Why temperature-dependent fission barriers should not be included in statistical model calculations

R. J. Charity
Department of Chemistry, Washington University, St. Louis, Missouri 63130
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The reduced nuclear density in the surface layer of a nucleus gives rise to an increase in the level density parameter and also makes the fission barrier of the nucleus temperature dependent. The manner in which these effects can be consistently incorporated in the transition state formalism for statistical model calculations is discussed. The naive replacement of zero temperature with temperature-dependent fission barriers in the standard formula to obtain the level density at the saddle-point configuration is incorrect.

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In statistical model calculations, fission decay rates are usually determined from the transition state formalism. The important quantity in this formalism is the level density at the saddle-point configuration, which is usually calculated assuming a Fermi gas form, where the appropriate thermal excitation energy is reduced from the value for the ground state by the fission barrier. Theoretical studies [1–5] have also addressed the dependence of fission barriers on temperature, concluding that they are expecting to decrease with increasing temperature. Newton, Popescu, and Leigh [6] have taken such temperature-dependent fission barriers and included them in the standard Fermi gas formula for the saddle-point level density. More recently, Hofman, Back, and Paul [7], in one of their statistical model calculations, have also included a temperature-dependent fission barrier. Other studies have also briefly mentioned temperature-dependent barriers as a possible reason for the low fission barriers needed to fit data [8,9] or as a possible way of enhancing intermediate fragment production [10].

The purpose of this Brief Report is point out that, for the determination of the thermal excitation energy at the saddle-point configuration, the naive replacement of the zero-temperature fission barrier with the temperature-dependent value is incorrect. Specifically, theoretical temperature-dependent fission barriers are determined from the difference between the Helmholtz free energy at the ground-state and at the saddle-point configuration. This free energy would be appropriate for determining the potential energy surface for isothermal processes. However, the thermal excitation energy at the saddle point is the energy above the zero-temperature saddle-point energy (ground-state energy plus zero-temperature fission barrier) and not the value above the saddle-point free energy.

To further elucidate these points, less us briefly reconsider the basic results of the Fermi gas model [11]. The level density is approximately proportional to e^S, where S is the entropy. The entropy and the thermal excitation energy U are related to the temperature T by the level density parameter a: $S=2aT$ and $U=aT^2$. The total energy of a nucleus, $E(T,\epsilon)$, can be separated into its dependence on temperature and deformation ϵ by

$$E=E(0,\epsilon)+U \tag{1}$$

$$=E(0,\epsilon)+aT^2. \tag{2}$$

Now the free energy of the nucleus is given by

$$F=E-TS \tag{3}$$

$$=E(0,\epsilon)-aT^2. \tag{4}$$

Numerous theoretical studies have considered the effect of the reduced nuclear density in the surface layers of a nucleus on the level density parameter [2,3,12–15]. In general, the level density parameter can be written as a sum of volume and surface terms:

$$a(\epsilon)=\alpha_v A+\alpha_s B_s(\epsilon)A^{2/3}, \tag{5}$$

where $B_s(\epsilon)$ is the liquid drop model quantity giving the ratio of the surface area at deformation ϵ to that for a sphere ($\epsilon=0$), A is the nucleon number of the nucleus, and α_v and α_s are both constants. For simplicity, the curvature term will be ignored in this discussion; however, the conclusions of this work will not be altered if one were also to consistently include curvature terms in the free energy and other quantities.

As the deformed saddle-point configuration has a larger surface area than the spherical or near-spherical ground-state configuration, the free energy in Eq. (4) decreases faster with temperature at the saddle-point than at the ground-state configuration. Hence, the fission barrier defined in terms of the difference in the free energies is temperature dependent. However, the thermal excitation energy is still determined from Eq. (2) and hence the entropy at the saddle-point deformation ϵ_{SP} is

$$S=2\,a(\epsilon_{SP})T=2\sqrt{a(\epsilon_{SP})[E-E(0,\epsilon_{SP})]}. \tag{6}$$

Note that $E(0,\epsilon_{SP})$ is the zero-temperature saddle-point energy which is determined from the zero-temperature fission barrier.

Now, the saddle-point deformation is determined from the balance between the cohesive surface force and and the disruptive Coulomb force. The change in the surface free energy of the nucleus with temperature changes this balance and one should allow for a temperature dependence of the saddle-point deformation $\epsilon_{SP}(T)$. However, the quantity $E(0,\epsilon_{SP}(T))$ does not change with temperature to first order, as by definition, the saddle-point energy sits at a maxima as a function of the fission deformation parameter. Thus the effect of the reduced nuclear density in the surface layer, which is primarily responsible for the theoretical temperature-dependent barriers, is incorporated in the saddle-point level density formula via the level density parameter $a(\epsilon_{SP})$.

In most statistical model analyses of fission excitation functions, the quantity a_f/a_n, the ratio of level densities parameters for the saddle-point and ground-state configurations [which can be identified with ratio $a(\epsilon_{SP})/a(0)$ in the above discussion], is often used as a fit parameter. If a_f/a_n values greater than unity are obtained from such fits, then this would be consistent with the theoretical decrease of the fission barrier (as defined from the free energies) with temperature. However, fission barriers extracted from fitting fission excitation functions are zero-temperature barriers.

Some minor modifications to the temperature dependence of the fission barriers and the level density at the saddle point can result from the predicted expansion of nuclear matter and increase in surface diffuseness with temperature. One consequence of this is that the Coulomb energy of the nucleus becomes temperature dependent. To lowest order, this effect can be included by adding a Coulomb term (deformation dependent) to the level density parameter when used in Eq. (2). Note, however, that the change in Coulomb energy modifies the total and free energies by the same amount, and so when calculating the entropy from the temperature, this Coulomb term should not be included in the level density parameter. In any case, the magnitude of such terms is minor compared to the surface and volume terms [2,3].

For saddle-point shapes which have prominent necklike features, the consideration of the finite range of the nuclear force is important. Fission barriers calculated for such saddle-point shapes are reduced relative to predictions of the liquid drop model when this effect is included [16]. The magnitude of this finite range correction may be modified by the increase in the surface diffuseness. Although some theoretical studies have included the above effects, only temperature-dependent fission barriers are presented. Unfortunately, the entropy at the saddle-point configuration cannot be obtained from these barriers without further information. In the future it would be useful if such studies present the dependence of the saddle-point entropy on the excitation energy.

The small modifications to the saddle-point entropy from the changes in nuclear density and surface diffuseness may be difficult to investigate experimentally, especially if the level density parameter itself is temperature dependent as suggested by some experiments [17,18]. Theoretically, such an effect can be obtained from a temperature dependence of the effective mass. Prakash, Wambach, and Ma [19] argue that the frequency-dependent (ω) effective mass considerably enhances the value of α_s. However, this ω effective mass is predicted to decrease with temperature [20–22] and thus the deformation dependence of the level density parameter will depend on the temperature and the ratio a_f/a_n should decrease with increasing temperature.

In conclusion, a deformation dependence of the level density parameter implies that fission barriers calculated from the difference in free energies between the saddle-point and ground-state configurations are temperature dependent. However in statistical model calculations, when determining the saddle-point level density with the standard Fermi gas expression, one should not calculate the thermal excitation energy using the above temperature-dependent fission barriers. Future studies of the fission probability should consider both the temperature and deformation dependence of the level density parameter.

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