

for decay to the ground state of C^{12} , is less than 0.1%. This limit is an order of magnitude smaller than the best previous experimental limits. It is not, however, inconsistent with current descriptions of helium burning.

A limit on P can be used as a basis for a choice between the 0^+ and 2^+ assignments if the partial widths for the various decay processes can be estimated with sufficient accuracy. Previous estimates by Rasmussen,¹⁰ CFLL,³ and Salpeter¹ indicate that for a 0^+ state P is probably of the order of 10^{-2} or 10^{-3} , while for a 2^+ state transitions to the ground state should be comparable to, or predominate over, alpha decays. It was therefore concluded by CFLL, from an experimental upper limit on P of about 10^{-2} , that the state is very probably 0^+ .

A 2^+ assignment, however, could not be completely ruled out due to the uncertainties in the theoretical estimates of the decay rates. The present limit makes it still more unlikely that the state is 2^+ and this, together with the angular distribution results, strengthens the 0^+ assignment.

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Hydrodynamic Theory of Spontaneous Fission*

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The penetration factor for spontaneous fission has been calculated from the liquid-drop model. The transformation of the Gamow integral over the nucleon coordinates into an integral over the deformation parameters a_n has been carried out hydrodynamically, assuming irrotational motion. The transformation requires evaluation of the kinetic energy in terms of a_n and \dot{a}_n . Series expansions are used for the kinetic energy and for the potential energy of deformation. We have neglected all parameters but a_2 and carried the hydrodynamic calculations through terms in a_2^4 . While the potential barrier is subject to several uncertainties, it has nevertheless been possible to estimate the spontaneous fission hindrance factor for the highest Z elements. We find for $Z=100$ and $Z^2/A \approx 39$ that a 1-Mev increase in barrier height should correspond to a $10^{8.7}$ -fold increase in the half-life. This result agrees closely with the empirical hindrance factor formula deduced by Swiatecki from a correlation of fluctuations in half-lives with deviations of ground-state masses from the semiempirical mass formula. We have included some details of both the hydrodynamic and the electrostatic calculations.

1. INTRODUCTION

THE successes and limitations of the liquid-drop theory of nuclear fission are well known. In addition to the qualitative explanation of the fission process and the simple calculation of the energy released in fission, the drop model has had reasonable success in predicting approximate activation energies for nuclides, such as the uranium isotopes, which are not close to classical instability.^{1,2} However, the variation of the predicted activation energies with Z^2/A is more rapid than the variation indicated by measured thresholds for photofission and neutron-induced fission. The calculation of the activation energy is a difficult and laborious problem requiring the determination of

the potential energy in terms of the deformation parameters for large deformations. It is not surprising that the drop model should give a good account of these essentially classical aspects of the fission process.

The possible occurrence of spontaneous fission as a quantum-mechanical tunnel effect was first suggested by Bohr and Wheeler,³ who made, however, no attempt at a detailed calculation. In order to evaluate the half-life, the integral appearing in the exponent of the penetration factor, which is a multiple integral over the nucleon coordinates, must be transformed into an integral over the deformation parameters of the drop. The transformation can be carried out if one assumes that the motion of the nucleons during the deformation process can be represented as an irrotational flow of an ideal incompressible fluid along classical streamlines. The first attempt at such a calculation led to unsatisfactory results⁴; a later attempt, while more successful,

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¹ Present, Reines, and Knipp, *Phys. Rev.* **70**, 557 (1946).

² S. Frankel and N. Metropolis, *Phys. Rev.* **72**, 914 (1947).

³ N. Bohr and J. A. Wheeler, *Phys. Rev.* **56**, 426 (1939).

⁴ F. Reines, doctoral thesis, New York University, December 1943 (unpublished); see also reference 1.

was based on an incomplete analysis of the hydrodynamics of the deformation process.²

It may seem an unwarranted extension of the drop model to employ classical hydrodynamics in the treatment of an effect so intrinsically quantum-mechanical as barrier penetration. Justification rests on the circumstance that the exponent of the penetration factor is related directly to the characteristic action function of classical mechanics and can therefore be calculated classically. The further assumption of an irrotational flow is justified by the fact that it leads to the greatest possible penetration factor for a given barrier. Since the potential energy terms of the liquid-drop model determine the barrier to be penetrated, it is then possible in principle to calculate the half-life for spontaneous fission. Because of the sensitivity of the exponential factor to slight variations of the exponent, if the assumptions and approximations were not reasonable ones, the answer could disagree with experiment by many powers of ten. This type of calculation therefore constitutes a severe test of the liquid-drop model.

The present work is based on a more complete analysis of the hydrodynamic problem than has hitherto been given, but is restricted in two essential respects: (1) only one deformation parameter, a_2 , is taken into account (the surface of the drop is the locus $r=R_0[1+\sum a_n P_n(\cos\theta)]$) and (2) the kinetic and potential energies are expanded in powers of a_2 . Both of these are good assumptions if the nuclide in question has a high enough Z to be close to classical instability. Since instability sets in first with respect to second harmonic (P_2) deformations, the potential barrier for such a nuclide will correspond to small values of a_2 and much smaller values of the other a_n . The results will be applied to fermium, which is the highest Z element for which spontaneous fission half-lives have been measured.

2. THE PENETRATION FACTOR

The penetration factor is obtained from the solution of the Schrödinger equation for a system of A nucleons by means of the WKB approximation method. The customary substitution

$$\psi = e^{iS/\hbar} \quad \text{with} \quad S = S_0 + (\hbar/i)S_1 + (\hbar/i)^2 S_2 + \dots \quad (1)$$

gives the successive orders of approximation, and it is a familiar result that S_0 satisfies the Hamilton-Jacobi partial differential equation. The identification of S_0 with the characteristic action function makes it possible to use Jacobi's form of this function for a parameterized motion of the particles: $\mathbf{r}_i = \mathbf{r}_i(\alpha)$ where α is a parameter completely specifying the path of the system point in configuration space. Thus

$$\begin{aligned} S_0 &= \int [2(E-V)]^{\frac{1}{2}} \left[\sum_i m_i (ds_i/d\alpha)^2 \right]^{\frac{1}{2}} d\alpha \\ &= \int [2m^*(E-V)]^{\frac{1}{2}} d\alpha, \end{aligned} \quad (2)$$

where $V(\alpha)$ is the potential energy and $m^*(\alpha)$ is the *effective mass*, which is related to the kinetic energy T by $T = \frac{1}{2}m^*\dot{\alpha}^2$. Since the kinetic energy is a homogeneous quadratic function of the velocities, the effective mass is independent of $\dot{\alpha}$. The exponential part of the first-order WKB wave function is then given by

$$\begin{aligned} \psi &\sim \exp \left\{ \pm \frac{i}{\hbar} \int [2m^*(E-V)]^{\frac{1}{2}} d\alpha \right\}, \quad E > V \\ &\sim \exp \left\{ \pm \frac{1}{\hbar} \int [2m^*(V-E)]^{\frac{1}{2}} d\alpha \right\}, \quad E < V. \end{aligned} \quad (3)$$

In the absence of a theory of barrier-penetration in many dimensions (the WKB connection formulas hold only for one-dimensional problems or three-dimensional problems reducible to one dimension), we proceed by analogy. The one-dimensional WKB functions take on the same form as (3) if one replaces the particle coordinate x by the parameter α and the particle mass m by the effective mass m^* . We assume that the penetration factor for the present case can be obtained by substituting m^* for m and α for x in the formula for the one-dimensional case. The desired formula for the probability of penetrating the barrier is then

$$P = \exp \left\{ -\frac{2}{\hbar} \int_{\alpha_1}^{\alpha_2} [2m^*(V-E)]^{\frac{1}{2}} d\alpha \right\}, \quad (4)$$

where α_1 and α_2 are turning points marking the entrance and exit of the barrier, respectively.

In the application of Eq. (4) to spontaneous fission, the values of the parameter α specify a sequence of shapes for the fissioning droplet. In the hyperspace of the deformation parameters a_n , defined in Sec. 1, the system point follows a path defined by the relations $a_n = a_n(\alpha)$. The barrier to be penetrated corresponds to a potential energy surface in many dimensions, and the largest possible penetration factor is obtained for the functions $a_n(\alpha)$ that characterize the saddle-point path. The potential energy $V(a_n)$ is the sum of the Coulomb and surface energies of deformation, calculated from the liquid-drop model. In order to simplify our calculation, we neglect all deformation parameters except a_2 and set $\alpha = a_2$. The barrier given by $V(a_2)$ vs a_2 is theoretically too large since the saddle-point path is not followed; an adjustment, to be described later, will be made to compensate for this.

Many possible parameterized motions of the nucleons are consistent with any given change in the droplet surface—e.g., with the sequence of shapes obtained by varying a_2 . Of all these possible motions consistent with a prescribed motion of the boundary, there is one motion which minimizes the kinetic energy and this is an irrotational motion of the particles along the classical streamlines (Kelvin's theorem). The smallest value of the effective mass and the largest value of the

penetration factor are obtained by assuming an irrotational flow of the incompressible nuclear fluid. The possibility of a classical calculation of V and of m^* rests, of course, on the fact that these quantities are defined through the classical action function of Eq. (2).

3. HYDRODYNAMIC CALCULATION OF THE EFFECTIVE MASS

Since the effective mass can be obtained directly from the kinetic energy, we shall take up next the classical hydrodynamic calculation of the kinetic energy of the fluid in the deforming droplet. The motions of the fluid particles are described by a velocity field, which is uniquely determined by the condition of irrotational flow and the boundary condition for the free liquid surface. Because of the assumed incompressibility, the velocity field \mathbf{v} is solenoidal. Since \mathbf{v} is also irrotational, we introduce the velocity potential Φ . Thus

$$\begin{aligned}\mathbf{v}(\mathbf{r},t) &= \nabla\Phi(\mathbf{r},t), \\ \nabla^2\Phi &= 0.\end{aligned}\quad (5)$$

The changing surface of the deformed drop can be represented as the locus: $F(\mathbf{r},t)=0$. Since a fluid particle in the surface must remain in the surface

$$\mathbf{v} \cdot \nabla F + \partial F / \partial t = 0, \quad (6)$$

where \mathbf{v} represents the velocity field at the surface. The first term in (6) arises from the motion of the fluid particles and the second from the changing parameters of the surface. The general boundary condition is then

$$\nabla\Phi \cdot \nabla F + \partial F / \partial t = 0. \quad (7)$$

We now assume that the drop has an axis of symmetry along the z axis and represent the radius vector from an origin at the center of the undeformed drop to an arbitrary point on the deformed surface by the series of Legendre polynomials:

$$r = R(\mu, t) \equiv R_0 \left[1 + \sum_{n=0} a_n(t) P_n(\mu) \right], \quad (8)$$

where $\mu = \cos\theta$. Since the velocity potential is a harmonic function, finite at the origin, we have

$$\Phi = \sum_{n=1} b_n(t) r^n P_n(\mu). \quad (9)$$

Inserting $F(\mathbf{r},t) = r - R(\mu, t)$ in Eq. (7), we obtain

$$\frac{\partial\Phi}{\partial r} = \frac{\partial R}{\partial t} + \frac{\partial R}{r\partial\theta} \frac{\partial\Phi}{r\partial\theta}. \quad (10)$$

This simpler form of the boundary condition can be obtained more directly by observing that the radial component of velocity of a surface particle is given by

$$v_r = \frac{\partial\Phi}{\partial r} = \lim_{\delta t \rightarrow 0} \left(\frac{\delta_a R}{\delta t} + \frac{\delta_\theta R}{\delta t} \right), \quad (11)$$

where $\delta_a R$ is the increment in r resulting from the change in $a_n(t)$ and $\delta_\theta R$ is due to the change in the θ coordinate of the particle. Evidently

$$\begin{aligned}\lim_{\delta t \rightarrow 0} \frac{\delta_a R}{\delta t} &= \frac{\partial R}{r\partial\theta} \frac{\partial\theta}{\partial t} = \frac{\partial R}{r\partial\theta} \frac{\partial\Phi}{r\partial\theta}, \\ \lim_{\delta t \rightarrow 0} \frac{\delta_\theta R}{\delta t} &= \frac{\partial R}{\partial t},\end{aligned}\quad (12)$$

leading again to Eq. (10). It is convenient to set $R_0 = 1$ in Eq. (8). Substitution of (8) and (9) into Eq. (10) then gives

$$\begin{aligned}\sum_n n b_n R^{n-1}(\mu) P_n(\mu) - \sum_l \sum_n a_l b_n R^{n-2}(\mu) P_l^1(\mu) P_n^1(\mu) \\ = \sum_l \dot{a}_l P_l(\mu),\end{aligned}\quad (13)$$

where P_l^1 is the associated Legendre function and $\dot{a}_l = da_l/dt$. If we assume the deformation shapes to have reflection symmetry about the equatorial (x, y) plane, then $R(\mu)$ and $\Phi(\mu)$ will be even functions of μ and the sums in Eqs. (8), (9), and (13) will be over the even integers.

Equation (13), which provides the relation between the deformation parameters a_n and the coefficients b_n of the velocity potential, can be solved by successive approximations. If the deformation is specified by certain nonvanishing a_n , Eq. (13) can be used to obtain the corresponding b_n to various orders of approximation in the a_n . Conversely, if the velocity field is specified by certain nonvanishing b_n , Eq. (13) can be used to determine the corresponding deformation parameters a_n . We shall assume in the following that $a_n = 0$ for $n > 2$; thus the deformation shapes are restricted to

$$r = R(\mu) = 1 + a_0 + a_2 P_2(\mu), \quad (14)$$

where a_0 is adjusted to maintain constant volume. If a_2 is small, the condition for this is

$$\begin{aligned}a_0 = -\frac{a_2^2}{5} \left(1 + \frac{2}{3 \times 7} a_2 + 0 \times a_2^2 + \frac{2}{3 \times 5 \times 7} a_2^3 \right. \\ \left. - \frac{127}{3^2 \times 5^2 \times 7^2} a_2^4 + 0 \times a_2^5 - \frac{107}{3^2 \times 5^3 \times 7^2} a_2^6 \dots \right).\end{aligned}\quad (15)$$

Equation (13) becomes

$$\begin{aligned}\sum_{n=2} \{ n b_n R^{n-1}(\mu) P_n(\mu) - a_2 b_n R^{n-2}(\mu) P_2^1(\mu) P_n^1(\mu) \} \\ = \dot{a}_0 + \dot{a}_2 P_2(\mu).\end{aligned}\quad (16)$$

Evidently the b_n 's are linear in \dot{a}_2 and successive approximations for small a_2 correspond to retaining successively higher terms of the form $a_2^m \dot{a}_2$. The zero-order solution of (16) is seen immediately to be

$$b_2 = \dot{a}_2/2, \quad b_n = 0 \quad \text{for } n > 2.$$

The first-order approximation is obtained by substituting

$$b_2 = \frac{1}{2}\dot{a}_2 + \beta_2^{(1)}a_2\dot{a}_2, \quad b_n = \beta_n^{(1)}a_2\dot{a}_2 \quad (n > 2), \quad (17)$$

in Eq. (16), neglecting terms of order $a_2^2\dot{a}_2$ and higher, and expressing all powers of μ in terms of Legendre polynomials. The resulting equation is an identity in which the coefficients of all the P_n are separately equal to zero. It is readily found that

$$\beta_2^{(1)} = \frac{1}{2 \times 7}, \quad \beta_4^{(1)} = -\frac{27}{2 \times 5 \times 7}, \quad \beta_n^{(1)} = 0 \quad (n > 4). \quad (18)$$

The calculation of the higher order approximations is somewhat lengthy and we have not gone beyond fourth-order terms (see Appendix 1). The determination of the b_n to terms in a_2^4 makes it possible to calculate the kinetic energy T also to the same approximation. Since the mass density κ is assumed to be uniform in the liquid-drop model, the kinetic energy can be written as

$$\begin{aligned} T &= \frac{1}{2}\kappa \int v^2 d\tau \\ &= \pi\kappa \int_{-1}^1 d\mu \int_0^{R(\mu)} dr r^2 |\nabla\Phi|^2 \\ &= \pi\kappa \int_{-1}^1 d\mu \int_0^{R(\mu)} dr r^2 \left\{ \left[\sum b_n n r^{n-1} P_n(\mu) \right]^2 \right. \\ &\quad \left. + (1-\mu^2) \left[\sum b_n r^{n-1} \frac{dP_n}{d\mu} \right]^2 \right\} \\ &= \pi\kappa \int_{-1}^1 d\mu \int_0^{R(\mu)} dr r^2 \left\{ \sum_l \sum_n b_l b_n r^{n+l-2} \right. \\ &\quad \left. \times [n l P_l(\mu) P_n(\mu) + P_l^1(\mu) P_n^1(\mu)] \right\} \\ &= \pi\kappa \sum_l \sum_n \frac{b_l b_n}{l+n+1} \int_{-1}^1 d\mu R^{n+l+1}(\mu) \\ &\quad \times [n l P_l(\mu) P_n(\mu) + P_l^1(\mu) P_n^1(\mu)]. \quad (19) \end{aligned}$$

The evaluation of T to first order is very simple since only the terms in b_2^2 and $b_2 b_4$ contribute and the latter term vanishes upon integration. The result to first order is

$$T = (4/5)\pi\kappa b_2^2(1+a_2), \quad (20)$$

and, using (17) and (18),

$$T = \frac{\pi\kappa\dot{a}_2^2}{5} \left(1 + \frac{9}{7}a_2 \right) = \frac{1}{2}m^*\dot{a}_2^2, \quad (21)$$

where m^* is the effective mass. Hence to first order

$$m^* = \frac{3}{10}mA \left(1 + \frac{9}{7}a_2 \right), \quad (22)$$

where A is the mass number and m the mass of one nucleon.⁵ Carrying the calculation of the effective mass to fourth order (see Appendix 1), the following result is obtained:

$$m^* = \frac{3}{10}mA \left(1 + \frac{9}{7}a_2 - \frac{134}{245}a_2^2 - 1.395225a_2^3 - 0.171995a_2^4 \right). \quad (23)$$

This value of $m^*(a_2)$ will be used in calculating the penetration factor.

4. THE SURFACE AND COULOMB ENERGIES OF DEFORMATION

The next step involves the determination of the barrier represented by $V(a_2)$ vs a_2 where $V(a_2)$ is the sum of the surface and Coulomb energies of deformation. Denoting the surface tension by O , the surface energy of the axially symmetric deformed drop is given by

$$U^s = 2\pi O \int_{-1}^1 d\mu r^2 [1 + (1-\mu^2)(dr/r d\mu)^2]^{\frac{1}{2}}. \quad (24)$$

Upon introducing (8) into (24) and expanding the radical, the surface energy can be obtained as a power series in the deformation parameters.

The electrostatic self-energy is

$$U^c = \frac{1}{2} \int \int \frac{\rho_1 d\tau_1 \rho_2 d\tau_2}{r_{12}}, \quad (25)$$

where $\rho_1 = \rho_2 = \rho = Ze/(4\pi R_0^3/3)$. The evaluation of (25) is less straightforward than in the case of (24), and will be discussed in some detail since it has not yet appeared in the literature. We restrict the deformation shapes to prolate figures of rotation possessing reflection symmetry about the equatorial (x, y) plane; thus $R(-\mu) = R(\mu)$. We further assume that any sphere drawn about an origin at the center of the drop, if it intersects the surface, will intersect at only two values of θ , namely θ_0 and $\pi - \theta_0$. Referring to Fig. 1, we see that a sphere of radius $b = R(0)$ is the largest sphere that can be inscribed within the figure of the drop. The electrostatic potential must be calculated differently for points that lie inside and outside of the sphere of radius b . Thus

$$\begin{aligned} U^c &= \frac{1}{2}\rho \int_0^{2\pi} d\phi_1 \int_{-1}^1 d\mu_1 \left[\int_0^b dr_1 r_1^2 V(r_1 < b) \right. \\ &\quad \left. + \int_b^{R(\mu_1)} dr_1 r_1^2 V(r_1 > b) \right], \quad (26) \end{aligned}$$

⁵ Equation (22) is given erroneously in reference 2 as $m^* = (3/10)mA(1+a_2)$.

where $V(r_1 < b)$ is obtained by using the expansion for $1/r_{12}$ in terms of $P_l(\mu_{12})$ where μ_{12} is the cosine of the angle between \mathbf{r}_1 and \mathbf{r}_2 :

$$V(r_1 < b) = \rho \int_0^{2\pi} d\phi_2 \int_{-1}^1 d\mu_2 \times \left[\int_0^{r_1} dr_2 r_2^2 \sum_{l=0}^{\infty} (r_2^l / r_1^{l+1}) P_l(\mu_{12}) + \int_{r_1}^{R(\mu_2)} dr_2 r_2^2 \sum_{l=0}^{\infty} (r_1^l / r_2^{l+1}) P_l(\mu_{12}) \right]. \quad (27)$$

Equation (27) cannot be used for $r_1 > b$ because it is incorrect in the range $\theta_0 < \theta < \pi - \theta_0$ or $\mu_0 > \mu > -\mu_0$ (shaded region of Fig. 1). It is convenient to express $V(r_1 > b)$ as

$$V(r_1 > b) = V(r_1 < b) + \Delta V, \quad \Delta V = \rho \int_0^{2\pi} d\phi_2 \int_{-\mu_0(r_1)}^{\mu_0(r_1)} d\mu_2 \times \left[- \int_{R(\mu_2)}^{r_1} dr_2 r_2^2 \sum_{l=0}^{\infty} (r_2^l / r_1^{l+1}) P_l(\mu_{12}) - \int_{r_1}^{R(\mu_2)} dr_2 r_2^2 \sum_{l=0}^{\infty} (r_1^l / r_2^{l+1}) P_l(\mu_{12}) \right], \quad (28)$$

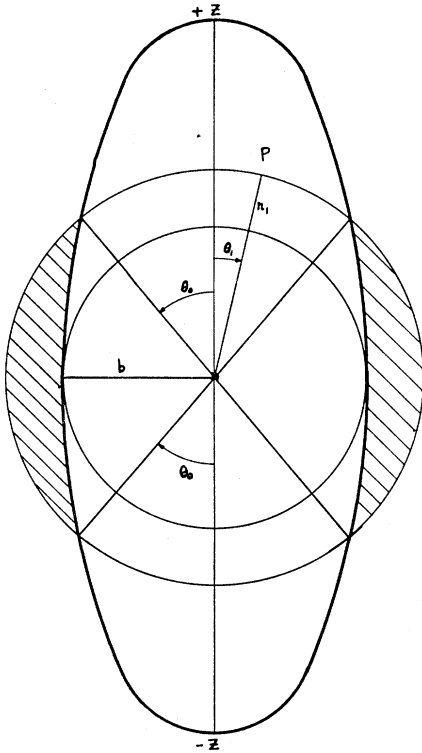


FIG. 1. Diagram used in calculating the electrostatic potential.

where $\mu_0(r_1)$ defines the intersection of the surface with a sphere of radius r_1 , i.e., $R(\mu_0) = r_1$, and μ_0 is taken to be positive. The first term in brackets in (28) corrects the first term in brackets in (27) for the shaded region of Fig. 1, which is not part of the drop. The second term in brackets in (28) cancels the second term of (27), which is spurious between $-\mu_0$ and $+\mu_0$. One can now write for the total Coulomb energy

$$U^c = U_0^c + U_1^c, \quad (29)$$

where

$$U_0^c = \frac{1}{2} \rho \int_0^{2\pi} d\phi_1 \int_{-1}^1 d\mu_1 \int_0^{R(\mu_1)} dr_1 r_1^2 V(r_1 < b) = 2\pi^2 \rho^2 \sum_{l=0}^{\infty} \int_{-1}^1 d\mu_1 P_l(\mu_1) \int_{-1}^1 d\mu_2 P_l(\mu_2) \int_0^{R(\mu_1)} dr_1 r_1^2 \times \left[r_1^{-(l+1)} \int_0^{r_1} r_2^{l+2} dr_2 + r_1^l \int_{r_1}^{R(\mu_2)} r_2^{1-l} dr_2 \right] \quad (30)$$

upon using the addition theorem to expand $P_l(\mu_{12})$ in terms of μ_1 , μ_2 , ϕ_1 , and ϕ_2 . The second term of (29) is given by

$$U_1^c = \frac{1}{2} \rho \int_0^{2\pi} d\phi_1 \int_{-1}^1 d\mu_1 \int_b^{R(\mu_1)} dr_1 r_1^2 \Delta V = 2\pi^2 \rho^2 \sum_{l=0}^{\infty} \int_{-1}^1 d\mu_1 P_l(\mu_1) \int_b^{R(\mu_1)} dr_1 r_1^2 \times \int_{-\mu_0(r_1)}^{\mu_0(r_1)} d\mu_2 P_l(\mu_2) \int_{R(\mu_2)}^{r_1} dr_2 r_2^2 \left(\frac{r_1^l}{r_2^{l+1}} - \frac{r_2^l}{r_1^{l+1}} \right) = 2\pi^2 \rho^2 \sum_{l=0}^{\infty} \int_{-1}^1 d\mu_1 P_l(\mu_1) \int_{-|\mu_1|}^{|\mu_1|} d\mu_2 P_l(\mu_2) \times \int_{R(\mu_2)}^{R(\mu_1)} dr_1 r_1^2 \int_{R(\mu_2)}^{r_1} dr_2 r_2^2 \left(\frac{r_1^l}{r_2^{l+1}} - \frac{r_2^l}{r_1^{l+1}} \right), \quad (32)$$

where the addition theorem has again been used and the order of integration inverted. Since $R(\mu)$ is an even function of μ , this becomes

$$U_1^c = 8\pi^2 \rho^2 \sum_{\text{even } l} \int_0^1 d\mu_1 P_l(\mu_1) \int_0^{\mu_1} d\mu_2 P_l(\mu_2) \times \int_{R(\mu_2)}^{R(\mu_1)} dr_1 r_1^2 \int_{R(\mu_2)}^{r_1} dr_2 r_2^2 \left(\frac{r_1^l}{r_2^{l+1}} - \frac{r_2^l}{r_1^{l+1}} \right). \quad (33)$$

It is readily seen that U_1^c contains no linear or quadratic terms in the deformation parameters a_n . While the individual terms of the infinite sum in (33) do not separately vanish, the series of terms has been found to converge to zero for third-order to seventh-order deformations. This is further discussed in Appendix 2

where the convergence proof is given explicitly for the third-order a_2^3 terms. For all deformations considered in this paper, the contribution of U_1^c to the electrostatic energy vanishes.

Returning to Eq. (30), we set

$$U_0^c = \sum_{l=0}^{\infty} u_l^c,$$

where

$$\begin{aligned} u_l^c &= -\frac{2\pi^2\rho^2}{(l+3)(l-2)} \int_1^1 d\mu P_l(\mu) R^{2-l}(\mu) \\ &\quad \times \int_{-1}^1 d\mu' P_l(\mu') R^{l+3}(\mu') \quad \text{for } l \neq 0, 2, \\ u_0^c &= \frac{\pi^2\rho^2}{3} \int_{-1}^1 d\mu R^2(\mu) \int_{-1}^1 d\mu' R^3(\mu') \\ &\quad - \frac{2\pi^2\rho^2}{15} \int_{-1}^1 R^5(\mu) d\mu, \\ u_2^c &= \frac{2\pi^2\rho^2}{5} \int_{-1}^1 d\mu P_2(\mu) R^5(\mu) \int_{-1}^1 d\mu' P_2(\mu') \ln R(\mu'). \end{aligned} \quad (34)$$

Introducing the dimensionless quantities $\xi^c = U^c / (3Z^2e^2/5R_0)$ and $\xi_l^c = u_l^c / (3Z^2e^2/5R_0)$, one finally obtains

$$\begin{aligned} \xi^c &= \sum_{\substack{l=0 \\ l \neq 2}}^{\infty} \xi_l^c - \frac{1}{8} \int_{-1}^1 [1 + \sum_n a_n P_n(\mu)]^5 d\mu \\ &\quad + \frac{3}{8} \int_{-1}^1 d\mu P_2(\mu) [1 + \sum_n a_n P_n(\mu)]^5 \\ &\quad \times \int_{-1}^1 d\mu' P_2(\mu') \ln [1 + \sum_n a_n P_n(\mu')], \end{aligned} \quad (35)$$

where, for $l \neq 2$,

$$\begin{aligned} \xi_l^c &= -\frac{15}{8(l+3)(l-2)} \int_{-1}^1 d\mu P_l(\mu) [1 + \sum_n a_n P_n(\mu)]^{2-l} \\ &\quad \times \int_{-1}^1 d\mu' P_l(\mu') [1 + \sum_n a_n P_n(\mu')]^{l+3}. \end{aligned}$$

If only even harmonics are included in $\sum a_n P_n$ (corresponding to symmetric deformations), ξ_l^c will vanish for odd l . The number of terms to be taken in the sum $\sum \xi_l^c$ will depend on the number of harmonics included in $\sum a_n P_n$ and the order of approximation desired for a power series in the deformation parameters a_n .

The potential energy of deformation is given by

$$\begin{aligned} \Delta U &= U^s + U^c - 4\pi R_0^2 O - 3Z^2e^2/5R_0, \\ \xi &= \Delta U / (4\pi R_0^2 O) = (\xi^s - 1) + 2x(\xi^c - 1), \end{aligned} \quad (36)$$

where $\xi^s = U^s / (4\pi R_0^2 O)$ and $2x = (3Z^2e^2/5R_0) / (4\pi R_0^2 O)$. Classical instability with respect to second harmonic (P_2) deformations sets in for $x=1$. Since the value of x for ${}_{100}\text{Fm}^{256}$ is in the neighborhood of 0.815, it is clear that large amplitude deformations must be investigated. This may be done by developing Eqs. (24) and (35) into a multiple power series in the a_n . We shall again assume that $a_n=0$ for $n>2$; then a straightforward calculation⁶ gives the following results for ξ^s and ξ^c :

$$\begin{aligned} \xi^s &= 1 + \frac{2}{5}a_2^2 - \frac{4}{3 \times 5 \times 7}a_2^3 - \frac{38}{5^2 \times 7}a_2^4 + \frac{36}{5 \times 7 \times 11}a_2^5 \\ &\quad + 0.053162a_2^6 - 0.088832a_2^7 + 0.0043574a_2^8 \dots, \end{aligned} \quad (37)$$

$$\begin{aligned} \xi^c &= 1 - \frac{1}{5}a_2^2 - \frac{4}{3 \times 5 \times 7}a_2^3 + \frac{157}{5^2 \times 7^2}a_2^4 + \frac{216}{5^3 \times 7^2 \times 11}a_2^5 \\ &\quad - 0.042411a_2^6 + 0.0057942a_2^7 + 0.0027802a_2^8 \dots, \end{aligned} \quad (38)$$

where a_0 has been eliminated with the aid of Eq. (15). Through substitution of Eqs. (37) and (38) in Eq. (36), the potential energy of deformation is obtained as a function of a_2 . The resulting expression for $\Delta U(a_2)$, which has been previously denoted by $V(a_2)$ in Sec. 2, will be used to calculate the penetration factor.

5. APPLICATION TO THE SPONTANEOUS FISSION OF FERMIVM

The penetration factor for spontaneous fission is obtained from Eq. (4) on substituting the effective-mass expression of Eq. (23) and the potential energy of deformation given by Eqs. (36), (37), and (38). Since the effective mass and the deformation energy are both expressed as power series in the deformation parameter a_2 , the applicability of the formulas is limited to nuclides for which the value of a_2 at the exit of the barrier is small enough to ensure good convergence. No known nuclide satisfies this condition at the present time; however, *faute de mieux*, the formulas will be applied to ${}_{100}\text{Fm}^{256}$ whose spontaneous fission half-life of 3.5 hours is the shortest that has yet been measured.⁷

The deformation energy is expressed in terms of $4\pi R_0^2 O$ and x for the nuclide in question. In terms of the usual parameters r_0 and γ of the Weizsäcker semiempirical mass formula, we have: $R_0 = r_0 A^{1/3}$, $4\pi R_0^2 O = \gamma A^{2/3}$, and $x = (Z^2/A) / (10\gamma r_0/3e^2)$. The values of r_0 and γ are not well fixed. The barrier depends sensitively on x and thus on γr_0 , which is uncertain by about 3%. The value of x appropriate to Fm^{256} is about 0.815. Two further difficulties arise in attempting

⁶ These terms were first calculated by one of the authors (R.D.P.) in collaboration with J. K. Knipp; an error in the earlier work, first noticed by M. E. Rose, has now been corrected.

⁷ Ghiorso, Harvey, Choppin, Thomson, and Seaborg, Phys. Rev. 98, 1518 (1955).

to apply the formulas; one of these is inherent in the liquid-drop model and the other is a result of approximations made here. It has been mentioned in Sec. 1 that the liquid-drop model activation energies or barrier heights fall off more rapidly with increasing Z^2/A than experiment indicates. The barrier height for the Fm^{256} fission should be about 3.0 Mev according to the calculations of references 1 and 2 with a reasonable choice of γ and r_0 . In view of the observed trend of fission thresholds with Z^2/A , this barrier height is undoubtedly too low. The second difficulty arises from our neglect of the deformation parameters a_n for $n > 2$; since the saddle path is not followed, the barrier obtained is theoretically too large. A barrier height close to 8 Mev is obtained for Fm^{256} from the a_2 terms given in Eqs. (37) and (38).

The absence of threshold data for photofission and neutron-induced fission of the highest Z elements makes it difficult to estimate the true barrier height. Some attempts^{8,9} to use spontaneous fission half-lives to estimate fission thresholds have been based on a semi-empirical formula which gives fairly good results in the neighborhood of $Z=92$. The extrapolation of this formula to $Z \sim 100$ is very questionable, however, in spite of some interesting correlations with slow neutron fissionabilities. The barrier height estimates of reference 8 give 4.9 Mev for Fm^{256} ; this value, which is probably too large, includes an uncertain allowance for the induced fission threshold being below the top of the barrier.

Because of the sensitive dependence of the penetration factor on the height of the barrier, no reasonable value of the half-life can be expected from a barrier whose height is not approximately correct. We have therefore proceeded by treating x as an *ad hoc* parameter whose value is adjusted to give the theoretical barrier a reasonable height for the Fm isotopes. Although the height of the barrier is made empirical, its width and general shape are determined by Eqs. (37) and (38). It is found by comparison with the saddle point parameters of reference 2 that the adjusted barrier is somewhat too wide for its height, and that it is therefore better to underestimate than overestimate the height. In view of these uncertainties, we shall not attempt to assign a definite barrier height or x value to the Fm isotopes, but we shall assume that the barrier heights do lie between the liquid-drop value of 3.0 Mev and the estimate of 4.9 Mev from reference 8. It should be noted that uncertainties in the liquid-drop values arise from zero-point effects, which have not been taken into account, and also from a possible nonspherical shape of the nucleus before undergoing fission. Further uncertainty in the barrier for any specific nuclide comes from

possible effects of shell structure on the ground state energy, similar effects being absent from the activated state.

The considerations of the preceding paragraph might appear to preclude the possibility of a significant comparison between the theoretical formulas of this paper and experiment. While such a comparison would not be very meaningful for the half-life of a specific nuclide, e.g., Fm^{256} , we are nevertheless able to make a reasonably reliable estimate of the variation of the penetration factor and half-life produced by a variation in the barrier height for nuclides of the highest Z values, e.g., the Fm isotopes. If T denotes the spontaneous fission half-life and $\tau \equiv \log_{10} T$, then the change $\delta\tau$ associated with a change in barrier height $\delta\epsilon$ can be written as: $\delta\tau = k\delta\epsilon$, where k is defined¹⁰ as the *spontaneous fission hindrance factor*. In general k will be expected to vary with Z^2/A and this variation has been studied in reference 10; we shall now calculate k for the fermium isotopes ($Z^2/A = 39$).

The evaluation of the penetration factor is straightforward when the choice of x has been made. The values of x were selected to give barrier heights of 3.0 Mev and 4.9 Mev. Since the effective mass has been determined through terms in a_2^4 and since the deformation energy expansion begins with the a_2^2 term, consistency requires that terms through a_2^6 in Eqs. (37) and (38) be included in the integrand of the penetration integral. The additional terms in a_2^7 and a_2^8 have been used only to fix more accurately the upper limit of the integral, which corresponds to the exit of the barrier. The exit values of a_2 are so large (~ 0.8 to 0.9) that the convergence is not satisfactory. Some indication of the error that can arise in this way is given by dropping the a_2^8 term in the deformation energy and also by omitting the a_2^4 term in the effective mass. The ratio of the penetration factors or reciprocal half-lives corresponding to barrier heights of 3.0 Mev and 4.9 Mev is found to be $10^{7.4 \pm 1.6}$ where the limits of error correspond to the convergence uncertainties mentioned above. The resulting value of the hindrance factor is 3.7 ± 0.8 per Mev—i.e., an increase of 1 Mev in the barrier height corresponds to an increase of $10^{3.7}$ in the half-life. This result disagrees with the half-life formula proposed in reference 2, which gives a constant hindrance factor of 7.85 per Mev. While the latter formula has received some confirmation for the uranium isotopes, our results indicate that it must fail badly for higher Z values.

The hindrance factor has been deduced from experimental data in an ingenious way by Swiatecki.¹⁰ He finds that deviations of individual τ values from a straight-line graph of τ vs Z^2/A are correlated with deviations of the nuclear masses from the Weizsäcker semiempirical mass formula. The latter deviations are probably associated with shell-structure effects

⁸ G. T. Seaborg, Phys. Rev. **88**, 1429 (1952); R. Vandenbosch and G. T. Seaborg, Phys. Rev. **110**, 507 (1958).

⁹ J. R. Huizenga, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955* (United Nations, New York, 1956), Vol. 2, p. 208.

¹⁰ W. I. Swiatecki, Phys. Rev. **100**, 937 (1955).

that are presumably absent in the activated state at the top of the barrier; hence the mass fluctuations correspond to fluctuations in barrier heights and these are connected with fluctuations in the half-lives. A study of 28 spontaneous fissions of elements from $Z=90$ to $Z=100$ has shown a variation of the hindrance factor with Z^2/A which is well represented by¹⁰: $k=42.5-Z^2/A$ per mMU. Our calculated value of k for the fermium isotopes is in good agreement with this formula.

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APPENDIX 1

The calculation of the velocity field for the liquid drop was begun in Sec. 3. Equation (16) is used to determine the coefficients b_n of the velocity potential; any b_n can be written as $b_n = \dot{a}_2 \sum_{m=0}^n \beta_n^{(m)} a_2^m$. One obtains the k th order approximation by terminating the sum with $m=k$. Equation (16) is an equation in two different sets of linearly independent functions; one set is the set of Legendre polynomials. All products of Legendre polynomials can be removed by the combination laws; one then equates the coefficients of the separate $P_n(\mu)$ for the left and right sides of Eq. (16). The other set of independent functions is the set of powers of a_2 ; this set also leads to equating of coefficients for each member of the set. The $\beta_n^{(m)}$ are chosen to satisfy these requirements for the two linearly independent sets.

Two combination laws are used in reducing the left side of Eq. (16) to a linear combination of the $P_n(\mu)$. The two are

$$P_2 P_n = -\frac{3}{2} \frac{(n+1)(n+2)}{(2n+1)(2n+3)} P_{n+2} + \frac{n(n+1)}{(2n-1)(2n+3)} P_n$$

$$+ \frac{3}{2} \frac{n(n-1)}{(2n-1)(2n+1)} P_{n-2},$$

$$P_2^2 P_n^2 = \frac{3n(n+1)}{2n+1} \left[-\frac{n+2}{2n+3} P_{n+2} + \frac{2n+1}{(2n-1)(2n+3)} P_n \right.$$

$$\left. + \frac{n-1}{2n-1} P_{n-2} \right].$$

When all powers of a_2 greater than a_2^k are discarded from the reduced form of Eq. (16), the coefficient of each $P_n(\mu)$ is reduced to a finite number of terms. For large N , $P_N(\mu)$ in the original form of Eq. (16) contributes to $P_n(\mu)$ in the reduced form through combinations such as $[a_2 P_2(\mu)]^m P_N(\mu)$; in the k th approxima-

tion these products contribute to the coefficient of $P_n(\mu)$ only when $N \leq n+2k$.

We now calculate the b_n 's to the second approximation. In this approximation

$$R^m(\mu) \rightarrow 1 + m a_0 + m a_2 P_2(\mu) + \frac{1}{2} m(m-1) a_2^2 [P_2(\mu)]^2,$$

and

$$\dot{a}_0 = -\frac{2}{5} [1 + (a_2/7)] a_2 \dot{a}_2.$$

Equating the coefficients of P_0 in the reduced form of Eq. (16), one gets

$$2b_2(-\frac{2}{5}a_2) + 4b_4 a_2^2 [2/(5 \times 7)] = \dot{a}_0.$$

Since $\beta_4^{(0)} = 0$, $b_4 a_2^2$ is of order of a_2^3 and is therefore dropped. The resulting equation,

$$b_2 = \frac{1}{2} \dot{a}_2 [1 + (a_2/7)],$$

checks the first-order result for b_2 . Equating the coefficients of P_2 yields

$$2b_2 \left(1 - \frac{1}{7} a_2 - \frac{1}{5} a_2^2 \right) + 4b_4 \left(\frac{1}{7} a_2 \right) = \dot{a}_2,$$

and, using the first-order result for b_4 , one obtains

$$2\dot{a}_2 \left\{ \frac{1}{2} \left(1 + \frac{1}{7} a_2 \right) + \beta_2^{(2)} a_2^2 \right\} \left(1 - \frac{1}{7} a_2 - \frac{1}{5} a_2^2 \right)$$

$$+ \frac{4}{7} a_2 \left[-\frac{27}{2 \times 5 \times 7} a_2 \dot{a}_2 \right] = \dot{a}_2,$$

which is an identity in a_2 to quadratic terms if $\beta_2^{(2)} = 54/(5 \times 7^2)$. Equating P_4 coefficients leads similarly to the value of $\beta_4^{(2)}$ and equating P_6 coefficients leads to the first nonvanishing term ($\beta_6^{(2)}$) in b_6 . On equating P_n coefficients for $n \geq 8$, the corresponding b_n are found to vanish in second order. The second-order result is

$$b_2 = \dot{a}_2 \left(\frac{1}{2} + \frac{1}{2 \times 7} a_2 + \frac{54}{5 \times 7^2} a_2^2 \right),$$

$$b_4 = \dot{a}_2 \left(0 - \frac{27}{2 \times 5 \times 7} a_2 + \frac{459}{5 \times 7^2 \times 11} a_2^2 \right),$$

$$b_6 = \dot{a}_2 \left(0 + 0 + \frac{45}{7 \times 11} a_2^2 \right),$$

$$b_n = 0 \quad \text{for } n \geq 8.$$

In the higher orders of approximation it is found that (1) the k th order equation for the P_0 coefficient merely checks the value of b_2 found in the $(k-1)$ th order, (2) the first nonvanishing term ($\beta_n^{(l)}$) in b_n appears in the l th order equation for the coefficient of P_n , and (3) with each new order there is a single, new,

TABLE I. Coefficients of the velocity potential.

$n =$	2	4	6	8	10
$\beta_n^{(0)}$	$\frac{1}{2}$	0	0	0	0
$\beta_n^{(1)}$	$1/(2 \times 7)$	$-27/(2 \times 5 \times 7)$	0	0	0
$\beta_n^{(2)}$	$54/(5 \times 7^2)$	$459/(5 \times 7^2 \times 11)$	$45/(7 \times 11)$	0	0
$\beta_n^{(3)}$	-0.033439	-0.92474	-0.67367	-1.05734	0
$\beta_n^{(4)}$	0.133007	0.70465	2.8667	1.99330	2.0834

nonzero b_n . In the k th order approximation,

$$b_n = \dot{a}_2 \sum_{\frac{1}{2}(n-2)}^k \beta_n^{(m)} a_2^m,$$

and when $n > 2k+2$, b_n is zero in the k th order. The results to fourth order are given in Table I.

APPENDIX 2

We wish to evaluate formula (33) for U_1^c . The integrations over r_2 and r_1 are carried out and we then substitute $1+a_0+a_2P_2(\mu)$ for $R(\mu)$. Expanding in powers of a_2 and neglecting terms beyond third order, it is readily found that the terms in a_2 and a_2^2 vanish [as may also be seen by inspection of Eq. (33)] and that the a_2^3 term is given by

$$\begin{aligned}
 U_1^c &= \frac{4}{3}\pi^2\rho^2a_2^3 \sum_{\text{even } l} (2l+1) \int_0^1 d\mu P_l(\mu) \\
 &\quad \times \int_0^\mu d\mu' P_l(\mu') [P_2(\mu) - P_2(\mu')]^3 \\
 &= 2\pi^2\rho^2a_2^3 \int_0^1 d\mu \int_0^\mu d\mu' \left\{ \sum_{l=0}^{\infty} (2l+1)(\mu^2 - \mu'^2) \right. \\
 &\quad \left. \times P_l(\mu)P_l(\mu') \right\} [P_2(\mu) - P_2(\mu')]^2.
 \end{aligned}$$

It is readily seen that

$$\begin{aligned}
 &\sum_{l=0}^N (2l+1)(\mu^2 - \mu'^2) P_l(\mu) P_l(\mu') \\
 &= \frac{(N+1)(N+2)}{2N+3} [P_{N+2}(\mu)P_N(\mu') - P_N(\mu)P_{N+2}(\mu')].
 \end{aligned}$$

The a_2^3 term is then proportional to

$$\lim_{N \rightarrow \infty} \frac{(N+1)(N+2)}{2N+3} \int_0^1 d\mu \int_0^\mu d\mu' [P_{N+2}(\mu)P_N(\mu') - P_N(\mu)P_{N+2}(\mu')] [P_2(\mu) - P_2(\mu')]^2.$$

A typical term in the integrand is of the form: $P_{N+2}(\mu) \times P_N(\mu') \mu^{2m} \mu'^{2n}$. We set $\mu^{2m} P_{N+2}(\mu) = \sum A_M P_M(\mu)$ and $\mu'^{2n} P_N(\mu') = \sum B_L P_L(\mu')$ and note that, when N is large, L and M are large even integers and the coefficients A_M and B_L become independent of N . Thus one obtains a finite number of integrals of the form

$$\begin{aligned}
 &\frac{(N+1)(N+2)}{2N+3} \int_0^1 d\mu P_M(\mu) \int_0^\mu d\mu' P_L(\mu') \\
 &= \frac{(N+1)(N+2)}{(2N+3)(2L+1)} \int_0^1 d\mu P_M(\mu) [P_{L+1}(\mu) - P_{L-1}(\mu)].
 \end{aligned}$$

The integral $\int_0^1 P_M(\mu) P_K(\mu) d\mu$ with M even and K odd can be readily evaluated in closed form and, when M and K are both large and of order N , can be seen to vanish as N^{-1} as $N \rightarrow \infty$. The a_2^3 term therefore vanishes. A similar proof can be given to show that higher order terms also vanish.