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On the Division of Nuclear Charge in Fission*

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The theory of the most probable charge number of a fission fragment of given mass number is examined. Two hypotheses previously suggested are (1) that the charges divide in the same ratio as the masses and (2) that the most probable charges correspond to minimum energy of two droplets in contact. These hypotheses predict results at variance with each other and with preliminary experiments. In this paper the actual division of charge is calculated for the final configuration of spheres in contact on the basis of a general nuclear model in which the charge distribution in the nucleus is non-uniform. The tendency of the protons to spread outward results in the smaller fragment of an asymmetric fission having a greater proton-neutron ratio than the larger fragment. In the most probable division (mass ratio 2:3) the most probable partners (of odd mass number) should both have chain lengths equal to 3.6. In the case of a 1:2 mass ratio, the probable chain lengths of the light and heavy partners should be 2.5 and 4.1, respectively. Data on the independent fractional yields of particular chain members are found to lie near a smooth (error) curve when plotted with the aid of the theoretical results.

1. INTRODUCTION

THE division of nuclear charge in fission depends on the extent to which the proton and neutron densities and spatial distributions are altered during the fission process.

Feenberg[1] and Wigner[2] independently have studied the effect of electrostatic repulsion of the protons on the nuclear particle density for normal nuclei. The tendency of the protons to spread outward is accompanied by a tendency of the neutrons to follow the protons because of the exclusion principle and well-known properties of nuclear forces. Whether the neutron density at the surface of the nucleus is greater or less than at the center depends on the compressibility of nuclear matter. Wigner has made the "liquid-drop" assumption of incompressibility or uniform total density, and in the case of uranium his method gives the result that the proton-neutron ratio at the surface exceeds the same ratio at the center by 36 percent. Feenberg obtains a lower result (21 percent for uranium) by a more elaborate calculation in which the nuclear compressibility is taken explicitly into account. While in the former method the proton density is 21 percent larger at the surface than at the center, the latter method gives a 49 percent increase in proton density accompanied by a 23 percent increase in neutron density in going to the surface. The associated corrections to the binding energy are small, and the corrections to the nuclear radius are negligible. So far there has appeared to be no way in which to verify the existence of these effects. It is suggested here that there must be a small but possibly observable effect on the nuclear charge distribution of the primary fission fragments and the most probable number of β-decays of a fragment of given mass number.

If the proton density were uniform throughout the nucleus, the charges of the fragments would be in the same ratio as the masses. However, if the proton density is greater at the surface, the smaller fragment of an asymmetric fission must have a greater proton-neutron ratio than the larger fragment. This effect is enhanced in the Wigner model by the decrease in neutron density from center to surface. On the Feenberg model where the neutron density varies in the opposite fashion, the effect is a differential one caused by the proton density varying more steeply than the neutron density. Since the effect depends on the relative shift of the proton spatial distribution with respect to the neutron distribution, the method of Wigner will give somewhat larger results.

A rough estimate of the size of the effect can be made as follows: The critical shape of a dividing uranium nucleus, corresponding to the saddle

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[1] E. Feenberg, Phys. Rev. 59, 593 (1941).

[2] E. Wigner, University of Pennsylvania Bicentennial Conference (1941).

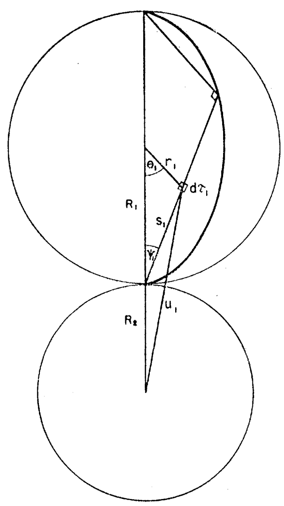

FIG. 1. Diagram of the coordinate system employed.

point of the potential energy *versus* deformation parameter hypersurface, is not very different from a prolate spheroid of eccentricity $\sqrt{3}/2$ with a slight flattening at the equator preliminary to "pinching-in."[3] We assume that the proton and neutron densities vary only with the distance from the center, and use a parabolic fit to the radial dependence of the density computed by Feenberg, the total charge being normalized to that of the undeformed nucleus. The spheroid is sliced into two unequal parts, and the charge on each fragment computed. The calculation is an elementary one and, in view of the more precise calculation to be described in the next section, no details will be given here. The results can be expressed in terms of the charge numbers, Z_1 and Z_2, of the light and heavy fragments and their mass numbers, A_1 and A_2, before neutron emission. Setting γ equal to $[Z_1/(Z_1+Z_2)]/[A_1/(A_1+A_2)]$, one finds that for a 2:3 ratio of the fragment masses, γ is 1.010, and for a 1:2 division, γ is 1.020. Somewhat larger values are obtained if Wigner's densities are used, γ being 1.014 and 1.023 in the two cases, respectively. These results cannot be very accurate in view of (1) our use of the density variation appropriate to the sphere for the critical spheroid and (2) the further elongation of the drop beyond the critical shape accompanied by a further spreading of charge.

[3] R. D. Present, F. Reines, and J. K. Knipp, Phys. Rev. **70**, 557 (1946).

2. ASSUMPTIONS AND FORMULATION

In this section we calculate the charge distribution in the deformed nucleus just before scission into two unequal fragments. This configuration is approximated by two spheres in contact, and the proton and neutron densities are taken to be functions of the distance from the point of contact and of the angle with the axis of symmetry. The nuclear model is that already described by Wigner.[2] We outline the principal assumptions of the calculation:

(A) The charge redistributes itself rapidly during the deformation time for fission so that the charge density has its equilibrium value for every shape of the dividing drop. This follows if the redistribution of the charge is accomplished via the meson field and requires no redistribution of the heavy particles (nucleons).

(B) The total particle density is assumed to be uniform throughout the volume of the deformed nucleus. This is the liquid-drop assumption of incompressibility of nuclear matter.

(C) The decrease of electrostatic energy when the charge goes to the surface is counterpoised by an increase in the "isotopic spin energy." The isotopic spin energy arises from the exclusion principle and the character of nuclear forces and is represented in the mass defect formula by the term or terms in $N-Z$. (The isotopic spin $T_\zeta=(N-Z)/2$.) The mass defect formula proposed by Weizsäcker and employed by Bethe and Bacher, Bohr and Wheeler, and others, contains a term in $(N-Z)^2$. Wigner[4] has shown that the potential energy of the nucleus computed from the symmetrical Hamiltonian contains a term in $(N-Z)/A$ and has included both linear and quadratic terms in $N-Z$ in his mass defect formula. The presence of the linear term considerably reduces the magnitude of the quadratic term. Wigner's mass defect formula is used in the following.

(D) It is assumed that the isotopic spin energy is a "volume energy," i.e., that it can be expressed as an energy density, depending on the local difference in proton and neutron density, integrated over the volume of the nucleus. The numerical coefficient of this energy density is obtained from the empirical coefficients of the

[4] E. Wigner, Phys. Rev. **51**, 947 (1937).

$(N-Z)$ and $(N-Z)^2$ terms in the mass defect formula. Since these coefficients are approximately independent of nuclear size, it is assumed that the same numerical coefficient can be used for the configuration of spheres in contact as for normal separated nuclei.[5]

Figure 1 shows the coordinate system used. Spherical polar coordinates with origin at the point of tangency and polar axis along the axis of symmetry are denoted by s, ψ, and φ; the same coordinates with origin taken at the center of one of the spheres are denoted by r, θ, and φ. R_1 and R_2 are the radii of the two spheres in contact and R the radius of the original nucleus. The ratio R_1/R is denoted by λ_1; hence $\lambda_1^3+\lambda_2^3=1$. Let $\rho_Z(s,\psi)$ be the proton density, $\rho_N(s,\psi)$ the neutron density, and ρ the uniform total particle density. Z, N, and A represent as usual the total numbers of protons, neutrons and nucleons; and Z_1, N_1, and A_1 refer to the numbers in sphere 1. Since the effects to be calculated depend on the difference in proton and neutron densities, we introduce the "local isotopic spin" $t(s,\psi)$ defined by $t=(\rho_N-\rho_Z)/2\rho$. The average values $\bar{\rho}_Z$, $\bar{\rho}_N$, and $\bar{t}$ are the values appropriate to a uniform charge distribution. We have

$$T_\zeta=A\bar{t}=\rho\iiint_{1+2}t\,d\tau=\rho\iiint t(s_1,\psi_1)d\tau_1+\rho\iiint t(s_2,\psi_2)d\tau_2, \quad (1)$$

whence

$$\iiint_{1+2}(t-\bar{t})d\tau=0. \quad (2)$$

It is necessary to evaluate the change in the electrostatic self-energy of each sphere and in the mutual electrostatic energy of the two in going from uniform to non-uniform charge density. Since the deviation from uniform density is small, second-order terms in $\epsilon=\rho_Z-\bar{\rho}_Z=\rho(\bar{t}-t)$ will be neglected. Then the change in the self-energy of each sphere is given by

$$\begin{aligned}\Delta E_i^c&=\frac{e^2}{2}\iiiint\!\!\iint\left\{\frac{\rho_Z(\mathbf{r}_a)\rho_Z(\mathbf{r}_b)-\bar{\rho}_Z^2}{r_{ab}}\right\}d\tau_a d\tau_b \quad i=1,2\\ &=\frac{e^2}{2}\iiiint\!\!\iint\frac{\bar{\rho}_Z\epsilon(\mathbf{r}_a)}{r_{ab}}d\tau_a d\tau_b+\frac{e^2}{2}\iiiint\!\!\iint\frac{\bar{\rho}_Z\epsilon(\mathbf{r}_b)}{r_{ab}}d\tau_a d\tau_b\\ &=e^2\iiiint\!\!\iint\frac{\bar{\rho}_Z\epsilon(\mathbf{r}_a)}{r_{ab}}d\tau_a d\tau_b=e\iiint\epsilon(\mathbf{r}_a)V_0(r_a)d\tau_a,\end{aligned}$$

where $V_0(\mathbf{r}_a)$ is the potential of a uniform distribution. Hence,

$$\Delta E_i^c=\frac{Z_i^{(0)}e^2\rho}{R_i}\iiint\left\{\frac{3}{2}-\frac{1}{2}\frac{r_i^2}{R_i^2}\right\}\cdot(\bar{t}-t_i)d\tau_i. \quad (3)$$

Similarly, the change in the mutual electrostatic energy is

$$\begin{aligned}\Delta E_{12}^c&=e^2\iiint d\tau_1\iiint d\tau_2\left\{\frac{\rho_Z(\mathbf{r}_1)\rho_Z(\mathbf{r}_2)-\bar{\rho}_Z^2}{r_{12}}\right\}\\ &=e^2\iiint d\tau_1\iiint d\tau_2\left\{\frac{\bar{\rho}_Z\epsilon(\mathbf{r}_1)+\bar{\rho}_Z\epsilon(\mathbf{r}_2)}{r_{12}}\right\}\\ &=e\iiint d\tau_1\epsilon(\mathbf{r}_1)V_2^{(0)}(\mathbf{r}_1)+e\iiint d\tau_2\epsilon(\mathbf{r}_2)V_1^{(0)}(\mathbf{r}_2)\\ &=Z_2^{(0)}e^2\rho\iiint\frac{(\bar{t}-t_1)d\tau_1}{u_1}+Z_1^{(0)}e^2\rho\iiint\frac{(\bar{t}-t_2)d\tau_2}{u_2},\end{aligned} \quad (4)$$

[5] A further assumption is that the kinetic energy correction terms arising from the gradients of proton and neutron densities (Weizsäcker terms) can be neglected. Feenberg has included these terms explicitly. However, the effect

where $V_2^{(0)}(\mathbf{r}_1)$ is the potential at $\mathbf{r}_1$ because of a uniform distribution over sphere 2, and $u_1 = (s_1^2+R_2^2+2R_2s_1\cos\psi_1)^{\frac{1}{2}}$ is the distance to $\mathbf{r}_1$ from the center of sphere 2.

We next consider the evaluation of the isotopic spin energy from the mass defect formula. The latter involves the partition quantum numbers $(PP'P'')$ of the ground state, where P is equal to the isotopic spin T_ζ, and P' and P'' are of the order of unity.[2, 4, 6] Neglecting[7] the terms in P' and P'', the mass defect formula is found to contain two terms depending on the isotopic spin of the form: $c_1(N-Z)/A+c_2(N-Z)^2/A$. These correspond to an energy density g_T given by

$$g_T=\frac{c_1\rho}{A}\cdot\left(\frac{N-Z}{A}\right)+\frac{c_2\rho}{A}\cdot\frac{(N-Z)^2}{A}$$

$$=\frac{c_1\rho}{A}\cdot\frac{\rho_N-\rho_Z}{\rho}+c_2\rho\cdot\left(\frac{\rho_N-\rho_Z}{\rho}\right)^2$$

$$=(2c_1\rho/A)t+4c_2\rho t^2. \tag{5}$$

The relation (5) is now assumed to hold when g_T and t are functions of position in the deformed nucleus. The difference in isotopic spin energy between the non-uniform and uniform charge distributions is then given by

$$\Delta E_T=\iiint_{1+2} g_T d\tau-\iiint_{1+2}\bar{g}_T d\tau$$

$$=(2c_1\rho/A)\iiint_{1+2}(t-\bar{t})d\tau+4c_2\rho\iiint_{1+2}(t^2-\bar{t}^2)d\tau$$

$$=4c_2\rho\iiint_{1+2} t(t-\bar{t})d\tau \tag{6}$$

in view of Eq. (2). The term $c_1(N-Z)/A$ makes no contribution to the energy difference. The indirect effect of this term is very considerable, however, since its omission from the mass-defect formula leads to a larger value for c_2. In the Weizsäcker-Bethe formula which contains no term in $(N-Z)/A$, the coefficient c_2 is approximately 20 Mev. The value of c_2 in Wigner's formula is about 12.5 Mev.

The spatial variation of t throughout the two spheres is determined by minimizing the energy difference ΔE between the non-uniform and uniform charge distributions where

$$\Delta E=\Delta E_T+\Delta E_C=\Delta E_T+\Delta E_1{}^C+\Delta E_2{}^C+\Delta E_{12}{}^C. \tag{7}$$

We develop $t(s, \psi)$ in a power series in s and $\cos\psi$ and, because of the smallness of the effect, use only the first terms. Assuming continuity of the densities and their derivatives at the point of contact of the spheres, one omits linear terms. Hence

$$t_i(s_i, \psi_i)=\bar{t}+\delta-\alpha_i(s_i/R)^2+\beta_i(s_i/R)^2\cos\psi_i,\quad i=1, 2. \tag{8}$$

The parameter δ is expressed in terms of α_1, α_2, β_1, and β_2 by means of Eq. (2), and the latter are determined from the minimizing conditions

$$\frac{\partial\Delta E}{\partial\alpha_1}=\frac{\partial\Delta E}{\partial\alpha_2}=\frac{\partial\Delta E}{\partial\beta_1}=\frac{\partial\Delta E}{\partial\beta_2}=0. \tag{9}$$

The charge number of sphere 1 is altered from $Z_1{}^{(0)}=Z\lambda_1{}^3$ for uniform density to Z_1 when the density is not uniform. Thus

$$Z_1-Z\lambda_1{}^3=\iiint_1(\rho_Z-\bar{\rho}_Z)d\tau=\rho\iiint_1(\bar{t}-t_1)d\tau_1 \tag{10}$$

gives the distribution of charge between the fragments. Equations (1) through (10) suffice for the calculation of $\gamma=[Z_1/(Z_1+Z_2)]/[A_1/(A_1+A_2)]$ for any ratio of fragment sizes (provided that α_i and β_i are not too large).

3. PRELIMINARY ESTIMATE

A preliminary estimate of the size of the effects can be simply obtained by replacing Eqs. (8) and (9) by

$$t_i=\bar{t}+\delta_i,\quad \frac{\partial\Delta E}{\partial\delta_i}=0,\quad i=1, 2, \tag{8'–9'}$$

of these terms is partly taken into account when the isotopic spin energy is taken from the mass defect formula with its empirically determined coefficient. One can regard the Weizsäcker terms as expanded in powers of $(N-Z)/A$ and thus partly included in the other terms when empirical coefficients are used.

[6] E. Feenberg and E. Wigner, "Reports on progress in physics," Phys. Soc. London **8**, 274 (1941).

[7] One neglects P', P'^2, and P''^2 compared to P^2. In the case of an even-even nucleus $P'=P''=0$ in the ground state.

corresponding to uniform but different charge densities for the two spheres. Equations (2), (3), (4), and (6) reduce to

$$\delta_1\lambda_1^3+\delta_2\lambda_2^3=0, \tag{11}$$

$$\Delta E_i^c=-(8/5)\pi R^2\cdot Ze^2\rho\cdot\delta_i\lambda_i^5, \tag{12}$$

$$\Delta E_{12}^c=-(4/3)\pi R^2\cdot Ze^2\rho\cdot(\delta_1+\delta_2)\cdot[\lambda_1^3\lambda_2^3/(\lambda_1+\lambda_2)], \tag{13}$$

$$\Delta E_T=4c_2\rho\cdot(4/3)\pi R^3\cdot(\delta_1^2\lambda_1^3+\delta_2^2\lambda_2^3). \tag{14}$$

In the case of the uranium and plutonium isotopes the quantity $Ze^2\rho\cdot 4\pi R^2\cdot A^{-1}$ is slightly less than 50 mMU (milli-mass units). The coefficient $4c_2$ is slightly greater than 50 mMU. In the following we take both quantities equal to 50 mMU. Then,

$$\frac{\Delta E}{100A}=\frac{\delta_1^2\lambda_1^3+\delta_2^2\lambda_2^3}{2}-\frac{\delta_1\lambda_1^5+\delta_2\lambda_2^5}{5}-\frac{\delta_1+\delta_2}{6}\cdot\frac{\lambda_1^3\lambda_2^3}{\lambda_1+\lambda_2} \tag{15}$$

which is to be minimized subject to (11). The charges are then computed from

$$Z_1=Z\lambda_1^3-\delta_1\lambda_1^3A, \tag{16}$$

giving the following results:

$$\begin{aligned} \lambda_1^3:\lambda_2^3&=2:3 & \gamma&=1.0194\\ &=1:2 & &=1.0360\\ &=1:3 & &=1.0607 \end{aligned} \tag{17}$$

For the 1:2 division of uranium $\Delta E=-1.16$ mMU. This simple calculation gives values of γ slightly larger than those obtained from the more accurate calculation described below.

4. DETAILED ESTIMATE

The detailed estimate of the effect is based on the use of (8) to represent the departure from uniform charge distribution. On substituting (8) into (2), (3), (4), and (6) we find

$$\delta=(3/4\pi R^5)\cdot\{(\alpha_1L_1+\alpha_2L_2)-(\beta_1A_1+\beta_2A_2)\}, \tag{18}$$

$$\Delta E_i^c=(Ze^2\rho/2R^3)\cdot\{-(16/5)\pi\delta\lambda_i^5R^5+3\alpha_iL_i\lambda_i^2-\alpha_iN_iR^{-2}-3\beta_iA_i\lambda_i^2+\beta_iD_iR^{-2}\}, \tag{19}$$

$$\Delta E_{12}^c=Ze^2\rho R^2\cdot\{-(8\pi\delta/3)\cdot[\lambda_1^3\lambda_2^3/(\lambda_1+\lambda_2)]+(\alpha_2\lambda_1^3P_2+\alpha_1\lambda_2^3P_1)R^{-4}-(\beta_2\lambda_1^3Q_2+\beta_1\lambda_2^3Q_1)R^{-4}\}, \tag{20}$$

$$\Delta E_T=-4c_2\rho\delta R^{-2}\cdot\{(\alpha_1L_1+\alpha_2L_2)-(\beta_1A_1+\beta_2A_2)\}+4c_2\rho R^{-4}\cdot\{(\alpha_1^2M_1+\alpha_2^2M_2)-2(\alpha_1\beta_1B_1+\alpha_2\beta_2B_2)+(\beta_1^2C_1+\beta_2^2C_2)\}, \tag{21}$$

where

$$\begin{aligned} L_1&=\iiint s_1^2d\tau_1=2\pi(16/15)R_1^5,\\ M_1&=\iiint s_1^4d\tau_1=2\pi(16/7)R_1^7,\\ N_1&=\iiint r_1^2s_1^2d\tau_1=2\pi(24/35)R_1^7,\\ A_1&=\iiint s_1^2\cos\psi_1d\tau_1=2\pi(32/35)R_1^5,\\ B_1&=\iiint s_1^4\cos\psi_1d\tau_1=2\pi(128/63)R_1^7,\\ C_1&=\iiint s_1^4\cos^2\psi_1d\tau_1=2\pi(64/35)R_1^7,\\ D_1&=\iiint r_1^2s_1^2\cos\psi_1d\tau_1=2\pi(544/3^3\cdot 5\cdot 7)R_1^7,\\ P_2&=\iiint(s_2^2/u_2)d\tau_2=2\pi R_1^4K(\lambda), \qquad P_1=2\pi R_2^4K(\lambda^{-1}),\\ Q_2&=\iiint(s_2^2/u_2)\cos\psi_2d\tau_2=2\pi R_1^4W(\lambda), \quad Q_1=2\pi R_2^4W(\lambda^{-1}). \end{aligned} \tag{22}$$

TABLE I. Calculated numerical values.

$\lambda_1^3:\lambda_2^3$	α_1	α_2	δ	$\gamma-1$	$\Delta E/100A$
2:3	0.0364	0.0187	0.0254	0.0158	−.000127
1:2	0.0465	0.0153	0.0244	0.0290	−.000145
1:3	0.0658	0.0122	0.0225	0.0493	−.000169

The integrals $K(\lambda)$ and $W(\lambda)$ with $\lambda=\lambda_1/\lambda_2$ are given by

$$K(\lambda)=\int_0^{2/\lambda}dyy^4\int_{\lambda y/2}^{1}\frac{dx}{(2yx+y^2+1)^{\frac{1}{2}}}$$
$$=4\lambda^{-4}+\frac{32}{5}\lambda^{-5}-\frac{2}{15(1+\lambda)^2}\cdot\left\{\left(\frac{\lambda+2}{\lambda}\right)^3\cdot\left(\frac{6+6\lambda-\lambda^2}{\lambda^2}\right)+1\right\}, \quad (23)$$

$$W(\lambda)=\int_0^{2/\lambda}dyy^4\int_{\lambda y/2}^{1}\frac{xdx}{(2yx+y^2+1)^{\frac{1}{2}}}$$
$$=-\frac{32}{9}\lambda^{-6}-\frac{8}{9}\lambda^{-3}+\frac{2(2-\lambda)m^3}{9(1+\lambda)\lambda^3}+\frac{1+(5\lambda/2)}{48(1+\lambda)^{5/2}}\cdot\{2nm^3-nm-\log(n+m)\}, \quad (24)$$

where $m=(\lambda+2)/\lambda$ and $n=2\lambda^{-1}(1+\lambda)^{\frac{1}{2}}$. The evaluated integrals are substituted into (18), (19), (20), and (21) to give

$$\delta=(8/5)(\alpha_1\lambda_1^5+\alpha_2\lambda_2^5)-(48/35)(\beta_1\lambda_1^5+\beta_2\lambda_2^5), \quad (25)$$

$$\begin{aligned}\Delta E/100A = &-\delta^2/2-(\delta/5)(\lambda_1^5+\lambda_2^5)-(\delta/3)(\lambda_1^3\lambda_2^3/\lambda_1+\lambda_2)+(12/7)(\lambda_1^7\alpha_1^2+\lambda_2^7\alpha_2^2)\\ &+(11/35)(\lambda_1^7\alpha_1+\lambda_2^7\alpha_2)+(1/4)(K(\lambda)\lambda_1^7\alpha_2+K(\lambda^{-1})\lambda_2^7\alpha_1)\\ &-(64/21)(\lambda_1^7\alpha_1\beta_1+\lambda_2^7\alpha_2\beta_2)+(48/35)(\lambda_1^7\beta_1^2+\lambda_2^7\beta_2^2)-(256/3^3\cdot5\cdot7)(\lambda_1^7\beta_1+\lambda_2^7\beta_2)\\ &-(1/4)(W(\lambda)\lambda_1^7\beta_2+W(\lambda^{-1})\lambda_2^7\beta_1). \quad (26)\end{aligned}$$

Numerical values are inserted in (25) and (26) and the minimizing conditions (9) give four simultaneous equations for α_1, α_2, β_1, and β_2. Since the determinants are close to vanishing, greater accuracy is obtained by solving these equations by the method of elimination. When $\lambda_1^3:\lambda_2^3=1:2$ the minimum energy occurs at: $\alpha_1=0.184$, $\alpha_2=0.099$, $\beta_1=0.140$, $\beta_2=0.085$, $\delta=0.0377$, and $\Delta E=-4.7$ mMU for uranium. The signs of the α's and β's correspond to a charge density increasing with distance s from the point of contact and with increasing angle ψ with the symmetry axis. The charges on the fragments are obtained from (10) and (22) which give

$$Z_1-Z\lambda_1^3=-\rho\delta\cdot(4/3)\pi R_1^3+\rho L_1\alpha_1/R^2-\rho A_1\beta_1/R^2$$
$$=(4/3)\pi R^3\rho\cdot\{-\lambda_1^3\delta+(8/5)\lambda_1^5\alpha_1-(48/35)\lambda_1^5\beta_1\}. \quad (27)$$

When $\lambda_1^3=\frac{1}{3}$ we find from the numbers given above that $\gamma=1.029$. This is to be compared with the value of 1.036 of the preliminary estimate. If the angular variation of charge density is neglected (i.e., if $\beta_1=\beta_2=0$), the results are changed to: $\alpha_1=0.0466$, $\alpha_2=0.0153$, $\delta=0.0244$, $\Delta E=-3.4$ mMU, and $\gamma=1.029$. As would be expected, the angular variation has a negligible effect on the division of charge but leads to an appreciable lowering of the energy. For our present purposes it is sufficiently accurate to omit all terms in β_1 and β_2 from the preceding formulae. Figure 2 is a plot of $(\gamma-1)$ *vs.* λ_1^3 obtained in this way, and Table I gives some of the numerical results.

DISCUSSION OF RESULTS

The theoretical results are represented in Fig. 2 which enables one to calculate the division of nuclear charge between fragments of given sizes. For the range of sizes shown, the higher order terms in α_1 and α_2 are negligible, as has previously been assumed.

The principal sources of uncertainty in these results are (1) the assumption of uniform total particle density and (2) the use of Wigner's mass defect formula with the large term in $(N-Z)/A$. In both respects our calculation gives an upper limit for $(\gamma-1)$. Since no definite knowledge of the compressibility of nuclear matter is at hand, the first assumption is reasonable. However, Feenberg's method, in which a roughly estimated value of the compressibility is used, would give smaller results probably closer to the truth. No definite confirmation of Wigner's term in $(N-Z)/A$ for heavy nuclei is yet at hand. The size of this term relative to the conventional term in $(N-Z)^2/A$ is a consequence of the symmetrical Hamiltonian. If this term should prove to be small or absent (as in the Weizsäcker-Bethe formula) the value of $(\gamma-1)$ would be considerably reduced. It is possible but unlikely that the calculated values of $(\gamma-1)$ are too large by as much as 50 percent.

Figure 2 enables one to predict the most probable initial charge of a fission fragment of given mass. We distinguish the primary fission fragment nucleus of mass number A_i from the fission product nucleus of mass number A_i' where $A_i'=A_i-1$. Experimental measurements deal with the product nucleus formed from the fragment nucleus by neutron emission. Since the fraction of fragment nuclei emitting more than one neutron apiece is small, this possibility will be neglected. Figure 3 represents the charge number Z_i of the fragment or of the product nucleus plotted against the mass number A_i' of the product nucleus. The charge numbers $Z_i^{(0)}$ following from the hypothesis of uniform charge distribution $(\gamma=1)$ are shown for comparison. In a very asymmetric division the Z_i and $Z_i^{(0)}$ curves differ by as much as one charge number, the smaller fragment having a higher, and the larger a lower, charge than would be expected if the charges divided in the same ratio as the masses. Still greater differences would be obtained for divisions more asymmetric than those shown in Fig. 3. The dots in Fig. 3 represent stable isotopes of odd mass number. A fairly well defined curve can be drawn through these points; such a curve would be nearly identical with a plot of Z_A *vs.* A in the notation of Bohr and Wheeler. The periodicities of the dots or of the Z_A curve are of course not reflected in the theoretical curve for Z_i (the finer details of nuclear binding represented by the terms in the Hamiltonian which depend on the partition quantum numbers P' and P'' have been neglected). Because of this and also because the theoretical curve gives fractional rather than integral charge numbers, no exact predictions for particular isotopes can be made. Nevertheless, some predictions are possible concerning the average behavior of the number ν of β-decays of a fragment of given mass and most probable charge for that mass. Fragments of the same mass but different charge are formed independently in the fission process, and their yield as primary fission products can be measured in a few instances. It is thus possible to estimate ν for several neighboring odd masses and

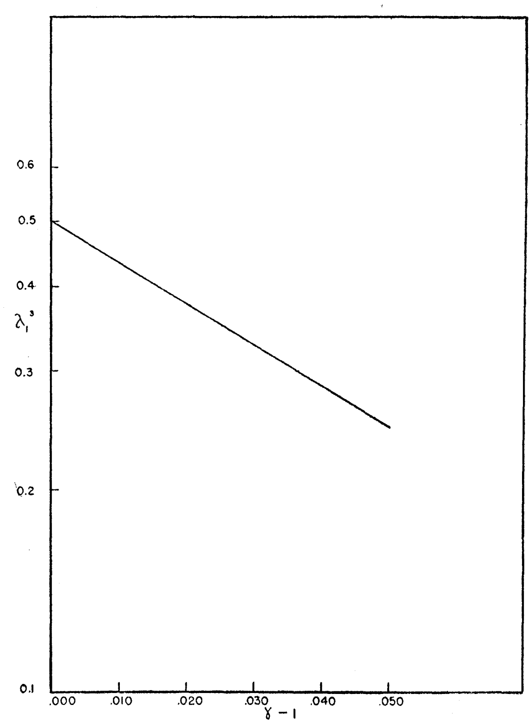

FIG. 2. Calculated curve of $(\gamma-1)$ *vs.* λ_1^3.

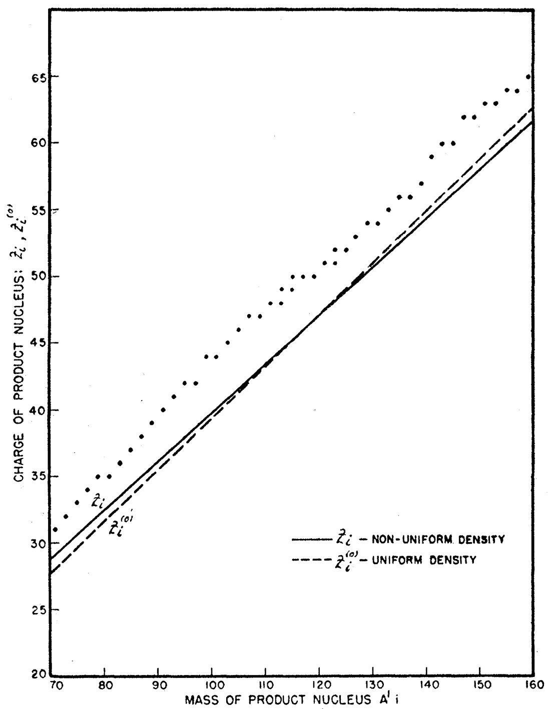

FIG. 3. Charge *vs.* mass of the product nuclei.

compare the result with Fig. 3 where ν is the mean vertical distance from the Z_i curve to the dots. The quantity ν will be referred to as the "probable chain length" for a given mass ratio of the fragments. The following conclusions can then be drawn from the theoretical curve for Z_i:

(1) For that mass ratio which occurs with greatest frequency in slow neutron uranium fission (about 2:3 or 93:141) the probable chain lengths of the complementary fragments are both equal to 3.6.
(2) In the case of a 1:2 or 78:156 division, the probable chain lengths for the light and heavy partners are 2.5 and 4.1, respectively.
(3) In the case of a 1:3 or 58:176 division (not shown in Fig. 3), the values of ν would be 2.4 and 3.5 for the light and heavy partners. Decay chains for such an extremely asymmetric split have not been observed but might be found in very fast neutron fission.

The foregoing conclusions refer to odd mass numbers A_i'. The stable isobars of even mass number have not been included in Fig. 3 because the dots would scatter so widely that the probable chain length ν would undergo large fluctuations from one mass number to the next. The probable chain lengths for the even mass numbers are on the average appreciably smaller than for the odd mass numbers, since the even mass dots lie mostly below a curve through the odd mass dots. Data on even masses will be utilized below.

The energy correction ΔE caused by spreading of the charge is small for all observed modes of division (cf. Table I), and the difference between the values of ΔE for symmetric and the most probable (2:3) asymmetric fission is of the order of one Mev. Hence the considerations of this paper make no appreciable change in fission calculations involving the energy of two spheres in contact or a deformed ellipsoidal nucleus.

Wigner and Way[8] have suggested that the charge might divide in such a way that the total decay energy of the two chains would be a minimum. Extending this idea to take into account the electrostatic repulsion of the fragment nuclei, Way[9] has calculated the division of charge by assuming that the most probable division is that in which the energy of the two product nuclei in contact is a minimum. While this assumption is a reasonable one, it can hardly be expected to be accurate, particularly in view of the fact that the most probable mass ratio of the fragments observed in the division of various fissionable nuclei does not in general correspond to minimum energy of the product nuclei in contact.[9,10] The results obtained by Way, after minimizing the total decay plus electrostatic energy using the Bohr-Wheeler mass defect formula, are nearly the same as ours for the 1:2 mode but are significantly different for the 2:3 mode (the probable chain length for 93 is 3.1 compared to our value of 3.6).

Some confirmation of these results is provided by the chemical investigation of fission product decay chains. It appears from the work of Glendenin, Coryell, and others[11] that the known facts about decay chains, with mass numbers corre-

[8] E. Wigner and K. Way, Report CC-3032 (1945); Plutonium Project Reports **9B**, 6.4 (1946).
[9] K. Way, private communication.
[10] S. Flügge and G. v. Droste, Zeits f. physik. Chemie [B], **42**, 274 (1939).
[11] Summarized in Glendenin, Coryell, Edwards, and Feldman, CL-LEG-No. 1 (1946). Most of the data have been published in the J. Am. Chem. Soc. **68**, 2411 (1946).

sponding to the most likely modes of fission, are consistent with the result of equal probable chain lengths (3.6) for the complementary fragments. Insufficient data are available, however, to draw conclusions about the chain lengths in a 1:2 division. In the case of a few mass numbers, it has been found possible to measure the independent yield of a particular member of the decay chain, formed directly from the fission process. This is possible when the preceding member of the chain has a long half-life or is stable (belonging to a pair of stable isobars). Assume that the total yield of the chain is known. Then the fraction of the chain of a given mass formed with a particular charge is determined in a few instances. If these fractions are plotted against $Z-Z_P$, where Z_P is the most probable charge for a fission fragment of the given mass, it is reasonable to expect the points to lie near a smooth (error) curve symmetrical about $Z=Z_P$. Such a plot has been made by Glendenin, Coryell, Edwards, and Feldman;[11] they find that if Z_P is determined either from unchanged charge distribution ($Z_i^{(0)}$), or from minimum energy of nuclei in contact (Way's method), the resulting points show systematic deviations from a symmetrical error curve, the lighter mass points (Br^{82} and Rb^{86}), and the heavier mass points (Xe^{135} and Cs^{136}) appearing to lie on separate curves displaced by about one unit of charge in both cases. On the other hand, if the values of Z_P are obtained by an empirical postulate of equal probable chain lengths for complementary fragments, all points appear to lie near a smooth curve. We have replotted their data taking Z_P equal to the theoretical value Z_i from Fig. 3. The resulting points lie reasonably close to a single error curve with some scatter but no systematic deviation, i.e., there is no evidence of separate curves for the lighter and heavier fission products. Some scatter is to be expected in view of the neglect of finer details of nuclear binding in the theoretical treatment; however, all the points lie within one-fifth of a charge unit of the best drawn error curve symmetrical about $Z=Z_i$. This can be considered as a preliminary check on the theory; evidently more experimental points, corresponding to a greater range of mass numbers, are needed to provide definitive confirmation.