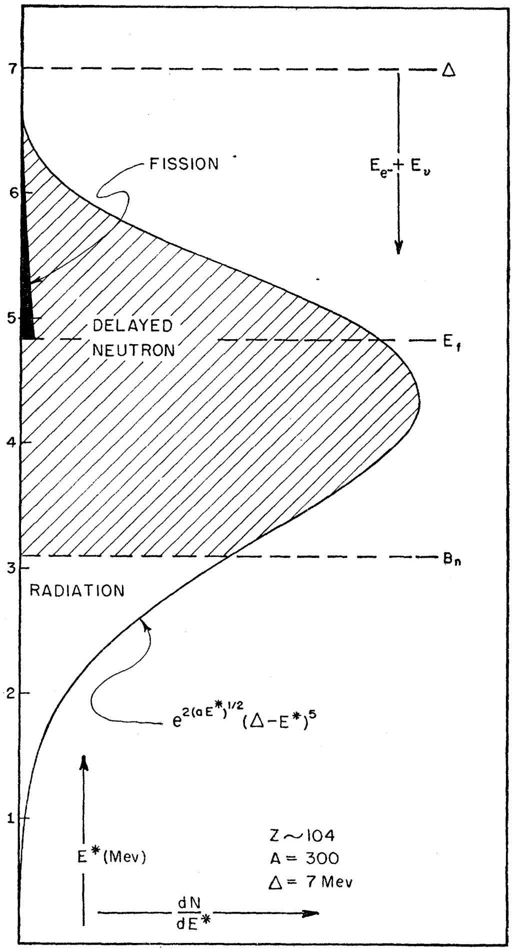


FIG. 10. Competition between simple beta decay to $(Z+1, A)$, "delayed" neutron emission, and fission as affected by the excitation E^* of the residual nucleus. Here $\Delta = c^2[M(Z,A) - M(Z+1, A)]$. B_n is neutron binding energy and E_f is the fission barrier height.

where $\frac{1}{2}$ Mev is a round number adopted for the effective temperature. The probability of any given excitation E^* of the residual nucleus is taken to be proportional to the expression

$$(\Delta - E^*)^5 \exp[2(aE^*)^{\frac{1}{2}}] \tag{53}$$

as in the theory of delayed neutron emission.[23] The branching ratios φ_β, φ_n, and φ_f for simple beta decay and "delayed" neutron emission and fission are represented by the respective fractions of the excitation spectrum shown in Fig. 10 and are calculated from the formulas

$$\varphi_s = \frac{\int_0^\Delta w_s(E^*)(\Delta - E^*)^5 \exp[2(aE^*)^{\frac{1}{2}}]dE}{\int_0^\Delta (\Delta - E^*)^5 \exp[2(aE^*)^{\frac{1}{2}}]dE},$$

[23] N. Bohr and J. A. Wheeler, Phys. Rev. 56, 426 (1939).

where

$$w_\beta(E^*) = \begin{cases} 1 & \text{for} \quad E^* < B_n (< E_f) \\ 0 & \text{for} \quad E^* > B_n, \end{cases}$$

$$w_n(E^*) = \begin{cases} 0 & \text{for} \quad E^* < B_n \\ 1 & \text{for} \quad B_n < E^* < E_f \\ \{1 + \exp[-(E_f - B_n)/(\frac{1}{2}\text{ Mev})]\}^{-1} & \text{for} \quad E^* > E_f, \end{cases}$$

$$w_f(E^*) = \begin{cases} 0 & \text{for} \quad E^* < E_f \\ \{1 + \exp[(E_f - B_n)/(\frac{1}{2}\text{ Mev})]\}^{-1} & \text{for} \quad E^* > E_f. \end{cases} \tag{54}$$

Typical evaluations of the branching ratios are summarized in Table VI. This analysis of the elementary processes in heavy-element building permits rough predictions of the outcome of exposure of uranium, for example, to a constant flux of Q neutrons per cm^2 second for a time t. The transport equation has the form

$$dn_{Z,A}/dt = Q \times (\sigma n)_{Z,A-1} + (\lambda_\beta \varphi_\beta n)_{Z-1,A} + (\lambda_\beta \varphi_n n)_{Z-1,A+1} - Q \times (\sigma n)_{Z,A} - (\lambda_\beta n)_{Z,A}. \tag{55}$$

We define the averages

$$\bar{Z}(t) = \sum_{Z,A} Z n_{Z,A}/n, \quad \bar{A}(t) = \sum_{Z,A} A n_{Z,A}/n,$$

where

$$n(t) = \sum_{Z,A} n_{Z,A}. \tag{56}$$

The transport equation allows a calculation of the time rates of change of $\bar{Z}$, $\bar{A}$, and n. In the approximation where the spread of the statistical distribution of nuclei

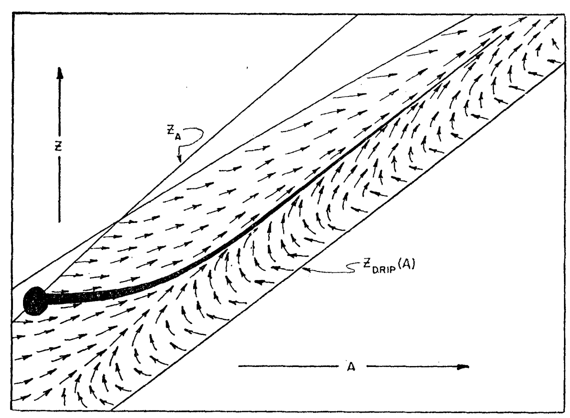


FIG. 11. Qualitative sketch of flow of representative point $(\bar{Z},\bar{A})$ in the (Z,A) diagram of Fig. 1(b) during massive neutron bombardment. The upward component of the flow is due to beta decay proceeding at its natural rate which becomes less as the point moves up towards the line of beta stability. The horizontal component towards smaller A values is due to emission of "delayed" neutrons from nuclei left excited in beta decay. This "backsliding" is greatest near the neutron drip line. The horizontal component of the flow towards greater A values is due to radiative neutron capture. It is least near the neutron drip line where the cross section drops off. Attrition due to fission of nuclei left excited in beta decay is least near the neutron drip line. Nuclei which start as U[238] would tend to follow a path represented by the heavy line whose diminishing width represents the diminishing portion of these nuclei left after fission attrition.

TABLE VI. Branching ratios for simple beta decay and for "delayed" neutron emission and fission calculated from Eq. (54). Nonintegral values of Z appear only because integral values for Δ made the numerical calculation easier to arrange. The last two columns give the beta-decay constant λ_β from Eq. (41) and $\sigma(n,\gamma)$ from Eq. (51).

A	Z	B_n (Mev)	E_f (Mev)	Δ (Mev)	φ_β	φ_n	φ_f	$\lambda_\beta(\mathrm{sec}^{-1})$	$\sigma(n,\gamma)(10^{-24}\ \mathrm{cm}^2)$
300	105.6	3.4	3.9	6.0	0.45	0.47	0.082	100	6.1×10^{-2}
	104.4	3.1	4.8	7.0	0.15	0.84	0.0079	220	2.5×10^{-2}
	103.2	2.8	5.9	8.0	0.037	0.96	0.00031	430	9.4×10^{-3}
	102.0	2.5	7.1	9.0	0.0082	0.99	1.0×10^{-5}	760	3.4×10^{-3}
350	117.3	2.4	2.3	8.0	0.010	0.45	0.54	530	4.4×10^{-3}
	114.6	1.9	3.9	10.0	0.00034	0.85	0.15	1600	4.3×10^{-4}
400	127.9	1.5	1.5	10.0	3.9×10^{-5}	0.53	0.47	1900	8.1×10^{-5}

is treated as small compared to the ranges of Z and A over which σ, φ_s, and λ_β vary strongly, these rates take the form

$$d\bar{Z}/dt=(\varphi_\beta+\varphi_n)\lambda_\beta,\quad d\bar{A}/dt=Q\sigma-\varphi_n\lambda_\beta,\quad dn/dt=-\varphi_f\lambda_\beta n, \tag{57}$$

where the right-hand side is evaluated at the "center of gravity" $(\bar{Z},\bar{A})$ of the cluster of transforming nuclei.

Equation (57) makes it clear that a critical neutron flux is required for building superheavy nuclei. The process of delayed neutron emission lowers $\bar{A}$ at a rate $\varphi_n\lambda_\beta$ which, according to Table VI, is typically of the order of $10^3\ \mathrm{sec}^{-1}$. Merely to balance this backsliding tendency, a flux is needed of the order

$$Q_{\mathrm{crit.}}\sim10^3\ \mathrm{sec}^{-1}/\sigma\sim2\times10^{29}/\mathrm{cm}^2\ \mathrm{sec}. \tag{58}$$

Such a flux is not adequate for buildup. $\bar{A}$ stays constant but $\bar{Z}$ increases, Eq. (57). The fission threshold soon drops below B_n. The nuclei are all destroyed by fission. To avoid this fate, the nuclear cluster in its motion in the (Z,A) diagram (Fig. 11) must stay below the line $Z=Z_{nf}(A)$ where E_f drops to equality with B_n. The slope of this line (Table V) is of the order $dZ_{nf}/dA=0.22$. To this we equate the slope of the flow in the (Z,A) diagram and find a formula for the minimum flux needed for element building:

$$0.22=d\bar{Z}/d\bar{A}=(\varphi_\beta+\varphi_n)/(\lambda_\beta^{-1}\sigma Q_{\mathrm{build}}-\varphi_n). \tag{59}$$

For example, for $A=350$, $Z=114.6$ (Table VI) we find

$$Q_{\mathrm{build}}\sim2\times10^{31}/\mathrm{cm}^2\ \mathrm{sec}. \tag{60}$$

A still higher flux will increase $\bar{A}$ even more than five times as fast as $\bar{Z}$. The representative point will approach the line of particularities. The cross section will diminish. Despite this drop in $\sigma(n,\gamma)$, a sufficiently high flux will carry the representative across the line of particularities. Then it will move along in the direction of increasing A nearly parallel to the neutron drip line, $Z=Z_{\mathrm{drip}}(A)$, but a little above this line. Element building is guaranteed to the extent that our extrapolations are valid. Moreover, this building procedure will take place in a strip of the (Z,A) diagram where the fission branching ratio is least and the attrition is smallest (Fig. 11).

For a quantitative estimate of the yield under the most favorable circumstances of high neutron flux, so that transport occurs near the neutron drip line, we find it most convenient to speak of the fractional loss, L, from the nuclear cluster per unit increase in the average nuclear charge:

$$L=-dn/nd\bar{Z}=\varphi_f/(1-\varphi_f). \tag{61}$$

Along the optimum buildup path, the neutron drip line, this quantity is estimated to have the following values (Table VII). From this quantity we calculate the fraction F of the nuclei not destroyed by "delayed" fission after buildup from a charge $Z_{1\ \mathrm{drip}}$ to a charge $Z_{2\ \mathrm{drip}}$:

$$F=\exp\left(-\int_{Z_1}^{Z_2}LdZ_{\mathrm{drip}}\right). \tag{62}$$

Table VII indicates that there is a reasonable yield of nuclei of mass number $A=350$ under sufficiently intense neutron irradiation, and a small but still quite appreciable yield of nuclei of mass numbers as high as $A=500$.

Fission as neutron source gives a borderline flux. Sudden fission of an indefinitely large mass of uranium of density 19 g/cm³ will produce a neutron density of only $(2.5n/U)(6.02\times10^{23}U/\mathrm{Avogadro})$ [(19/238) Avogadro/cm³]$=1.2\times10^{23}n/\mathrm{cm}^3$. Assuming a velocity of 10^8 cm/sec, we calculate a flux of $10^{31}n/\mathrm{cm}^2$ sec. The presence of transplutonium elements in the November 1, 1952 thermonuclear test debris has been observed and discussed.[24]

TABLE VII. Attrition by "delayed" fission in building up very heavy elements by massive neutron irradiation along the optimum path—the neutron drip line.

A	300	350	400	500
Z_{drip}	91	105	119	147
E_f (Mev)	20.3	16.0	8.4	0.7
Δ (Mev)	18.4	17.4	15.9	13.2
φ_f	0	4.3×10^{-15}	4.9×10^{-8}	0.2
L	0	4.3×10^{-15}	4.9×10^{-8}	0.25
$\int LdZ_{\mathrm{drip}}$	0	$<5\times10^{-14}$	$<7\times10^{-7}$	<3
F	1	$>1-5\times10^{-14}$	$>1-7\times10^{-7}$	>0.05

[24] Fields, Studier, Diamond, Mech, Inghram, Pyle, Stevens, Fried, Manning, Ghiorso, Thompson, Higgins, and Seaborg, Phys. Rev. **102**, 180 (1956).

Mechanisms for the production of still higher fluxes in stars have been discussed. It has been pointed out that matter at the center of a sufficiently massive and cool star can be compressed to such a high density that the electrons are squeezed onto the nuclei and a neutron core is formed.[25] Formation of heavy elements by boil-off of pieces from such a core and their subsequent beta decay has been discussed.[26] The 55-night half-life of the light curves of Type I supernovae and its identity with the half-life of Cf^{254} has been argued as evidence for the production in stars "on a fast time scale of heavy elements by neutron capture processes."[27] Buildup of the known heavy elements in stars by such irradiation at fluxes of the order of 10^{32} neutrons/cm^2 sec has been considered, and found to account for many of the features of the observed abundance curve.[28] In a star one can achieve fluxes much higher than on earth, not of course by neutron velocities substantially in excess of 10^8 cm/sec, but by densities far higher than the $10^{23}n$/cm^3 coming from complete fission of substances obtainable on earth. Therefore there seems to be no difficulty on this score in considering the production of superheavy nuclei.

The real issue is the stability of superheavy nuclei: How far can one trust the extrapolations of the semi-empirical mass formula which form the basis of this paper? On this score it will be surprising if future experiments do not bring future surprises.

ACKNOWLEDGMENTS

Gratitude is here expressed to R. N. Euwema for many helpful discussions and his constant encouragement during this work. We also wish to thank Francis Porzel, David Hill, and Thomas Shanley for their participation with us about 1948 in preliminary calculations like those summarized in Fig. 1(a).

APPENDIX. FIRST-ORDER VACUUM POLARIZATION BY A FINITE NUCLEUS

Wichmann and Kroll[29] have considered the charge and potential induced by a pure Coulomb field to the third order in the charge, Ze, that induces this polarization. The first-order vacuum polarization had been calculated by Uehling, the second-order effect vanishes identically, and the level shift due to the third-order vacuum polarization for a charge as great as $Z=95$ is only 6% of the first-order term, according to Wichmann and Kroll. Accordingly, we neglect all terms of order higher than the first in the *strength* of the field. The *wave number* of the field is also assumed small in Uehling's original analysis, for only low wave numbers affect the energy of an electron in an orbit of substantial size. However, when the charge is high, the electron is pulled in to distances where high wave numbers contribute or where the space dependence of the polarization potential has to be analyzed more closely. Kroll and Wichmann have made such a detailed analysis for the case of a *point* nucleus. In our case the finite size of the nucleus is all important. Therefore we have determined the polarization potential for a finite nucleus to first order in the field strength.

We treat the charge, Ze, as spread uniformly over a sphere of radius $R=r_0A^{\frac{1}{3}}$. Here $r=r^*(\hbar/mc)$ and $R=R^*(\hbar/mc)$ are position and radius. We represent the electric charge in the form

$$\rho_{\text{driving}}(r)=\begin{cases}3Ze/4\pi R^3, & (r<R)\\ 0\quad, & (r>R)\end{cases}$$

$$=(2\pi)^{-\frac{3}{2}}\int\rho_{\text{driving}}(k)\exp(ikr)d^3k, \quad \text{(A1)}$$

where

$$\rho_{\text{driving}}(k)=(3Ze/4\pi R^3)(2\pi)^{-\frac{3}{2}}(4\pi/k^3)\times(\sin kR-kR\cos kR). \quad \text{(A2)}$$

The induced polarization charge is obtained by multiplication with the polarization coefficient,[30] $-h_1(k)$,

$$\rho_{\text{driven}}(k)=-h_1(k)\rho_{\text{driving}}(k), \quad \text{(A3)}$$

$$h_1(k)=(e^2/\pi\hbar c)\int_0^1 x^2(1-x^2/3)\times\left(x^2-1-\frac{4m^2c^2}{\hbar^2k^2}\right)^{-1}dx.$$

The substitution,

$$x\equiv(1-y^{-2})^{\frac{1}{2}},$$

leads to the alternative expression,

$$h_1(k)=(e^2/\pi\hbar c)(-\tfrac{1}{3})\int_{y=1}^{\infty}(2-y^{-2}-y^{-4})\times\left(1+\frac{4y^2m^2c^2}{\hbar^2k^2}\right)^{-1}(y^2-1)^{-\frac{1}{2}}dy. \quad \text{(A4)}$$

This charge creates a supplementary potential under the action of which the potential energy, $V(r)$, of an atomic electron is changed by the amount

$$\delta V(r)=(2\pi)^{-\frac{3}{2}}\int_0^{\infty}(4\pi e/k^2)h_1(k)\rho_{\text{driving}}(k)\times(\sin kr/kr)(4\pi k^2dk). \quad \text{(A5)}$$

[25] L. Landau, Physik. Z. Sowjetunion **1**, 285 (1932); J. R. Oppenheimer and R. Serber, Phys. Rev. **54**, 540 (1938); J. R. Oppenheimer and G. M. Volkoff, Phys. Rev. **55**, 374 (1939).

[26] M. Mayer and E. Teller, Phys. Rev. **76**, 1226 (1949); Peierls, Singuri, and Wroe, Phys. Rev. **87**, 46 (1952).

[27] Burbidge, Hoyle, Burbidge, Christy, and Fowler, Phys. Rev. **103**, 1145 (1956).

[28] Fowler, Hoyle, Burbidge, and Burbidge, Science **125**, 747 (1957). See also P. Fong, Bull. Am. Phys. Soc. Ser. II, **2**, 15 (1957).

[29] E. H. Wichmann and N. M. Kroll, Phys. Rev. **101**, 843 (1956).

[30] First derived by W. Pauli and M. E. Rose, Phys. Rev. **49**, 462 (1936); for a dispersion theoretic derivation see G. Källèn and A. Sabry, Kgl. Danske Videnskab. Selskab. Mat.-fys. Medd. **B29**, No. 17 (1955) or R. N. Euwema and J. A. Wheeler, Phys. Rev. **103**, 803 (1956).

Into this expression we substituted the integral formulas (4) for the polarization coefficient and (2) for the driving charge and found the formula

$$\delta V(r) = -(mc^2)(3Z/4\pi R^3)(e^2/\hbar c)^2(\hbar/mcr) \times (4/3\pi)\int_{y=1}^{\infty} dy\,(y^2-1)^{-\frac{1}{2}}(2-y^{-2}-y^{-4}) \times \int_{-\infty}^{\infty} (\sin kR - kR\cos kR)k^{-2} \times (k^2+4y^2m^2c^2/\hbar^2)^{-1}\sin kr\,dk. \quad \text{(A6)}$$

The integral over wave numbers was performed by the method of evaluation of residues, and gave the result

$$(\hbar/mc)^3(\pi/4y^2) \times \begin{cases} [r^*-(2^{-1}y^{-1}+R^*)e^{-2yR^*}\sinh(2yr^*)], & r^*\leqslant R^* \\ [R^*\cosh(2yR^*)-2^{-1}y^{-1}\sinh(2yR^*)]e^{-2yr^*}, & r^*\geqslant R^*, \end{cases} \quad \text{(A7)}$$

where r^* and R^* are the position coordinate and radius in units $\hbar/mc$. Inserting this expression into (6) and integrating with respect to y, we found the following expression for the electronic potential energy due to polarization:

$$\delta V(r) = -(mc^2)(3Z/4\pi R^{*3})(e^2/\hbar c)^2(12r^*)^{-1} \times \begin{cases} \left\{\begin{array}{l} 4r^*M_6(0)-M_7[2(R^*-r^*)] \\ +M_7[2(R^*+r^*)]-2R^*M_6[2(R^*-r^*)] \\ \quad +2R^*M_6[2(R^*+r^*)] \end{array}\right\} & \text{for } r^*\leqslant R^* \\ \left\{\begin{array}{l} -M_7[2(r^*-R^*)]+M_7[2(r^*+R^*)] \\ +2R^*M_6[2(r^*-R^*)]+2R^*M_6[2(r^*+R^*)] \end{array}\right\} & \text{for } r^*\geqslant R^*. \end{cases} \quad \text{(A8)}$$

In the limit where the nuclear radius goes to zero, this expression goes over to the form

$$\delta V(r) = -(mc^2)(3Z/4\pi)(e^2/\hbar c)^2(4/9r^*)M_4(2r^*) \sim -(mc^2)Z(e^2/\hbar c)^2(2/\pi)^{\frac{1}{2}}(2r^*)^{-\frac{5}{2}}\exp(-2r^*), \quad \text{for large } r^*. \quad \text{(A9)}$$

We have not seen the polarization potential for a point charge expressed as it is here in terms of tabulated functions. Uehling gives $\delta V(r)$ as a double integral. In (8) and (9) we have used the abbreviation

$$M_m(a) = 2\,\mathrm{Ki}_{m-4}(a)-\mathrm{Ki}_{m-2}(a)-\mathrm{Ki}_m(a) \quad \text{(A10)}$$

for the combination of integrals of the form

$$\mathrm{Ki}_n(a) = \int_0^{\infty} \frac{\exp(-a\cosh\theta)}{\cosh^n\theta}d\theta = \int_1^{\infty} y^{-n}e^{-ay}(y^2-1)^{-\frac{1}{2}}dy, \quad \text{(A11)}$$

$\mathrm{Ki}_0(a) = K_0(a)$ (standard Bessel function).

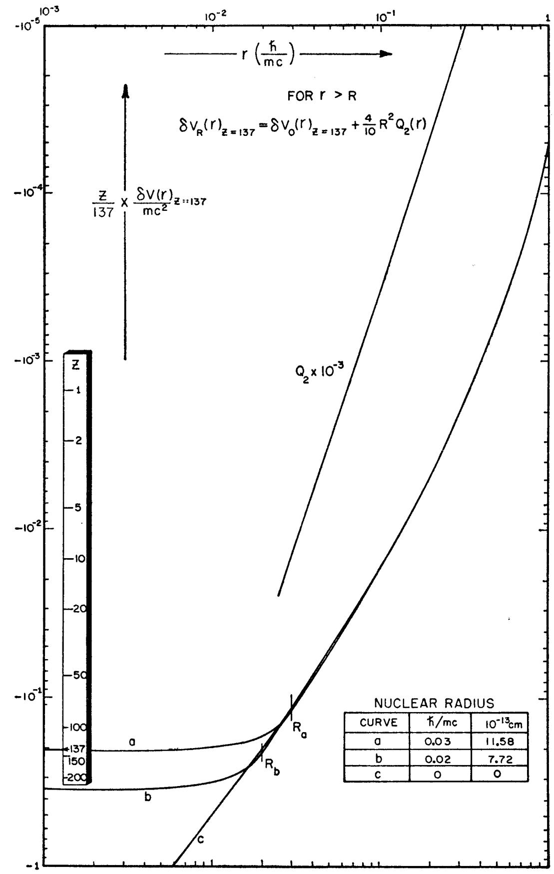


FIG. 12. First-order vacuum polarization contribution to electronic potential energy for two uniform finite spherical charge distributions and for point charge, all for charge $Z=137$. Since these first-order potentials are linear in the nuclear charge they can be scaled to any other nuclear charge. If the arrow at 137 on the scale ruler is carried along the appropriate curve, the division mark for the desired charge Z will trace out the curve for this charge and that radius.

These integrals have been tabulated[31] for values of a from $a=0.05$ at intervals of 0.05 up to $a=0.2$ and intervals of 0.1 from there up to $a=2$. Values of these integrals for smaller a we computed by integration of the appropriate series.

Figure 12 shows the contribution of polarization to the electronic potential energy for the cases $Z=137$, $R=0.02$, $0.03\hbar/mc=7.7$, 11.6×10^{-13} cm$=0.912$, $1.37\times10^{-13}(600)^{\frac{1}{3}}$ cm; and also, for comparison, the corresponding curve for the first order effect of a point nucleus. Both first-order potentials are linear in the nuclear charge and can therefore be scaled to any other nucleus, for two special values of the nuclear radius: $R=0$, 0.02, and 0.03 $\hbar/mc$. In this way one can estimate without tedious computation the order of magnitude of the polarization potential for any Z and any R that are likely to be of interest.

The energy shift due to vacuum polarization was

[31] Bickley and Nayler, Phil. Mag. **20**, 343 (1935).

calculated from the formula

$$\delta E_{\text{vac. pol.}}=\int \delta V(r)(F^2+G^2)dr \Big/ \int (F^2+G^2)dr. \quad \text{(A12)}$$

Here $F(r)$ and $G(r)$ are the two radial components of the K-electron wave function as obtained by numerical integration for $Z=170$, $R=11.3\times10^{-13}$ cm. In this way we obtained the value

$$\delta E_{\text{vac. pol.}}=-0.0185\ mc^2, \quad \text{(A13)}$$

as reported in the text.

PHYSICAL REVIEW VOLUME 109, NUMBER 1 JANUARY 1, 1958

Capture-to-Fission Ratios for Fast Neutrons in U^{235}†

B. C. DIVEN, J. TERRELL, AND A. HEMMENDINGER
Los Alamos Scientific Laboratory, University of California, Los Alamos, New Mexico
(Received September 13, 1957)

The ratio $\alpha=\sigma_c/\sigma_f$, where σ_c is the neutron capture cross section and σ_f the neutron-induced fission cross section, has been measured for U^{235} as a function of neutron energy. A pulsed and collimated neutron beam is passed through a U^{235} sample at the center of a large liquid scintillator. Captures and fissions are detected by means of their prompt gamma rays; elastic and inelastic scattering events are ignored because of smaller pulse heights. Fissions are distinguished from captures by means of delayed pulses from the capture of thermalized fission neutrons. It is found that in the neutron energy range $E_n=0.175$ to 1.0 Mev the value of α is given approximately by $\alpha=0.190$–$0.116E_n$. The accuracy of the determination of α is 10 to 15% in terms of the standard deviation of individual points.

INTRODUCTION

THE simplest and most widely used method of measuring neutron capture cross sections is activation, in which a radioactive end product is detected. For many nuclides, however, no radioactivity is produced. For capture reactions leading to stable or long-lived products, cross sections can be measured by the use of a mass spectrometer for determination of the product nuclides; this method usually requires the high fluxes present in reactors and does not seem to be suitable for fast monoenergetic neutrons. In addition to these methods involving detection of the end product of capture, two other basic methods are available. One is measurement of the change in neutron flux produced by capture, used, for example, in pile oscillator and reactivity measurements. The other basic method, used in the work reported here, is detection of the gamma rays emitted as a result of the capture process. For capture measurements with fast monoenergetic neutrons, this is in many cases the only practical method.

In the present experiment, neutron capture in U^{235} is detected by counting the capture gamma radiation in a large liquid scintillator[1] surrounding the sample; the cross section is determined by comparison with the fission rate of the same sample. Ideally the scintillator should be large enough to absorb all the energy of the gamma radiation emitted at its center. In this case, capture gamma rays would produce a pulse corresponding to the sum of the energies of the gamma rays, which is equal to the binding energy of a neutron in U^{236} (6.29 Mev) plus the kinetic energy of the incident neutron. Fission is also accompanied by prompt gamma emission and the total energy of fission gamma rays is nearly the same as the total energy of capture gamma rays.[2–4] Consequently, observation of the gamma pulses alone is not sufficient to distinguish between capture and fission. The prompt neutrons emitted in the fission process produce delayed pulses which enable us to identify a fission event. The scintillator is large enough to cause most of the fission neutrons to be thermalized and finally captured in the liquid. The addition of cadmium salt to the solution ensures that most neutrons will be captured in cadmium, and the resulting 9 Mev of gamma radiation provides an ample pulse for observation. The cadmium concentration is adjusted so that the mean life of neutrons in the solution is about 40 μsec. The pulses due to fission neutrons are then spread out in time so that they may be counted individually with almost negligible losses due to resolving time. The neutron beam is pulsed so that the neutrons which cause capture or fission in the sample arrive in bursts of 0.1-μsec duration every 100 μsec. Pulses in the scintillator which are caused by prompt gamma rays due to capture or fission are

† Work performed under the auspices of the U. S. Atomic Energy Commission.

[1] Liquid scintillator technique is described by Reines, Cowan, Harrison, and Carter, Rev. Sci. Instr. **25**, 1061 (1954).

[2] I. Francis and R. Gamble and also F. Maienschein *et al.*, Oak Ridge National Laboratory Report ORNL-1879, October, 1955 (unpublished).

[3] Smith, Fields, and Friedman, Phys. Rev. **104**, 699 (1956).

[4] Kinsey, Hanna, and Van Patter, Can. J. Research **26**, 79 (1948).