Transition State Rates and Complex Fragment Decay Widths

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Experimental excitation functions for the emission of complex fragments from compound nuclei are analyzed to search for atomic number Z and energy E dependent deviations from transition-state-method predictions. No Z and/or E dependent effects that could be attributed to an increased collectivity with increasing mass (charge) of the emitted fragment and associated with transient or stationary solutions of Kramers' diffusion equation are visible. Over seventy excitation functions, for complex fragments from four different compound nuclei, can be collapsed into a single universal straight line that is rigorously consistent with the transitions-state predictions.

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The rates for fission decay, as well as for chemical reactions, are calculated most often by means of the transition-state method [1]. In this approach, the reaction rate is equated to the flux of phase space density across a "suitably" located hyperplane normal to the "reaction coordinate." The suitable location is typically chosen at a saddle point in collective coordinate space, which corresponds to a bottleneck in phase space. A smart choice of the transition state location should minimize the number of phase space trajectories doubling back across the hyperplane.

The surprising success of the transition-state method in many subfields of physics and chemistry [2,3] has prompted attempts to justify its validity in a more fundamental way, and to identify regimes in which deviations might be expected. In particular, the intense debate regarding whether the observed prescission particle emission (n, p, α, and γ) should be interpreted as presaddle or postsaddle emission has a strong and direct bearing on the validity of the transition-state fission rates. In what follows we shall compare experimental decay rates for complex fragment emission with transition state predictions, and search for E and Z dependent deviations that can be expected to exist.

The transition state expression for the fission decay width is

$$\Gamma_f = \frac{1}{2\pi\rho(E)} \int \rho^*(E - B_f - \varepsilon)\, d\varepsilon \approx \frac{T_f}{2\pi} \frac{\rho^*(E - B_f)}{\rho(E)}, \quad (1)$$

where $\rho(E)$ is the level density of the compound nucleus, $\rho^*(E - B_f - \varepsilon)$ is the level density at the saddle point, B_f is the fission barrier, ε is the kinetic energy over the saddle along the fission coordinate, and $1/T_f = \partial[\ln\rho^*(x)]/\partial x|_{E-B_f}$.

For the one dimensional case, in which the only degree of freedom treated explicitly is the reaction coordinate, the decay width takes the form

$$\Gamma_f = \hbar\omega \frac{\rho^*(E - B_f)}{\rho^*(E)} \approx \hbar\omega e^{-B_f/T}, \quad (2)$$

where T is the temperature of the compound nucleus. Now both level densities correspond to the same number of degrees of freedom. The quantity $\hbar\omega$ is the oscillator phonon associated with the ground state minimum. In this simplest formulation, one can read the reaction rate in terms of its two factors: the frequency ω which gives the free rate of assault to the barrier and the Boltzmann factor which gives the probability per assault of making it over the barrier.

The emission of complex fragments can be treated in an analogous fashion by introducing the ridge line of conditional saddle points [4]. Each mass or charge emission can be associated with a conditional barrier that can be measured with techniques similar to those used to determine fission barriers [5]. Recently, nearly complete ridge lines have been determined for several nuclei: ^{75}Br [6], ^{90,94}Mo [7], and ^{110,112}In [5].

The emission rate of a fragment of a given mass or charge can still be described by an expression similar to that of Eq. (2). The quantity B_f becomes the conditional barrier B_Z, but what is now the meaning of $\hbar\omega$? Is there a single value of $\hbar\omega$ for all the channels or does each channel have its own characteristic frequency? In what follows, we shall endeavor to answer this question experimentally.

An additional aspect of the problem has been studied by Kramers in his seminal work [2]. Kramers considered the diffusion of the system from the reactant's region to the product's region from the point of view of the Fokker-Planck equation. The new parameter entering the problem is the viscosity coefficient, which couples the reaction coordinate to the heat bath. The stationary current solution found by Kramers leads to expressions for the reaction rates similar to that of the transition-state theory, differing only in the preexponential factor, which now includes the viscosity. More recent work has shown that if the system is forced to start at time $t = 0$ at the ground state minimum, a transient time τ_f exists during which the reaction rate goes from zero to its stationary value [8–14]. Both effects would decrease the overall fission rate compared to the transition state prediction.

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These transient effects have been advocated as an explanation for the large number of prescission neutrons [15–19], charged particles [20–24], and electric dipole γ rays [25,26] observed in the fission of many systems, in apparent contradiction with the predictions of the transition-state method. However, the prescission particles can be emitted either before the system reaches the saddle point, or during the descent from saddle to scission. Since only the former component has any bearing on possible deviations of the fission rate from its transition state value and the separation of the two components is fraught with difficulties, the experimental evidence is ambiguous. Furthermore, for some systems, the measured prescission charged particle multiplicities are consistent with statistical model calculations [27].

Recently, it has been suggested that the viscosity and the transient time may depend on the collectivity of the reaction coordinate [28]. More specifically, the reaction coordinate for a very asymmetric decay should have little collectivity, while that for a symmetric decay should be very collective. Studies of prescission particles as a function of the size of the emitted fragment claim to have observed such an effect [17,18,28,29]. However, a statistical model, incorporating mass-asymmetry-dependent fission delay times, could not simultaneously reproduce the prescission neutron multiplicities and the charge distributions [30].

In this Letter, we show that the presence or absence of the effects discussed above can be directly observable in the excitation functions for the emission of fragments with different Z values. This new technique can be used to search for systematic deviations from transition state predictions that would indicate the existence of a transient effect.

Our procedure uses the transition-state-method prediction as a null hypothesis, and involves only replotting experimental data without invoking a specific model. The cross section for the emission of a fragment of a given Z value can be written as

$$\sigma_Z = \sigma_0 \frac{\Gamma_Z}{\Gamma_T} = \sigma_0 \frac{\Gamma_Z}{\Gamma_n + \Gamma_p + \cdots}, \quad (3)$$

where σ_0 is the compound nucleus formation cross section and $\Gamma_T, \Gamma_n, \Gamma_p, \Gamma_Z$ are the total, neutron-, proton-, and Z-decay widths, respectively. Notice that Γ_T is essentially independent of Z if we confine our observations to the excitation energy region where the complex fragment emission probability is small.

We now rewrite Eq. (3) as follows:

$$\frac{\sigma_Z}{\sigma_0}\,\Gamma_T \frac{2\pi\rho(E - E_r^{gs})}{T_Z} = \rho^*(E - B_Z - E_r^s), \quad (4)$$

where T_Z is the temperature at the conditional saddle point, E_r^{gs} is the energy of the rotating ground state, and E_r^s is the saddle point rotational energy. In this way, the left-hand side of the equation contains the complex fragment cross section which can be measured, and other calculable quantities that do not depend on Z, except T_Z which is only weakly dependent. The right-hand side contains only the level density at the conditional saddle calculated at the intrinsic excitation energy over the conditional saddle, which is calculable if the barrier height is known.

By using the standard Fermi gas level density expression, one can rewrite Eq. (4) in the following way which takes out the A dependence of the level density:

$$\ln\left[\frac{\sigma_Z}{\sigma_0}\,\Gamma_T \frac{2\pi\rho(E - E_r^{gs})}{T_Z}\right] \Big/ 2\sqrt{a_n} = \frac{\ln R_f}{2\sqrt{a_n}} = \sqrt{\frac{a_Z}{a_n}(E - B_Z - E_r^s)}, \quad (5)$$

where a_Z, a_n are the saddle and ground state level density parameters. A plot of the left-hand side of this equation vs the square root of the intrinsic excitation energy over the saddle should give a straight line, and the slope should give the square root of a_Z/a_n.

Recently, the excitation functions for a large number of fragment Z values have been measured for the following systems: ^{75}Br [6], ^{90,94}Mo [7], and ^{110,112}In [5]. The corresponding conditional barriers have been extracted [5–7]

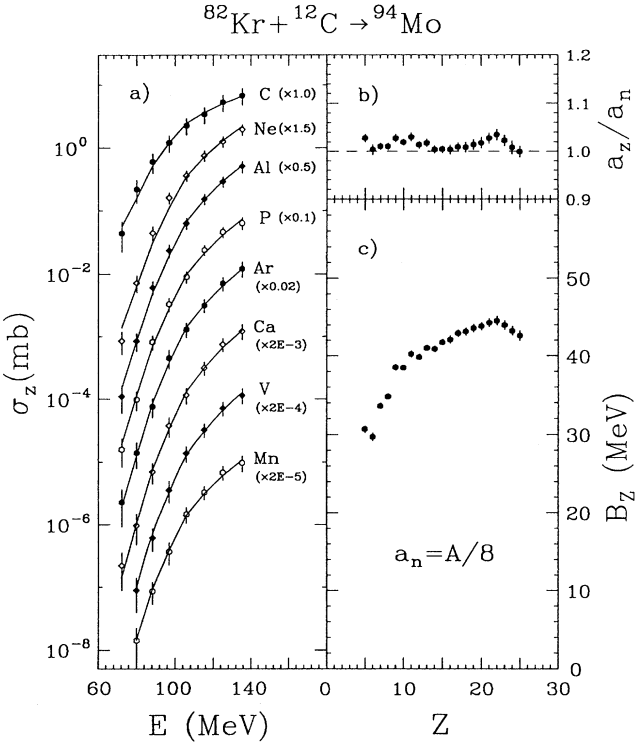

FIG. 1. (a) Excitation functions for representative complex fragments emitted from the compound nucleus ^{94}Mo. (b) The a_Z/a_n values and (c) conditional barriers B_Z, extracted by fitting the excitation functions with a transition-state formalism. The solid lines in (a) correspond to the fit using an energy level parameter $a_n = A/8$. Statistical error bars are shown when they exceed the size of the symbols.

by fitting the excitation functions with the transition-state formalism. A level density parameter $a_n = A/8$ was assumed. The conditional barrier B_Z and the level density parameter ratio a_Z/a_n were the only free parameters in the fitting. As an example, Fig. 1(a) shows excitation functions for representative fragments with Z values from 5 to 25 for the compound nucleus ^{94}Mo. The solid lines in Fig. 1(a) correspond to the best fit to the experimental data. The energy of the rotating ground state E_r^{gs} was calculated with the rotating finite-range model by Sierk [31] and the rotational energy of the saddle E_r^s was calculated assuming a configuration of two nearly touching spheres separated by 2 fm. Using the maximum angular momentum obtained from the best fit to the excitation functions, these values of ℓ_{max} agree with Bass model [32] predictions within $2\hbar$, one can calculate $\langle \ell^2 \rangle = \ell_{max}^2/2$ and then the averages of E_r^{gs} and E_r^s accordingly. The extracted ratios a_Z/a_n are close to unity for all Z values [see Fig. 1(b)]. The extracted conditional barriers increase from 30–45 MeV as the charge of the emitted fragment increases [see Fig. 1(c)].

Equation (5) suggests that it should be possible to reduce *all* the excitation functions for the emission of different complex fragments from a given system to a single straight line. In Fig. 2 all the excitation functions associated with each of four compound nuclei are plotted according to Eq. (5). There are 20, 21, 21, and 9 excitation functions for ^{75}Br, ^{90}Mo, ^{94}Mo, and ^{110,112}In, respectively. We see that all the excitation functions for each Z value fall with remarkable precision on a single line which is in fact straight, has a slope near unity, and passes closely through zero.

The sensitivity of the excitation functions to the mass A of the compound nucleus is vividly shown in the Fig. 3(a), where the logarithm of the reduced mass-asymmetric fission rate $\ln R_f$ is plotted vs the square root of the intrinsic excitation energy for $Z = 10$ and for four different compound nuclei. The excitation functions are straight lines, but with different slopes for different compound nuclei. After the A dependence is removed, as suggested in Eq. (5), the four lines collapse into a single straight line [see Fig. 3(b)]. Similar results are obtained for all the other Z values. We find this delicate sensitivity to the mass of the compound nucleus truly remarkable.

The normalized intercepts of the straight line fits for each Z value and for all four compound nuclei do not show a statistically significant correlation with Z value. For example, the linear correlation coefficient determined by Pearson's method is 0.1, where a value close to ± 1 would indicate a linear correlation [33]. This suggests that the quantity $\hbar\omega$ appearing in Eq. (2) does not depend on the Z value of the emitted fragment.

As a final virtuoso touch, we can try to collapse *all* the excitation functions for *all* Z values and for all compound nuclei into a single straight line. The resulting plot for four different compound nuclei is shown in Fig. 4. It includes a total of 71 excitation functions, for fragments ranging in Z from 3 to 25. The collapse of all the

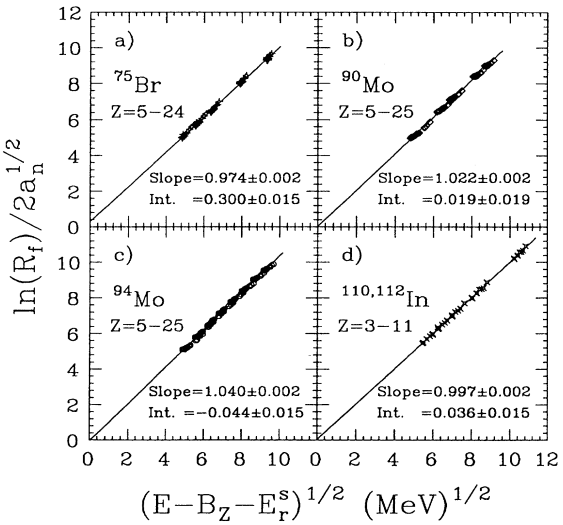

FIG. 2. The logarithm of the reduced mass-asymmetric fission rate R_f as defined in Eq. (5) divided by $2a_n^{1/2}$ vs the square root of the intrinsic excitation energy for four compound nuclei: ^{75}Br (a), ^{90}Mo (b), ^{94}Mo (c) and ^{110,112}In (d). The solid lines are the linear fits to the data. The error bars are smaller than the size of symbols.

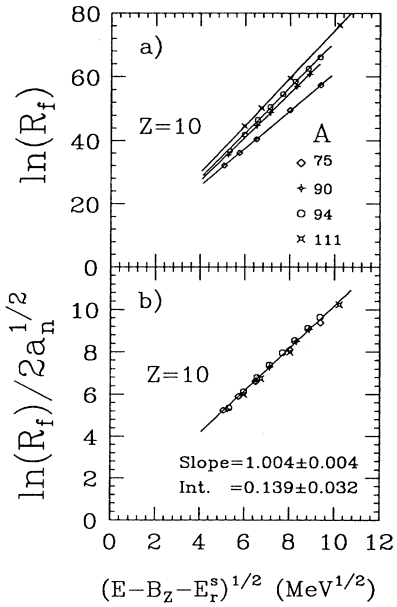

FIG. 3. (a) The quantities in $\ln R_f$ and (b) $\ln R_f$ divided by $2a_n^{1/2}$ vs the square root of the intrinsic excitation energy for $Z = 10$ fragments emitted from the compound nuclei: ^{75}Br, ^{90}Mo, ^{94}Mo, and ^{110,112}In. The A dependence of R_f via a_n is shown in (a), while this A dependence is removed in (b). See Fig. 2.

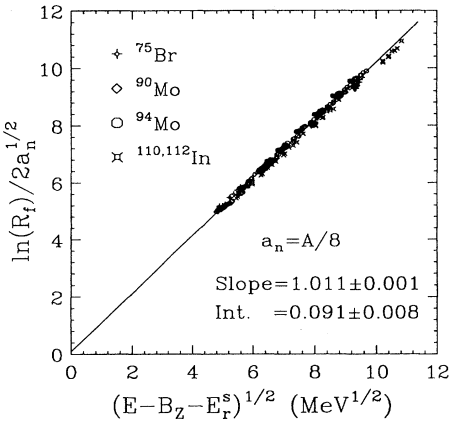

FIG. 4. The same as Fig. 2 with the data for all four nuclei shown in a single plot. The straight line is the linear fit to all the data points. See Fig. 2.

experimental excitation functions for all the systems onto a single straight line is strong evidence for the validity of the transition state formalism and for the absence of Z and E dependent deviations.

The present analysis shows unequivocally that there are no deviations from the transition-state formalism in the explored range of excitation energies and charges. The estimated compound nucleus lifetimes range from $\tau_{CN} \approx 3 \times 10^{-20}$ sec at the lowest excitation energies to $\tau_{CN} \approx 1 \times 10^{-21}$ sec at the highest excitation energies. This range is to be compared with transient times $\tau_{tr} \leq 10^{-19}$ sec that have been inferred from prescission particle evaporation. In particular, one is led to the following conclusions. (i) Once one removes the phase space associated with the nonreactive degrees of freedom at the conditional saddle point, the reduced rates are *identical* for fragments of all Z values. Within the experimental sensitivity, the quantity $\hbar\omega$ in Eq. (2) appears to be Z independent. (ii) For all fragments, there is no deviation from the expected linear dependence over the excitation energy range from 60–140 MeV. This seems to rule out, for all Z values, transient time effects which should become noticeable with increasing excitation energy. (iii) The slope, which corresponds to the $\sqrt{a_Z/a_n}$, is essentially unity for all Z values of all systems studied. (iv) The intercept of the straight line, which is associated with the channel frequency ω, is essentially zero and shows no obvious dependence on the fragment Z values (i.e., the collectivity).

We conclude that in this extended data set there is no evidence for transient effects either directly or through their expected dependence upon the mass of the emitted fragment. Furthermore it appears that the channel frequency is the same for all the different Z decay channels.

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