Surface Coupling Mechanism for Approaching Statistical Equilibrium in Compound Nucleus Formation, with Application to Fission*

LAWRENCE WILETS
Physics Department, University of Washington, Seattle, Washington
(Received May 27, 1959)

In a particular representation where the state vectors are not eigenstates of the Hamiltonian, coupling terms remain which cause "virtual" or "real" transitions among states. An appropriate choice of representation depends upon the physical processes involved. The decay of a compound nucleus, expecially by fission, is considered. The strong coupling representation of the unified model is employed, with surface oscillations inducing transitions among states of the representation. A diffusion equation is derived to describe the flow of probability among the states available within constraints. An estimate of the characteristic relaxation time for arriving at statistical equilibrium is obtained. Only when the relaxation time is short compared with a basic reaction time are statistical arguments valid to evaluate the reaction rate. As an example the relevant reaction time in the fission process is a collective vibrational period. The necessary condition appears to be satisfied for excitation energies more than a few Mev above threshold. Arguments are presented to show why equilibrium may not be maintained at lower energy. Thus the usual estimate of the number of open channels, $2\pi\bar{\Gamma}_f/D$, would give a number lower than what one would estimate simply from penetrability of the fission barrier. This seems to explain, at least in part, the anomalously low numbers of channels obtained in this manner. Problems relating to the validity of oft made statistical assumptions at scission are also discussed.

I. INTRODUCTION

THE compound nucleus of Niels Bohr may be regarded as an ensemble of intermediate states in a nuclear reaction, the nature of which is independent of the manner of formation. The ensemble is, in some sense, statistical, for the model admits only a few constants of the motion. These constants include energy, angular momentum, and parity—the "rigorous" invariants for nuclear motion—but they may also include such approximate constants of the motion as the component of angular momentum along a nuclear symmetry axis.

If one were able to write down a complete set of eigenstates of the system, then the wave function describing some particular process would be a linear combination, with certain phase relations, of such eigenstates. The statistical assumption in Bohr's model is that all states available within specified constraints (the constants of motion) are equally probable, and that the phase relations among the various components are immaterial. In examining the development of the system, one might be tempted to assume, as does Pauli[1] in somewhat different context, that the phase relations among the components are random for all times. However, one must remember that the phases are intimately related to the mode of formation, and only after the passage of some characteristic relaxation time is the "importance" of these phase relations lost. The compound nuclear model is useful only if subsequent decay occurs after this relaxation time.

An alternative description—that to be employed in this paper—is one which utilizes some appropriate representation other than eigenstates. As the system develops in time, off-diagonal matrix elements of the Hamiltonian induce transitions among various states of the representation. The transitions are classified as "real" or "virtual" depending upon whether or not energy is conserved. The classification is, of course, dependent upon the choice of representation. Through real transitions, excitations may pass among many states and via various paths before returning to the initial state. In such cases, interference terms are assumed to drop out. One then speaks in terms of probabilities rather than amplitudes. The rate of approach to statistical equilibrium is determined by a diffusion equation for probability flow.

Both descriptions are manifestly incomplete, as must be any irreversible description. The question is not whether one has a complete description of a nuclear reaction, but rather whether one has an adequate description for processes of interest. The basic questions are essentially the same as those which arise generally in discussing irreversible processes in a microscopically reversible system. We will not attempt to justify the procedures here in such detail as has been done for infinite systems (see Van Hove[2]), but it must be noted that whatever validity the description does possess improves (except for questions related to the validity of perturbation theory) with increasing size and excitation of the nucleus.

It is evident that a description based on a particular representation must lead to a characteristic relaxation time which is dependent upon the choice of representation, since this choice determines the magnitude of the off-diagonal matrix elements. Fortunately, the choice of representation is not completely arbitrary, since it is

* Supported in part by the U. S. Atomic Energy Commission.
[1] W. Pauli, *Festschrift zum 60. Geburstage A. Sommerfelds* (Hirzel, Leipzig, 1928), p. 30.

[2] L. Van Hove, Physica **23**, 441 (1957). Further references are given here.

governed by the physics of the process of interest, and by the requirement that the off-diagonal matrix elements not be so large that perturbation theory fails. Although there is no clear prescription for the choice, the present example may serve to illustrate some of the considerations.

We will be particularly interested in decay of the compound nucleus by fission, and therefore choose the unified model representation of strong coupling between particle and collective (surface) modes. This provides a very convenient framework in which to follow the fission process. Indeed, any essentially different representation I can think of requires resolution back into the strong-coupling representation in order to discuss the fission process, and this would then require following phase relations.

If we were interested in decay of the compound nucleus by neutron emission, we similarly might use the strong-coupling representation to find how much energy is concentrated in single particle modes. On the other hand, we might use a pure independent particle model for a representation, with "residual" two-body interactions inducing real transitions among the independent particle states.

Virtual transitions induced by surface coupling play an important role in the determination of the mass parameter in collective rotations and vibrations, as first demonstrated by Inglis[3] through his "cranking model." In the case of low-energy spectrum, "real" transitions are not possible, since all transitions involve an increase in both particle and collective energies.

Hill and Wheeler[4] discuss in some detail real transitions induced by surface coupling according to the time-dependent Hamiltonian formulation. They name the phenomenon "slippage," with reference to the jump of excitation from one level to another at a near-crossing. The present paper was influenced considerably by their work, and is, in part, an attempt to put their considerations on a quantitative basis for high excitation. The reader is referred to the Hill-Wheeler paper for a clear physical picture of the mechanism.

The surface coupling mechanism has a thoroughgoing analogy in atomic and molecular physics. In the collision of two atoms, for example, the interatomic (or internuclear) distance plays the role of a collective coordinate. If the collision is slow, the electronic wave functions adjust adiabatically to the changing potential. The adiabatic wave functions form a representation which for finite interatomic velocities are not solutions of the Schrödinger equation, although they do satisfy the equation long before or long after the collision. Real transitions induced during the collision give rise to atomic excitations which may persist after the atoms have separated.

Transitions are computed below using first-order perturbation theory. When the probability of making a transition becomes large (in a time compared with, say, a collective period divided by the quantum number of the collective state), the perturbation approach breaks down. When this happens, however, the relaxation time is already short compared with other physical processes. We are primarily concerned with obtaining an estimate of the relaxation time to determine if statistical equilibrium is reached before decay, or other processes, set in.

[3] D. R. Inglis, Phys. Rev. **96**, 1059 (1954); **97**, 701 (1955).
[4] D. L. Hill and J. A. Wheeler, Phys. Rev. **89**, 1102 (1953).

II. DERIVATION OF THE DIFFUSION EQUATION

A. The Strong-Coupling Representation

The nuclear system is to be described by a Hamiltonian

$$H=H_p+H_s+H', \tag{1}$$

where $H_p=H_p(\mathbf{x}_1\cdots\mathbf{x}_A;\ \boldsymbol{\nabla}_1\cdots\boldsymbol{\nabla}_A)$ describes the particle degrees of freedom and $H_s=H_s(\beta;\ \partial/\partial\beta)$ the collective (or surface or deformation) degrees of freedom.[5] There could be several collective coordinates, but we shall specifically refer to Bohr's symmetric quadrupole deformation parameter β. The interaction term $H'=H'(\mathbf{x}_1\cdots\mathbf{x}_A;\beta)$ is assumed to be independent of the particle or collective momenta. An alternative formulation of the problem in terms of a time-dependent Hamiltonian, such as Hill-Wheeler[4] and Inglis[3] have introduced, has certain conceptual advantages, but in the present context it would obscure the evaluation of certain important approximations. The time-dependent formulation treats $\beta=\beta(t)$ as a classical variable while treating the particle modes quantum mechanically. The development can be carried through in quite a parallel way as for the full quantum-mechanical treatment. The results are identical in the limit that the collective mass parameter becomes infinite (and the collective frequency goes to zero).

The strong-coupling representation (not *approximation*) is defined by the following equations:

$$(H_p+H')u_j(\mathbf{x};\beta)=\epsilon_j(\beta)u_j(\mathbf{x};\beta), \tag{2}$$

$$[\epsilon_j(\beta)+H_s]\phi_{j\lambda}(\beta)=E_{j\lambda}\phi_{j\lambda}(\beta), \tag{3}$$

where $\mathbf{x}=(\mathbf{x}_1\cdots\mathbf{x}_A)$. The functions $u_j(\mathbf{x};\beta)\phi_{j\lambda}(\beta)$ form a complete set in $(\mathbf{x},\beta)$-space. Thus a general wave function for the system may be written

$$\Psi=\textstyle\sum_{j\lambda} C_{j\lambda}(t)u_j(x;\beta)\phi_{j\lambda}(\beta)\exp(-i\hbar^{-1}\tilde{E}_{j\lambda}t). \tag{4}$$

The exponential time variation is introduced for

[5] The system possesses redundant variables, since the particle coordinates are sufficient. In principle subsidiary conditions should be stated, but we ignore the matter for two reasons. Firstly, the collective coordinates are few in number compared with the particle coordinates and the redundancy is relatively unimportant. Secondly, and perhaps more significantly, is the point that the particle degrees of freedom are not fully developed, and the collective coordinates may be regarded as describing the "unexcited," but adiabatically deformed, inner nucleonic shells.

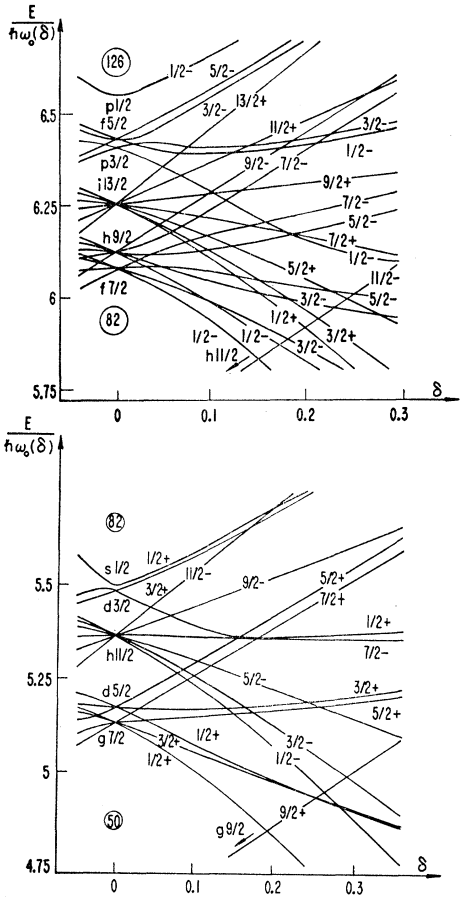

FIG. 1. Nilsson spectra for protons from $Z=50$ to $Z=82$ (below) and for neutrons from $N=82$ to $N=126$ as functions of nuclear deformation $\delta\approx 0.95\beta$ (reproduced from Nilsson, reference 6).

convenience at this point, with the explicit choice of $\tilde{E}_{j\lambda}$ made in (7) below.

Schrödinger's equation,

$$i\hbar\dot{\Psi}=H\Psi, \tag{5}$$

can now be written in the equivalent form

$$i\hbar\dot{C}_{j\lambda}=\sum_{j'\lambda'}C_{j'\lambda'}\int\phi_{j\lambda}{}^{*}u_{j}{}^{*}[H_{s},u_{j'}]\phi_{j'\lambda'}d^{3A}xd\beta \times\exp[i\hbar^{-1}(\tilde{E}_{j\lambda}-\tilde{E}_{j'\lambda'})t]. \tag{6}$$

The diagonal terms in the sum, $(j'\lambda')=(j\lambda)$, have been removed (hence the prime on $\sum$) by choosing

$$\tilde{E}_{j\lambda}=E_{j\lambda}+\int\phi_{j\lambda}{}^{*}u_{j}{}^{*}[H_{s},u_{j}]\phi_{j\lambda}d^{3A}xd\beta. \tag{7}$$

The treatment to this point [assuming (1)] is exact. A variety of approximations follow, of course. Although the nature of the approximations is mentioned in most cases, a critical discussion is deferred until Sec. IIF in order to give continuity to the development.

Some comments about the structure of $u_j(\mathbf{x};\beta)$ and $\epsilon_j(\beta)$ are appropriate. In an independent-particle model the *single-particle* energy level spectrum, as a function of deformation, would assume a form like that calculated by Nilsson.[6] A section of Nilsson's spectrum is shown in Fig. 1. The many-particle spectrum would be similar in nature except that it would be far denser and include frequent crossings of energy levels of the same character (spin and parity). This is represented schematically by the dotted lines in Fig. 2. The inclusion of residual two-body forces will quite generally remove the degeneracy of states of the same character, and therefore we expect no crossings. The anticipated spectrum is represented schematically by the solid lines in Fig. 2. The single-particle structure of the u_j (and here j labels a continuous solid line) changes rather abruptly at a near-crossing. This will be seen to be of considerable importance when discussing matrix elements of single-particle operators.

B. Probability Flux and Diffusion

The transition rate from state j to state j' (all λ') is given by the well-known expression[7]

$$R(j;j')=\frac{2\pi}{\hbar}\left|\int\phi_{j\lambda}{}^{*}u_{j}{}^{*}[H_{s},u_{j'}]\phi_{j'\lambda'}d^{3A}xd\beta\right|^{2}\frac{d\lambda}{dE}. \tag{8}$$

The formula is derived for a continuum (of collective

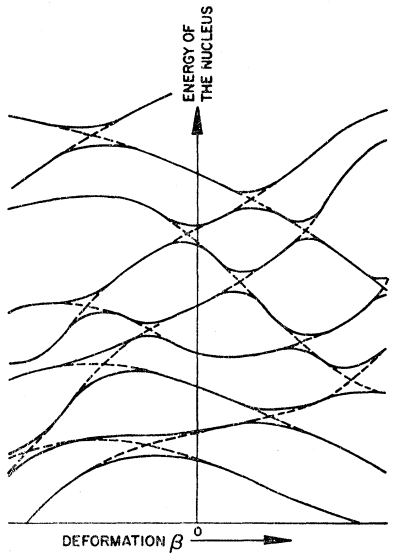

FIG. 2. A schematic representation of energy levels of an entire nucleus as a function of deformation. Levels of only one character (K and π) are considered. The broken lines correspond to an independent particle model, and exhibit possible level crossings. The solid lines include the effect of residual two-body interactions, which prevent level crossings.

[6] S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd, **29**, No. 16 (1955).

[7] Cf. L. I. Schiff, *Quantum Mechanics* (McGraw-Hill Book Company, New York, 1949), Chap. VIII.

states) and is valid only to first order in perturbation theory. The factor $d\lambda/dE$ is the level density of collective states. The transitions conserve energy (at least approximately in noncontinuum theory), so that $\bar{E}_{j'\lambda'}\approx\bar{E}_{j\lambda}$. (Henceforth, we will replace $\bar{E}_{j\lambda}$ by $E_{j\lambda}$.) The transitions are what we call real, and the continuum approximation will be seen to be valid (Sec. II F) only if the lifetime broadening of a state is larger than the collective level spacing.

We are now in a position to write down an equation for probability flow. Let the probability that the state j is occupied be denoted by $p(j)$. We need not specify λ, since it is determined by the requirement that $E_{j\lambda}$ approximately equals the total nuclear excitation energy E^*. Then the net flux, l, of probability from states with particle energies below $\epsilon_j(\beta)$ to states above $\epsilon_j(\beta)$ is given by

$$l(j)=\sum_{j'<j}\sum_{j''>j}R(j';j'')[p(j')-p(j'')]. \tag{9}$$

We wish to change from discrete indices $(j\lambda)$ to some variable which can go over to a continuum description conveniently. The particle energies $\epsilon_j(\beta)$ can fill this requirement, but the β-independence introduces some ambiguity. Therefore, we will label a particle state, j, by the energy $\epsilon=\epsilon_j(\beta_{\min})$, where $\beta_{\min}$ is the deformation where the particle energy is a minimum (equilibrium value of β). The collective wave functions $\phi_{j\lambda}$ will be denoted by $\phi_{E^*-\epsilon}$. Actually, we are most interested in what happens near the equilibrium position anyway since that is where the collective velocity is greatest and the transition rate fastest. We will often encounter the difference $\epsilon_j(\beta)-\epsilon_{j'}(\beta)$, which near equilibrium is a more slowly varying function of β than either $\epsilon_j(\beta)$ or $\epsilon_{j'}(\beta)$; we will generally set $\epsilon_j(\beta)-\epsilon_{j'}(\beta)=\epsilon-\epsilon'$.

As is made plausible below, $R(\epsilon';\epsilon'')$ is taken to be of the form $R(|\epsilon''-\epsilon'|)$ and is a generally falling function of the argument. To first order in the gradient of p, we have

$$l(\epsilon)=-W^2(\epsilon)\frac{\partial p}{\partial\epsilon}\int_\epsilon^{E^*}d\epsilon''\int_0^\epsilon d\epsilon'(\epsilon''-\epsilon')R(\epsilon''-\epsilon'). \tag{10}$$

By extending the outer limits to $\pm\infty$, we find

$$l(\epsilon)=-W^2\frac{\partial p}{\partial\epsilon}\int_0^\infty\eta^2R(\eta)d\eta, \tag{11}$$

where the dummy variable η is $\epsilon''-\epsilon'$. Here $W(\epsilon)$ is the particle energy level density and differs from the total level density. We define the diffusion coefficient to be

$$\sigma(\epsilon)\equiv W(\epsilon)\int_0^\infty\eta^2R(\eta)d\eta, \tag{12}$$

and thus

$$l(\epsilon)=-W\sigma(\partial p/\partial\epsilon). \tag{13}$$

We now write down a formal equation for the rate of change of the probability, $pWd\epsilon$, that some state in the energy range ϵ to $\epsilon+d\epsilon$ is occupied. Neglecting other effects which tend to change the state of the system (radiation, particle emission, fission, etc.), we find

$$W\left(\frac{\partial p}{\partial t}\right)_{\text{diffusion}}=-\frac{\partial l}{\partial\epsilon}=\frac{\partial}{\partial\epsilon}W\sigma\frac{\partial p}{\partial\epsilon}. \tag{14}$$

Integration of (14) over the available energy range reveals that total probability is conserved, since the flux must vanish at the end points. The steady state solution is $p=$constant, which follows immediately from the symmetry of the matrix elements.

C. Estimation of the Diffusion Coefficient

The explicit form of the collective Hamiltonian is taken to be the kinetic energy operator

$$H_s=T_s=-\frac{\hbar^2}{2B}\frac{\partial^2}{\partial\beta^2}, \tag{15}$$

with B assumed independent of β. The effective potential energy for surface vibration is obtained from $\epsilon_j(\beta)$ [see (3)]. Hence,

$$[H_s,u_{j'}]=-\frac{\hbar^2}{2B}\left(\frac{\partial^2u_{j'}}{\partial\beta^2}+2\frac{\partial u_{j'}}{\partial\beta}\frac{\partial}{\partial\beta}\right). \tag{16}$$

We will drop the first term in the parentheses compared with the second. Using the notation

$$\langle j''|O|j'\rangle\equiv\int u_{j''}{}^*Ou_{j'}d^{3A}x, \tag{17}$$

we find

$$\left\langle j''\left|\frac{\partial H'}{\partial\beta}\right|j'\right\rangle=\left\langle j''\left|\left[H_p+H',\frac{\partial}{\partial\beta}\right]\right|j'\right\rangle$$
$$=(\epsilon_{j''}-\epsilon_{j'})\left\langle j''\left|\frac{\partial}{\partial\beta}\right|j'\right\rangle. \tag{18}$$

By combining (8), (12), (16), and (18), we find for the diffusion coefficient,

$$\sigma(\epsilon)=\frac{2\pi\hbar^3W}{B^2}\int_0^\infty\frac{d\lambda}{dE}d(\epsilon''-\epsilon')$$
$$\times\left|\int\phi_{E^*-\epsilon''}{}^*\left\langle\epsilon''\left|\frac{\partial H'}{\partial\beta}\right|\epsilon'\right\rangle\frac{\partial\phi_{E^*-\epsilon'}}{\partial\beta}d\beta\right|^2. \tag{19}$$

Except for details in fluctuation, we assume that the $\langle\epsilon''|\partial H'/\partial\beta|\epsilon'\rangle$ do not depend on the energy difference $\epsilon''-\epsilon'$, and that the $\phi_{j\lambda}$ are independent of j. This

leads to the approximate sum rule

$$\sigma(\epsilon)\approx\frac{2\pi\hbar^3 W}{B^2}\sum_{\lambda''(>\lambda')}\left|\int\phi_{\lambda''}{}^*\left\langle\epsilon''\left|\frac{\partial H'}{\partial\beta}\right|\epsilon'\right\rangle\frac{\partial\phi_{\lambda'}}{\partial\beta}d\beta\right|^2$$

$$\approx\frac{2\pi\hbar W}{B}\int\phi_{\lambda'}{}^*\left(\left\langle\epsilon''\left|\frac{\partial H'}{\partial\beta}\right|\epsilon'\right\rangle^2\right)_{\text{Av}}H_s\phi_{\lambda'}d\beta$$

$$\equiv\frac{2\pi\hbar W}{B}[(\mathcal{K}_\beta)_{\epsilon''\epsilon'}{}^2]_{\text{Av}}\bar{H}_s, \tag{20}$$

where the abbreviation $(\mathcal{K}_\beta)_{\epsilon''\epsilon'}=\langle\epsilon''|\partial H'/\partial\beta|\epsilon'\rangle$ has been introduced. $\mathcal{K}_\beta$ is, of course, a function of β. A term of the form

$$\int\phi_{\lambda'}{}^*\mathcal{K}_\beta(\partial\mathcal{K}_\beta/\partial\beta)(\partial\phi_{\lambda'}/\partial\beta)d\beta$$

has been neglected compared with

$$\int\phi_{\lambda'}{}^*\mathcal{K}_\beta{}^2(\partial^2\phi_{\lambda'}/\partial\beta^2)d\beta.$$

A factor of one-half appears because the λ''-sum is only half complete.

If the ϕ_λ were oscillator functions and if the elements $\mathcal{K}_\beta$ were independent of β and $\epsilon''-\epsilon'$, we would have the selection rule $\lambda''=\lambda'\pm1$, with (20) holding exactly. [Indeed, if the $\mathcal{K}_\beta$ are constant, no matter what the ϕ's, (20) reduces to the dipole sum rule.] The selection rule is broken down both by the anharmonicity of the ϕ_λ and by the nonconstancy of $\mathcal{K}_\beta$. Since $\partial H'/\partial\beta$ is a sum of single-particle operators, it connects shell-model states which differ in at most one single particle constituent. Whenever the relative single-particle structure between states ϵ'' and ϵ' changes, as it does at near-crossings, the matrix element changes (unless it is zero anyway). Then $\mathcal{K}_\beta$ as a function of β consists of a series of segments. The shorter the segments, the less each contributes to (20), but then contributions come from further away in $(\epsilon''-\epsilon')$. For high collective states, where the WKB approximation is valid, (20) represents a Fourier analysis of the $\mathcal{K}_\beta$ elements, and then a resumming of the absolute square values of the Fourier coefficients. This leads to the integral over the square of the elements.

The derivation of (20) assumes only that the fluctuations in $\mathcal{K}_\beta$ are, on the average, independent of $\epsilon''-\epsilon'$. If anything, the fluctuations increase with $\epsilon''-\epsilon'$ because there are more opportunities for change, with β, of the single-particle structure. In that case, (20) represents a lower estimate of the diffusion coefficient.

In order to estimate $(\mathcal{K}_\beta{}^2)_{\text{Av}}$, we adopt Nilsson's form for the particle-deformation interaction:

$$H'=-\beta m\omega_0{}^2\sum_{p=1}^{A}r_p{}^2Y_{20}(\theta_p), \tag{21}$$

where ω_0 is the shell-model oscillator frequency: $\hbar\omega_0\sim40\ \text{A}^{-\frac{1}{3}}$ Mev. Then it follows that

$$\partial H/\partial\beta=-m\omega_0{}^2\sum r_p{}^2Y_{20}(\theta_p), \tag{22}$$

and for the mean square average of the matrix elements, we have

$$(\mathcal{K}_\beta{}^2)_{\text{Av}}\equiv\left(\left\langle\epsilon''\left|\frac{\partial H}{\partial\beta}\right|\epsilon'\right\rangle^2\right)_{\text{Av}}=\frac{1}{N}\sum_{\epsilon'}\left\langle\epsilon''\left|\frac{\partial H}{\partial\beta}\right|\epsilon'\right\rangle^2$$

$$=\frac{1}{N}\left[\left\langle\left(\frac{\partial H}{\partial\beta}\right)^2\right\rangle-\left\langle\frac{\partial H}{\partial\beta}\right\rangle^2\right], \tag{23}$$

where N is the number of states, ϵ', joined to states, ϵ'', by nonvanishing material elements. Since nonvanishing elements occur between oscillator shells differing in the major quantum number of 0 or ±2, we estimate

$$N\approx4\hbar\omega_0 W. \tag{24}$$

The quantity in square brackets is, up to a constant, the mean square fluctuation in the density quadrupole moment. In an independent-particle model, this is the sum of fluctuations for each particle, and these can be calculated easily for the anisotropic harmonic oscillator:

$$\sum_p[\langle(z^2-\tfrac{1}{2}x^2-\tfrac{1}{2}y^2)^2\rangle-\langle z^2-\tfrac{1}{2}x^2-\tfrac{1}{2}y^2\rangle^2]$$
$$\approx\tfrac{1}{3}\sum_p\langle z^4+\tfrac{1}{4}x^4+\tfrac{1}{4}y^4\rangle\approx(3/70)AR^4, \tag{25}$$

where A is the nuclear mass number and R the nuclear radius. Hence

$$(\mathcal{K}_\beta{}^2)_{\text{Av}}\approx\frac{3m^2\omega_0{}^3AR^4}{224\pi\hbar}=\frac{\omega_0V_0B_2}{14\hbar W}, \tag{26}$$

where

$$V_0=\tfrac{1}{2}m\omega_0R^2 \tag{27}$$

is the depth of the nuclear potential and

$$B_2=(3/8\pi)mAR^2 \tag{28}$$

is the irrotational mass parameter for deformations of second order. We set $B_2=B$. This gives, finally,

$$\sigma(\epsilon)\approx(1/7)\pi\omega_0V_0H_s. \tag{29}$$

D. Selection Rules

Axially symmetric deformations of the type assumed here preserve the projection, K, of angular momentum along the nuclear symmetry axis as a good quantum number. The reflectional symmetry, furthermore, preserves the parity, π, of particle states as a good quantum number. The symmetric β-coupling does not mix states of different character (here character means K and π).

Other coupling mechanisms are available to produce mixing of the various K-values and the two parity states. The Coriolis force due to nuclear rotation produces admixing of K-states with the selection rule[3] $\Delta K=\pm1$. Coriolis mixing decreases with increasing deformation, and is further limited by the requirement

$K \leq I$, with I a rigorous constant of the motion. Second-order non-axially-symmetric vibrations can produce admixtures of states differing by $\Delta K = \pm 2$. The parity states can be mixed by reflectionally non-invariant potentials (such as pear-shapes) or oscillations. This in no way violates the invariance of parity for the entire nucleus.

The expressions obtained for the diffusion coefficient and the relaxation time (next section) are for diffusion among the various states of a given character. When explicit expressions are needed for the level density (at high excitation), the tacit assumption is made that sufficiently strong couplings are available to maintain equilibrium among the sets of levels of different character (or at least of different K).

E. Relaxation Time

A relaxation time can be estimated if we take appropriate mean values for the parameters. Consider the mode

$$p(\epsilon,t) = \text{const} \exp(-\alpha t + ik\epsilon). \tag{30}$$

Insertion into (14) yields

$$\alpha = \sigma k^2 - i[W^{-1}\partial(W\sigma)/\partial\epsilon]_{\text{Av}} k. \tag{31}$$

The time dependence shows oscillation as well as decay, but it is only the decay which interests us here. The lowest mode, where the energy range is 0 to E^*, is with

$$k = k_0 = \pi/E^*. \tag{32}$$

The e^{-1} folding time is given by

$$t_r = (\bar{\sigma} k_0^2)^{-1} = E^{*2}/\pi^2\bar{\sigma} = \frac{7}{\pi^3}\frac{E^{*2}}{V_0 \bar{H}_s}\frac{1}{\omega_0}, \tag{33}$$

which is independent of the level density. Note that $\bar{H}_s$ is collective kinetic energy, so we set $\bar{H}_s = \frac{1}{2}E_v = \frac{1}{2}(E^* - \bar{\epsilon})$. We estimate $\bar{\epsilon}$ from the equilibrium distribution. The density of nucleonic levels is given by the statistical formula[8]

$$W(\epsilon) = W_0 \exp[2(a\epsilon)^{\frac{1}{2}}], \tag{34}$$

where $a \approx 12/\text{Mev}$ for heavy nuclei. The mean particle energy is thus

$$\bar{\epsilon} = \int_0^{E^*} \epsilon W d\epsilon \Big/ \int_0^{E^*} W d\epsilon = E^* - (E^*/a)^{\frac{1}{2}} + \cdots. \tag{35}$$

Thus, at high excitations

$$H_s = \tfrac{1}{2}(E^*/a)^{\frac{1}{2}}, \quad [E^* \gg a(\hbar\omega_v)^2], \tag{36a}$$

but at low excitations we must satisfy

$$\bar{H}_s > \tfrac{1}{2}\hbar\omega_v \approx \tfrac{1}{2}\ \text{Mev}, \tag{36b}$$

where ω_v is the collective vibrational frequency. Therefore

$$t_r < \frac{14}{\pi^3}\frac{E^{*2}}{V_0\hbar\omega_v}\frac{1}{\omega_0}, \tag{37a}$$

$$t_r \to \frac{14}{\pi^3}\frac{E^{*\frac{1}{2}}a^{\frac{1}{2}}}{V_0}\frac{1}{\omega_0}, \quad [E^* \gg a(\hbar\omega_v)^2]. \tag{37b}$$

The important thing to note is that the relaxation time is short. Indeed, for $E^* < 9$ Mev, (37a) yields $t_r < \omega_0^{-1}$. This is nonsense, of course, since the relaxation time cannot be shorter than the nucleonic period, which is the minimum time required for information to be transmitted through the nucleus. For a wide variety of reasons, the approximations which lead to (37a) break down for low excitation energy and 9 Mev is a reasonable lower limit for believing the statistical assumptions. However, either (37a) or (37b) give $t_r < \omega_v^{-1}$ for $E^* < 20$ Mev. In general, a mode of wave number $k = \pi/20$ Mev will decay in a time shorter than ω_v^{-1}.

F. Questions of Validity

1. The Collective Description

Although the collective and unified models have had great success in describing the low-lying levels of most nuclei, one might still question the appropriateness of such a description for high excitation. However, recall that the shell-model oscillator frequency is of the order of 6 Mev/$\hbar$ and in heavy nuclei excitation energies even up to 100 Mev do not involve an appreciable fraction of nucleons. Thus we may reasonably expect the collective description to maintain some validity for such energies.

At high excitations one should re-examine the evaluation of the collective parameters B, C (the restoring force constant), and $\omega_v = (C/B)^{\frac{1}{2}}$. The excitation process introduces an effect similar to viscosity which affects the vibrational period even if the parameters B and C are unaffected (and we will not attempt to re-evaluate B and C here).

Some estimate of the viscosity effect can be obtained in the following way. When the probability of making a transition within a collective oscillation period is large, the "path" of the system will tend to diffuse from one particle state to another, changing both its particle and collective energies to maintain constant total energy. The mean particle energy is given by (35) if we replace E^* by $E^* - \frac{1}{2}C\beta^2$. Thus the effective Hamiltonian for collective motion assumes the form

$$\begin{aligned} H_{\text{eff}} &= -\frac{\hbar^2}{2B}\frac{\partial^2}{\partial\beta^2} + \frac{1}{2}C\beta^2 + \bar{\epsilon}(\beta) \\ &\simeq -\frac{\hbar^2}{2B}\frac{\partial^2}{\partial\beta^2} + \frac{1}{(4aE^*)^{\frac{1}{2}}} \times \frac{1}{2}C\beta^2 + \text{const}, \end{aligned} \tag{38}$$

[8] Cf. J. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952), Chap. VIII.

which leads to a vibrational frequency

$$\omega_{v'}=\omega_v(4aE^*)^{-\frac{1}{2}},\quad [E^*\gg a(\hbar\omega_v)^2]. \tag{39}$$

2. Perturbation Theory

The use of first order perturbation theory to determine the real transition rate requires that the virtual (energy nonconserving) transition rate be small by comparison. When the virtual transition rate is large, the representation becomes inconvenient for following the development of the system: certain processes of importance may not be easily describable. For example, in Sec. IV we will discuss the possibility of rapid fracture of the nucleus into two fragments, a process described better in terms of the sudden (impulse) approximation than as a small deviation from adiabatic. Indeed, one might well calculate a very high diffusion rate in terms of the adiabatic representation, but in fact one is only calculating the virtual transitions required to describe the true state, and is throwing away phase relations which could be important in reanalyzing the wave function into the adiabatic representation later.

3. "Real" Transitions in Discrete Spectra

The equation for the transition rate (8) is based on a continuum of (here, collective) final states. The continuum assures the existence of energy-conserving final states, a condition not available in general for discrete spectra. We are therefore faced with energy-nonconserving transitions which are, therefore, not "real," in the sense that they feed back to the original state. Such "virtual" transitions persist a time of the order of $\hbar/\Delta E$, where ΔE is the energy difference between initial and final states and in present context, $\Delta E\sim\hbar\omega_v$. The distinction between real and virtual becomes unimportant if the state has a high probability of making another transition within the period of its virtual existence. This is equivalent to saying that its lifetime-broadening is greater than the level spacing, so that we have, in effect, a continuum. Let τ be the mean lifetime of a state against decay to all other states (as calculated). Then the consistency requirement is

$$\hbar/\tau\gg\hbar\omega_v,\quad \text{or}\quad \omega_v\tau\ll 1.$$

We estimate $(\omega_v\tau)^{-1}$ from the considerations of the previous section:

$$(\omega_v\tau)^{-1}=\omega_v^{-1}\sum_{j''}R(j'';j')\approx\frac{2W(\epsilon)}{\omega_v}\int_{\eta_c}^{\infty}\frac{\eta^2R(\eta)}{\eta^2}d\eta, \tag{40}$$

where η is the energy difference $|\epsilon''-\epsilon'|$ and η_c is a lower cutoff. The factor $\eta^2R(\eta)$ is a reasonably well-behaved falling function of η (except for fluctuations). Let us approximate

$$\eta^2R(\eta)\propto e^{-\eta/\eta_0}, \tag{41}$$

where η_0 is a mean range; the precise form is of little importance for a magnitude estimate. Then it follows that

$$(\omega_v\tau)^{-1}=\frac{2\sigma}{\omega_v\eta_c\eta_0}\approx\frac{2\pi}{7}\frac{\omega_0}{\omega_v}\frac{V_0\bar{H}_s}{\eta_c\eta_0}\sim\frac{120\ (\text{Mev})^2}{\eta_c\eta_0}. \tag{42}$$

The lower cutoff, η_c is given by the closest approach of single particle levels of the same character which, in the Nilsson scheme, is 2 or 3 Mev. Thus the requirements for validity of continuum theory is

$$\eta_0\ll 50\ \text{Mev}. \tag{43}$$

This criterion would seem to be easily satisfied, and is less restrictive than other (albeit less important) criteria which also depend upon η_0.

4. Energy Range of Single Probability Jumps

Although it does not enter explicitly into the calculations, it would be well to obtain some estimate of the range, η_0, of energy differences wherein the transition rate $R(\eta)$ is appreciable. This must be small compared with the wavelength (in energy space) of a relaxation mode in order that the diffusion formulation make sense. Also, the estimate of $(\mathcal{H}_\beta^2)_{\text{Av}}$ was based on the assumption that $\eta_0<2\hbar\omega_0$. Both of these are quantitative questions, rather than questions relating to the general validity of the approach.

The relevant integral

$$\int\phi_{\lambda''}{}^*\mathcal{H}_\beta\frac{\partial\phi_{\lambda'}}{\partial\beta}d\beta$$

is appreciable only so long as $\mathcal{H}_\beta$ is appreciably constant over a distance at least comparable to the reduced length of $\phi_{\lambda''}{}^*(\partial\phi_\lambda/\partial\beta)$,

$$\chi_\beta\sim(2\hbar^2E_v/B)^{\frac{1}{2}}/\eta. \tag{44}$$

This gives an implicit relationship for $\eta=\eta_0$ when χ_β is set equal to the sum of all single particle near-crossings per unit β within a (total) particle excitation of η_0. We do not have careful estimates of η_0 available, but indications are that it is of the order of magnitude of 10 Mev, which satisfies our conditions, although a closer estimate would be desirable.

III. FISSION WIDTHS

We here estimate fission widths in the framework of the unified model. In Fig. 3 the spectrum of nuclear levels for an even-even nucleus is shown as a function of the symmetric deformation parameter, which may be thought of as β. The energy surface is actually many-dimensional in terms of collective parameters describing the various deformations. We may imagine that this curve is a cut through the surface at the minimum values in the energy for the other parameters.

The low-lying energy levels of the system correspond

to the rotational states of a deformed nucleus. The 2+ level is at about 50 kev. Also indicated are the odd parity (1−, 3−, ···) levels. At 1 Mev, single particle excitations appear and the level density increases exponentially with the square root of the energy. As the nucleus is deformed, energy is taken up in the potential energy of deformation. This curve actually is an envelope of nuclear energy levels. A saddle point is reached, after which further deformation gives up energy to either collective or particle modes. Finally scission takes place and the fragments fly apart.

Near the saddle point, the nucleus is relatively cool and strongly deformed, and thus collective states appear. The levels at the saddle point are indicated by diffuse lines to indicate that the nucleus does not remain long at the saddle point and, according to the uncertainty principle, the energies cannot be well defined. For relatively low energies—near threshold—the nucleus does spend appreciably more time at the saddle point than elsewhere.

In the region of the fission saddle point for the jth particle state, let the collective potential energy be given by the inverted parabola

$$\epsilon_j(\beta)=\epsilon_j{}^{\text{th}}-\tfrac{1}{2}B\omega_f{}^2(\beta-\beta_{\text{th}})^2. \tag{45}$$

Hill and Wheeler[4] have shown that the probability of barrier penetration can then be written

$$P_j=\{1+\exp[-2\pi(E^*-\epsilon_j{}^{\text{th}})/\hbar\omega_f]\}^{-1}, \tag{46}$$

where E^* is the total (excitation) energy of the system and $\epsilon_j{}^{\text{th}}$ is the "threshold" energy for the jth particle state. $\hbar\omega_f$ varies considerably from nuclide to nuclide, but is of the order of one Mev, so that the penetrability falls rapidly over a few tenths of a Mev.

There are two times of consequence involved in the determinaton of fission half-widths. One is the collective vibrational period $2\pi/\omega_v$ (or $2\pi/\omega_{v'}$). The other is the relaxation time, t_r, required to establish statistical equilibrium, and here we mean establishing equilibrium from a state of the system which is deficient in the probability that the fissioning channels ($\epsilon_j{}^{\text{th}}\lesssim E^*$) are occupied. This point requires special attention, since the lowest particle states are usually nonstatistical in some way.

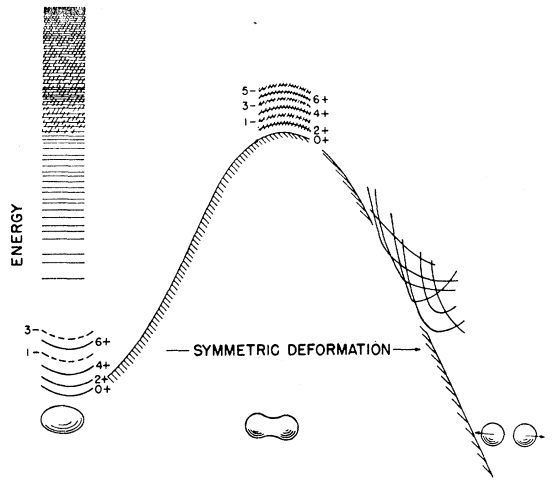

FIG. 3. Schematic representation of level spectrum of an even-even nucleus as a function of the symmetric deformation parameter (β) up to the point of scission. The level structure indicated beyond the saddle point shows crossings. Levels of the same character (K and π) will not cross.

A. Near Threshold

Let us consider the example where a nucleus is excited to some energy where 10^6 particle states are available; for one channel $P=1$, and for all others $P=0$. Further let us assume that the excitation mechanism produces equilibrium, but there is no coupling to maintain equilibrium. The probability is then 10^{-6} that the nucleus will fission promptly—within a time $2\pi/\omega_v$. If the coupling is finite but so small that $t_r\gg 2\pi/\omega_v$, then the fission rate is determined by the relaxation rate, and is equal to $t_r{}^{-1}$. If, however, $t_r\ll 2\pi/\omega_v$, the fission rate is $10^{-6}\ \omega_v/2\pi$. We take up the latter case in more detail first.

Let us assume that $t_r\ll 2\pi/\omega_v$, so that even though the fission channels are depleted in a time $2\pi/\omega_v$, statistical equilibrium is being maintained. Then the fission width is given by

$$\Gamma_f=\frac{\hbar\times(\text{effective No. of open channels})}{(\text{vibrational period})\times(\text{total No. of particle states})}$$
$$=\frac{\hbar}{2\pi/\omega_v}\frac{n}{N}, \tag{47}$$

where the effective number, n, of open channels is the sum over all channels (particle states) weighted by the penetrability P. The total number, N, of particle states is not the grand total of states since, in our representation, each particle state has vibrational states built upon it. Indeed, for large excitations, most of the states one sees are excited vibrational states, and so

$$N=\hbar\omega_v/D, \tag{48}$$

where D is the observed level spacing at the excitation energy E^* and N is, of course, the number of particle states with energy less than E^*. This leads to the expression for the effective number of open channels,

$$n=2\pi\Gamma_f/D, \tag{49}$$

which is identical with the expression first given by Bohr and Wheeler.[4,9,10] For thermal neutrons on fissionable nuclei, one sometimes finds that n, as given by (49), is less than unity, even though the excitation

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

[10] J. A. Wheeler, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955* (United Nations, New York, 1956), Vol. 2; *Proceedings of the International Conference on Nuclear Reactions, Amsterdam, 1956* (Nederlunde Natuurkundige Verenigig, Amsterdam, 1956), p. 1103.

TABLE I. Effective number of open channels in thermal neutron-induced fission as determined from two prescriptions: $2\pi\bar{\Gamma}_f/D$ is the Bohr-Wheeler[a–c] formula based on the magnitude of the fission widths; experimental errors in this column are of the order of 35%. The values of ν are obtained from the Porter-Thomas[b] expression for the statistical distribution of fission widths; these numbers are meaningful only to within a factor of about 2. In both cases, the numerical values are averages per spin state. Because one is dealing with two states ($I_0\pm\frac{1}{2}$, where I_0 is the target spin), the analyses should properly be carried out separately for each. In particular, the Porter-Thomas distribution should be a composite of two distributions[d] characterized by separate sets of parameters $\bar{\Gamma}_f$ and ν. The column labelled π gives the best available guess as to target parity.

	$I_0\pi$	$2\pi\bar{\Gamma}_f/D$	ν
U^{233}	$\frac{5}{2}+$	0.6[e]	4[e]
U^{235}	$\frac{7}{2}-$	0.21[f]	2[f]
Pu^{239}	$\frac{1}{2}+$	0.11[d]	1[d]

[a] See reference 4.
[b] See reference 9.
[c] See reference 10.
[d] Bollinger, Cote and Thomas, *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1959), P/687.
[e] Fluharty, Moore, and Evans, *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1959), P/645.
[f] W. W. Havens, Jr. and E. Melkonian, *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1959), P/655.

energy is a Mev or two over the lowest threshold, and one expects the effective number of channels to be greater than unity. Table I contains values of n determined by the Bohr-Wheeler formula (49) for three nuclides. Also listed are values for the effective number of open channels determined from statistical fluctuations in the fission widths, according to Porter and Thomas.[11] The Porter-Thomas number is generally larger than the Bohr-Wheeler number, but this can be understood by noting that partially open channels ($P<1$) can contribute fully to statistical fluctuations, but only partially to the absolute value of the fission widths.

The paradox of the small number of open channels, which has long been noted, can be resolved reasonably by observing that the coupling is, in all probability, insufficient to assure the condition $t_r\ll 2\pi/\omega_v$. Before delving into this further, let us re-examine an early (and more general) interpretation[8] of (49). h/D is the period of complete nuclear motion, that is, the time required for the nucleus to pass through all modes and return to some original configuration. Thus each open channel decays with the rate of D/h. (The present derivation is consistent with this interpretation.) We see that there is the implicit assumption that all modes are strongly coupled to one another. If, for example, the lowest fissioning channels are not strongly coupled to the other modes, the nucleus will not have equal probability of passing through them during each period D/h.

The lowest particle states are special in several ways. The compound nuclei corresponding to thermal neutron fissioners are even-even, and the lowest fissioning state belongs to the $K=0+$ band. The first 0+ particle excitation is separated from the lowest state by an energy gap[12–14] of at least a Mev, and probably significantly more. Although level density does not appear explicitly in the estimate of relaxation time, it has dropped out by cancellation. The cancellation was effected on the assumption that the level density is a smoothly varying function near the point where flux is calculated. This is not the case at the bottom end of the energy spectrum where the level density goes to zero (and there are no levels below the ground state). The diffusion coefficient has in the numerator a level density factor corresponding to the region across which transitions occur, and in the denominator a level density factor used in computing $(\mathcal{K}_\beta^2)_{Av}$ corresponding to the average over several $\hbar\omega_0$[(23) and (24)]. At the bottom end, the ratio of the level density factors can be very considerably less than unity.†

One must also consider whether there is sufficient coupling to states of different K to maintain equilibrium (see Sec. II.D). Presumably states are excited with all K-values up to $I_0+\frac{1}{2}$, where I_0 is the target spin. This consideration becomes progressively more important with increasing I_0. If only a $K=0$ channel can fission, insufficient coupling would lead to a small fission width. Table I does not, however, indicate any systematic dependence of $2\pi\bar{\Gamma}_f/D$ on target spin.

These effects can be compounded by the failure of the continuum criterion (Sec. II.F.3) when the lifetimes of the states are smaller than the collective level spacings.

It is not surprising, then, that the coupling to the lowest states of an even-even nucleus should be anomalously small, and that $2\pi\bar{\Gamma}_f/D$ overestimates the number of open (defined according to penetrability) channels.

B. High Excitation

At energies which are sufficiently high to employ statistical considerations (a few Mev above threshold), we expect $t_r\ll 2\pi/\omega_v$ for most open channels, and evaluate the number of open channels by the formula

$$n=\int_0^{E^*} W(\epsilon)P(E^*-\epsilon^{\text{th}}-\epsilon)d\epsilon, \qquad (50)$$

where $\epsilon^{\text{th}}=\epsilon_j{}^{\text{th}}-\epsilon_j(\beta\approx 0)$ is assumed to be the same for

[11] C. Porter and R. G. Thomas, Phys. Rev. **104**, 483 (1956).
[12] Bardeen, Cooper, and Schrieffer, Phys. Rev. **108**, 1175 (1957).
[13] Bohr, Mottelson, and Pines, Phys. Rev. **110**, 936 (1958).
[14] C. De Dominicis and P. Martin, Bull. Am. Phys. Soc. **3**, 224 (1958).

† *Note added in proof.*—The lowest lying particle states best fulfill the conditions necessary for enforcement of the selection rule $\Delta\lambda=\pm 1$: the level structure $\epsilon_j(\beta)$ is very nearly harmonic near $\beta=\beta_{\min}$; the character of the states and hence $\mathcal{K}_\beta$ does not change abruptly with β. This means that the partial energy jump during a transition is of order $\hbar\omega_v$, which is about 1 Mev. If the energy gap is appreciably greater than 1 Mev, the diffusion of probability to the lowest particle state should be significantly inhibited.

all particle states, and also represents the lowest threshold energy. By closely approximating the integration in (50), it was found that replacing P by a step function results in only a few percent error at a few Mev above threshold and progressively less at higher energies. This is sufficient for present purposes. Denoting by $N(E)$ the total number of particl states with energy less than E, we obtain

$$\frac{n}{N}=\frac{N(E^*-\epsilon^{\rm th})}{N(E^*)}\simeq\exp\{2[a(E^*-\epsilon^{\rm th})]^{\frac{1}{2}}-2[aE^*]^{\frac{1}{2}}\}; \quad (51a)$$

$$\frac{n}{N}\rightarrow\exp[-\epsilon^{\rm th}(a/E^*)^{\frac{1}{2}}], \quad (E^*\gg\epsilon^{\rm th}) \quad (51b)$$

or

$$\Gamma_f\rightarrow\frac{\hbar\omega_v}{2\pi}\exp[-\epsilon^{\rm th}(a/E^*)^{\frac{1}{2}}], \quad (E^*\gg\epsilon^{\rm th}). \quad (52)$$

The fission width does not increase indefinitely, but rather approaches the limiting value $\hbar\omega_v/2\pi\sim150$ kev. It seems hardly necessary to caution the reader to regard (52) as no more than an order of magnitude estimate. For one thing, we have already seen that at high excitation energies ω_v should be replaced by $\omega_{v'}$ (39).

IV. FROM SADDLE TO SCISSION

Once the nucleus has crossed the saddle point (top of the barrier) configuration, it appears unlikely (although by no means impossible) for it to return to a relatively undeformed state, since most nucleonic states do turn down.

A unique feature which enters the picture beyond the saddle point is the dominance of the Coulomb field. Internucleonic forces are of short range, and a nucleonic period is required to "feel" out the positions of the various nucleons. The Coulomb field, on the other hand, is long range and acts instantaneously (as far as the velocities here are concerned) on all protons. Here, in principle, is a mechanism capable of disrupting the nucleus within a time which could be shorter than either the vibrational or nucleonic periods. The question is not whether the collective velocities are so slow that the motion is adiabatic but rather whether they are so rapid—within a time less than a nucleonic period—that the nucleus is disrupted impulsively. In the latter case, the nuclear "path" follows the independent particle states (dotted lines in Fig. 2; see Hill and Wheeler[4]).

Three nuclear models have been proposed to describe the fission process beyond the saddle point. The first and oldest is the liquid drop model of Bohr and Wheeler.[9] The nuclear motion is assumed to be that of an irrotational, viscosity-free liquid, and in the latter sense the motion is adiabatic.

A second is the statistical model proposed by Fong,[15] and refined by Newton[16] and Cameron.[17] In this model, statistical equilibrium is assumed at the time of scission, and the probability of fissioning into a particular mass distribution is determined by statistical weights alone.

The third model proposed by Bohr[18] and Wheeler,[10] invokes the unified model and would say that certain constraints—K and π—are determined by saddle point considerations. The model has had considerable success with respect to angular distribution of fragments,[19–21] but without further assumptions does not provide a real theory of mass distributions. The model is consistent with almost any descent from the saddle—adiabatic, statistical, or impulsive—so long as the constraints are maintained. (This means K, since π is a "rigorously" good quantum number.)

The quantity of interest is the ratio between the time from saddle to scission and a nucleonic (reduced) period. This is given by

$$\sim(Ad^2/55E_{\rm coll})^{\frac{1}{2}},$$

where $E_{\rm coll}$ is a mean collective kinetic energy in Mev and d is the relative distance in fermis, which the "fragment centers" travel from saddle to scission. When this ratio is appreciably less than unity, impulsive tearing at the saddle is likely to take place. For the thermal fissioners around uranium, $E_{\rm coll}$, which cannot exceed the total non-Coulomb energy available at scission, is <20 Mev. For d a few fermis, the ratio is greater than unity, but not by a large factor.

Experimental indications are that, for low excitations at least, the kinetic energy is independent of excitation energy and probably small in magnitude at scission[22]; this is consistent with statistical equilibrium.

Fairhall and Halpern[23] have emphasized that the fissions occurring with symmetric or asymmetric mass distributions may behave as two distinct "modes," with the symmetric mode increasing with energy more rapidly than the symmetric mode. In some light-heavy nuclei, such as radium, three distinct peaks in the mass distribution have been observed.[22] The relative

[15] P. Fong, Phys. Rev. **102**, 434 (1956).

[16] T. D. Newton, *Proceedings of the Symposium on the Physics of Fission*, Chalk River Report CRP-642-A (unpublished); Atomic Energy of Canada Limited Report AECL-329, 1956 (unpublished), p. 357.

[17] A. G. W. Cameron, *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1959), P/198.

[18] A. Bohr, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955* (United Nations, New York, 1956), Vol. 2, p. 151.

[19] L. Wilets and D. M. Chase, Phys. Rev. **103**, 1296 (1956).

[20] I. Halpern and V. M. Strutinskii, *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1959), P/1513.

[21] J. J. Griffin, Bull. Am. Phys. Soc. **3**, 337 (1958), and Phys. Rev. **116**, 107 (1959).

[22] A comprehensive review of experimental results with theoretical interpretation is given by I. Halpern, *Annual Review of Nuclear Science* [Annual Reviews, Inc., Palo Alto (to be published)], Vol. 59.

[23] A. Fairhall and I. Halpern, University of Washington, Seattle (private communication).

energy dependence of the two modes can be understood in a qualitative way if one is willing to assume that the saddle point favors equal distribution in mass between the undivided lobes, as the simple liquid drop model predicts, while at scission an asymmetric division is favored as far as phase space is concerned, as Fong predicts from shell structure considerations. At low energies, then, statistical equilibrium is expected to obtain for just the reasons given above. At higher energies, however, the collective motion may develop sufficient velocity to produce impulsive tearing soon after the symmetric saddle point is passed, with the result that symmetric fission obtains.

A test of this interpretation of symmetric and asymmetric fission can be sought in the kinetic energies of the fragments. As stated above, low-energy (asymmetric) fissions probably possess small kinetic energy at scission, consistent with equilibrium. If symmetric fission originates in the nonstatistical tearing process, the kinetic energy of such fragments should be considerably larger. This could be detected by separate measurement of the kinetic energies of the fragments from each mode, or by an examination of the mean kinetic energy when the relative intensities of the two modes are varying appreciably with excitation. These questions are currently being investigated by Nicholson and Halpern.[24]

V. ACKNOWLEDGMENTS

The author is indebted to several of his colleagues for helpful discussions on this paper. He is especially grateful to Dr. I. Halpern for extensive and stimulating discussions on fission phenomena; to Dr. S. Rodriguez for valuable suggestions and for a critical reading of the manuscript; and to Dr. J. S. Blair for providing cautious encouragement.

[24] W. Nicholson and I. Halpern, University of Washington, Seattle (private communication).

PHYSICAL REVIEW VOLUME 116, NUMBER 2 OCTOBER 15, 1959

Spallation-Fission Competition in Heavy-Element Reactions: $Th^{232}+He^4$ and $U^{233}+d$*

BRUCE M. FOREMAN, JR.,† WALTER M. GIBSON,‡ RICHARD A. GLASS,§ AND GLENN T. SEABORG
Lawrence Radiation Laboratory and Department of Chemistry, University of California, Berkeley, California

(Received May 18, 1959)

Cross sections and excitation functions have been determined for spallation and fission products from bombardments of Th^{232} with helium ions (15 to 46 Mev) and U^{233} with deuterons (9 to 24 Mev). This work extends a series of investigations of charged particle (α, d, and p) induced reactions in heavy elements ($Z \geq 88$). Radiochemical methods were employed to isolate products corresponding to the following spallation reactions: neutron emission, $(\alpha,4n)$, $(\alpha,5n)$, (d,n), $(d,2n)$, and $(d,3n)$; emission of one proton and neutrons (α,p), (α,pn), $(\alpha,p2n)$, and $(\alpha,p3n)$; and emission of two protons and neutrons, $(\alpha,2p)$, $(\alpha,2pn)$, and $(\alpha,\alpha n)$, and $(d,\alpha n)$. In addition, the following fission products were isolated from one or more bombardments: Zn^{72}, Ge^{77}, As^{77}, $Br^{82,83}$, Rb^{86}, $Sr^{89,91}$, Y^{93}, $Zr^{95,97}$, Nb^{96}, Mo^{99}, $Ru^{103,105,106}$, $Pd^{109,112}$, Ag^{111}, $Cd^{115,115m,117}$, $I^{131,133}$, Cs^{136}, $Ba^{139,140}$, La^{140}, $Ce^{141,143,144}$, Nd^{147}, Eu^{157}, and Gd^{159}.

The results show that fission is the predominant reaction at all energies for Th^{232} and to an even greater extent for U^{233}. The data for the surviving spallation products are consistent with several mechanisms of reaction, including compound-nucleus formation and evaporation, direct interactions between nucleons of the incoming helium ion or deuteron and nucleons of the nucleus, and a combination of these types of processes (direct interaction followed by evaporation). In general, the results confirm and extend previously established concepts.

The neutron-emission spallation reactions as well as fission are best explained as proceeding through compound-nucleus formation. The shapes and magnitudes of $(\alpha,4n)$, $(d,2n)$, and $(d,3n)$ excitation functions correlate well with a compound-nucleus treatment modified to include fission competition. According to this treatment, ratios of neutron to total-reaction level width, $\Gamma_n/\Sigma_i \Gamma_i$, are 0.49 for $U^{236-233}$ [from $Th^{232}(\alpha,4n)$], 0.17 for $Np^{235-234}$ [from $U^{233}(d,2n)$], and 0.20 for $Np^{235-233}$ [from $U^{233}(d,3n)$]. In addition the total-reaction excitation functions (consisting mostly of the fission excitation functions) are consistent with theoretical cross sections for compound-nucleus formation calculated with a nuclear radius parameter $r_0=1.5\times10^{-13}\,A^{\frac{1}{3}}$.

The fission mass-yield curves are similar to those found for other heavy target isotopes (for elements from thorium to plutonium). The minimum in the curves in the region of mass 120 tends to disappear as helium-ion or deuteron energy is increased.

The (α,pxn), $(\alpha,2pxn)$, $(\alpha,\alpha n)$, (d,n), and $(d,\alpha n)$ products are attributed to direct interactions, with complex particles emitted in preference to a series of protons and neutrons. Thus (α,d), (α,t), and (α,tn) mechanisms would account for most of the (α,pn), $(\alpha,p2n)$, and $(\alpha,p3n)$ products, respectively. In the case of the (α,t) and (α,tn) reactions, analysis of the ratio $\sigma(\alpha,tn)/\sigma(\alpha,t)$ leads one to the conclusion that with 35-Mev helium ions only 9% of outgoing tritons leave the residual nucleus with sufficient energy to evaporate a neutron or undergo fission, and with 44-Mev helium ions only 20% do so. The (d,n) product probably results from the stripping reaction.

* This work was performed under the auspices of the U. S. Atomic Energy Commission. It is based in part on the Ph.D. theses of Bruce M. Foreman, Jr., University of California, June, 1958, and Walter M. Gibson, University of California, June, 1957. One of us (W. M. G.) wishes to acknowledge the support of the U. S. Air Force Institute of Technology during this research.

† Present address: Brookhaven National Laboratory, Upton, New York.
‡ Present address: Bell Telephone Laboratories, Murray Hill, New Jersey.
§ Present address: Stanford Research Institute, Menlo Park, California.