New Type of Shape Instability of Hot Nuclei and Nuclear Fragmentation

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A novel mechanism of nuclear fragmentation is proposed. Assuming microcanonical equilibrium, it is shown that a strong enhancement of the accessible volume of the phase space due to the diffuseness of nuclear surface leads to dynamical instabilities of hot nuclei and to a fragmentation. Equations are derived for the transition temperature T_T for which the thermodynamical surface tension vanishes, as well as for the thermodynamical fissility parameter χ_{td}. [S0031-9007(99)09391-6]

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The understanding of properties and the behavior of hot nuclear matter, apart from its general scientific merit, is of key importance in studies of nuclear multifragmentation [1–3]. The latter studies have produced experimental evidence that, under stress generated by heavy-ion collisions, nuclei fragment into multiple pieces—intermediate-mass fragments. At the same time, theoretical effort has been undertaken to establish the nature of the stress necessary or sufficient for the loss of (shape) stability of finite nuclei. While theoretical modeling of thermostatic properties of finite nuclear matter [4] has led to the realization that above a certain critical temperature, $T_{\rm cr}$, nuclear matter cannot exist in its basic liquid phase, most models of nuclear multifragmentation [5–10] rely on the presence of some dynamical stimulus in addition to a purely thermal one. This is so because the predicted magnitude of $T_{\rm cr}$ appears to be significantly higher than the experimentally determined temperatures of multifragmenting systems. For example, calculations assuming Skyrme interactions predict $T_{\rm cr}$ in the range of 13–20 MeV for infinite matter. While for finite systems theoretical values of $T_{\rm cr}$ are significantly lower than those for infinite matter, they are still substantially higher than the experimentally observed "multifragmentation temperatures" of 4–5 MeV.

The present paper points out the existence of an effect that could lead to the loss of macroscopic stability of finite nuclei at excitation energies of a few MeV per nucleon even in the absence of dynamical (compressional, inertial) stimuli. Its findings derive from a realization of the importance of surface effects in thermodynamics of hot nuclei.

To demonstrate the essence of the new mechanism, a schematic model is adopted in which an excited nuclear system is allowed to assume one of only two macroscopic configurations (phases), that of a spherical mononucleus and that of a dinuclear configuration of two identical touching spheres. It is further assumed that the system is in microcanonical equilibrium, i.e., all microstates belonging to the allowed macroscopic configurations are populated with equal probabilities. Additionally, to simplify the calculations, it is assumed that the two constituents of the dinuclear configuration have approximately equal excitation energies

$$E_1^* = E_2^* = \frac{1}{2}\,(E_{\rm tot}^* - E_{\rm pot})\,, \tag{1}$$

where $E_{\rm tot}^*$ is the total excitation energy of the system and $E_{\rm pot}$ is the potential energy of the dinuclear configuration relative to the ground state of the mononuclear configuration ($E_{\rm pot} = 0$). The quantity $E_{\rm pot}$ can be calculated based on the ground-state binding energies of the spherical nuclei involved in both types of configurations and on the Coulomb repulsion energy of the dinuclear complex.

The role of the nuclear surface is described in the present model by the nuclear mass tables, by the liquid-drop mass formula, and by the surface term in the Fermi-gas model expression [11–13] for the level density parameter a. It is the latter term that leads to the effects discussed:

$$\begin{aligned} E &= E_V + E_S + E_C(\text{shape}) \\ &= \epsilon_V A + \epsilon_S A^{2/3} F_2 + E_C(\text{shape}) \quad \text{and} \end{aligned} \tag{2}$$

$$a = a_V + a_S = \alpha_V A + \alpha_S A^{2/3} F_2\,, \tag{3}$$

where A is the atomic number, $E_C(\text{shape})$ is the shape-dependent Coulomb energy, and F_2 is the surface area in units of its value for the spherical shape.

In microcanonical equilibrium, macroscopic states of the system are populated according to weight factors that can be expressed as $W_k \propto e^{S_k}$, where S_k is the entropy of the system in the kth macroscopic configuration. Within the Fermi-gas model, the entropy for the two allowed configurations can be approximated as

$$S_m = 2\sqrt{a_m E^*} \quad \text{and} \tag{4}$$

$$S_d = 4\sqrt{a_d (E^* - E_{\rm pot})/2}\,, \tag{5}$$

where subscripts m and d identify the mono- and dinuclear configurations, respectively, and the level density parameters are calculated from Eq. (3) for the mass number A (mononuclear) and for the mass number $A/2$ (dinuclear). In Eq. (5), the small contribution to the entropy from the degrees of freedom of relative motion of the constituents of the dinuclear complex has been neglected.

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The results of schematic calculations for a hypothetical nucleus with $a = 200$ and $Z = 80$ are shown in Fig. 1. In these calculations, a potential energy of $E_{\rm pot} = 62$ MeV was assumed, based on the nuclear mass tables and the Coulomb interaction of two point nuclei of charges $Z/2$, separated by a distance of $d = 2.6(A/2)^{1/3}$ fm. Note that the assumed potential energy is significantly higher than the actual saddle energy for this system. The use of such a high value of $E_{\rm pot}$ in the schematic calculations allows one to better illustrate the large magnitude of the discovered effect. For the level density parameter a, the parametrization of Tõke and Swiatecki [11] was employed with $\alpha_V = 0.068$ MeV^{-1} and $\alpha_S = 0.274$ MeV^{-1}.

As seen in the top panel of Fig. 1, at low total excitation energies, the system achieves the highest entropy when it assumes the mononuclear configuration. In other words, the accessible volume of the phase space is larger for the mononuclear than for the dinuclear configuration. However, the accessible phase space volume is enhanced due to the surface diffuseness of the nuclear matter distribution [reflected in the surface term in Eq. (2)]. This accessible volume grows faster with increasing total excitation energy for the dinuclear than for the mononuclear configuration. Eventually, at an excitation energy of $E^*/A \approx 3.3$ MeV, the two allowed configurations fill equal phase space volumes, i.e., correspond to equal entropies. Above this "crossover energy" the system has a higher entropy in the dinuclear state. Obviously, driven by Coulomb repulsion, the latter configuration will decay dynamically.

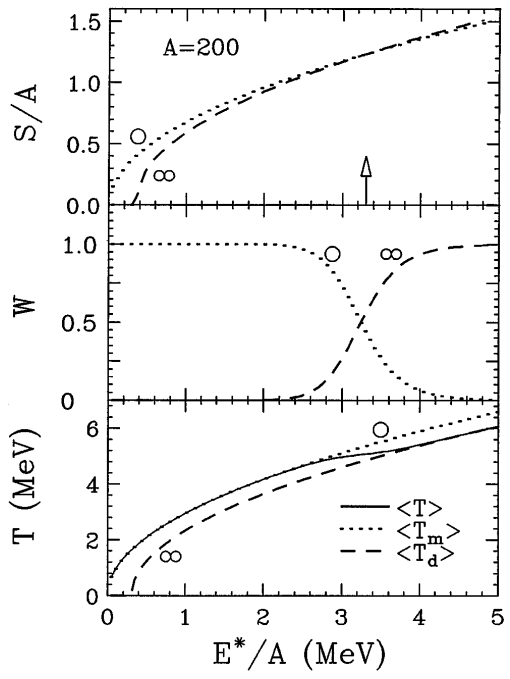

FIG. 1. Entropy per nucleon (top), normalized microcanonical population probability (middle), and temperature (bottom) are plotted vs total excitation energy per nucleon. Two competing geometries of a nuclear system of $A = 200$, $Z = 80$ are illustrated, a mononuclear (single circle) and a dinuclear (touching circles) configuration. The solid line in the bottom panel represents the weighted average temperature of the system, while the dotted and dashed lines illustrate the temperatures for the pure mononuclear and dinuclear configurations, respectively. See text.

The middle panel of Fig. 1 illustrates the dependencies of the normalized microcanonical weight factors $W_{\rm mc}$ for the mononuclear and the dinuclear configuration on the total excitation energy. A second-order phase transition from the mononuclear to the dinuclear phase is seen to occur in the smooth, gradual manner characteristic of small systems. This figure demonstrates that the present schematic system cannot survive in a microcanonically equilibrated mononuclear configuration when excited to energies in excess of 4 MeV/nucleon.

The bottom panel in Fig. 1 illustrates the predicted relation (solid line) between average temperature and total excitation energy of the system, i.e., the "caloric curve" for the system. The average temperature is defined as $T = W_m T_m + W_d T_d$, where W_m, W_d, T_m, and T_d are the microcanonical weight factors and average system temperatures for the mononuclear and dinuclear configurations, respectively. The latter temperatures are calculated from the Fermi-gas model relationship $E^* - E_{\rm pot} = aT^2$. For comparison, the temperatures T_m and T_d are also shown. As expected for a microcanonical system, the temperature is not a sharply defined quantity. For any excitation energy, the two-phase system assumes two different temperatures, with probabilities given by the weight functions depicted in the middle panel in Fig. 1. In the caloric curve, the mononuclear-to-dinuclear phase transition shows up as a quasiplateau around $E^*/A \approx 3.3$ MeV. This quasiplateau should not be confused with a plateau expected for a first-order phase transition such as, e.g., liquid-to-gas transition.

A very similar behavior is obtained when a canonical rather than a microcanonical equilibrium is considered for the present system. In that case, the transition from the mononuclear to the dinuclear phase is expected to occur at $T \approx 5$ MeV [14]. While a microcanonical description appears better suited [6] for isolated nuclear systems than a canonical one, the present schematic model does not reveal any qualitative or major quantitative differences in the behavior of the system in these two approximations.

It is remarkable that already an excitation energy of the order of 4 MeV/nucleon, corresponding to an average temperature of less than 6 MeV, is sufficient for the system to overcome a potential barrier of over 60 MeV. This should not be surprising when one realizes that the mechanism that allows the system in the present case to overcome a large potential barrier is fundamentally the same as that causing thermal expansion of nuclear matter. For example, in a schematic model such as the expanding emitting source model (EESM),

[7] the thermal pressure that causes the system to expand arises as a result of a strong dependence of the level density parameter a on the nuclear matter density ρ, $a = a_o(\rho/\rho_o)^{-2/3}$, where ρ_o is the ground-state nuclear matter density. In the EESM [7], this thermal pressure is equivalent to potential energies in the compressional degree of freedom of hundreds of MeV, already for temperatures below 10 MeV. Hence, in both cases, a shape-instability (considered in the present schematic model) and a density-instability (considered in the EESM [7]), it is the dependence of the level density parameter a on the "driven" observable (shape and ρ, respectively) that generates large effective thermodynamical driving forces and the associated destabilizing potential energies. In both cases, the latter energies are significantly larger than the temperature of the system.

The above ρ dependence of the level density parameter a is not included in the present schematic model, in order to isolate the destabilizing surface effects from other shape-destabilizing effects. In a more complete model, where both shape and density dependencies of the level density parameter are considered, the loss of shape stability is expected to occur for even lower excitations than indicated in Fig. 1. This is so because a self-similar [7] radial expansion leads to both a reduction of the surface energy coefficient ϵ_S and an enhancement of the surface term in the expression [Eq. (3)] for the level density parameter.

To gain a better understanding of the discovered surface effect and its role in generating a shape instability of finite nuclei, thermodynamical surface tension and thermodynamical fissility are discussed below. The derivation of the respective equations is based on the observation that a thermodynamical driving force F_β for a coordinate β is generally given by the gradient of the total energy with respect to β, taken at fixed value of the entropy S. Accordingly, one writes for the thermodynamical surface tension Λ_{td}

$$\Lambda_{td} = \frac{\partial E^*}{\partial \sigma}\bigg|_{S=\text{const}}, \tag{6}$$

where σ is the surface area.

The conditional partial derivative on the right-hand side of Eq. (6) can be calculated by noting that the condition $S = \text{const}$ implies

$$\Delta S^2 = 4a^0 E^* - 4\left(a^0 + \frac{1}{4\pi r^2}\,\alpha_S \Delta\sigma\right) \times \left(E^* + \Delta E^* - \frac{1}{4\pi r^2}\,\epsilon_S \Delta\sigma\right) = 0, \tag{7}$$

where a^0 is the ground-state value of the level density parameter, ϵ_S and α_S are defined via Eqs. (2) and (3), respectively, and r is the radius parameter.

By taking the limit of $\Delta E^* -> 0$ and $\Delta\sigma -> 0$, while omitting the terms that are quadratic in these two small quantities, one obtains from Eq. (7)

$$\Lambda_{td} = \frac{1}{4\pi r^2}\left(\epsilon_S - \frac{\alpha_S}{a^0}\,E^*\right) = \frac{1}{4\pi r^2}\,(\epsilon_S - \alpha_S T^2), \tag{8}$$

where the Fermi-gas model relationship between temperature T and excitation energy E^*, $E^* = a^0 T^2$, is utilized.

As seen from Eq. (8), the thermodynamical surface tension Λ_{td} decreases monotonically with increasing excitation energy, from its liquid-drop ground-state value of $\Lambda_{\text{ld}} = \epsilon_S/4\pi r^2$ to zero at a certain transition temperature T_T:

$$T_T = \sqrt{\frac{\epsilon_S}{\alpha_S}}\,. \tag{9}$$

Note that Eq. (9) is analogous to the Fermi-gas model expression for the temperature $T = \sqrt{E^*/a^0}$. A numerical estimate, using $\epsilon_S = 18$ MeV and [11] $\alpha = 0.274\ \text{MeV}^{-1}$, yields for the transition temperature $T_T \approx 8.1$ MeV, i.e., a value that is significantly lower than (the 13–20 MeV) predicted by standard nuclear-matter calculations for semi-infinite matter [4].

The shape stability of finite nuclei is commonly described by the fissility parameter χ_{ld}, rather than by the surface tension. The fissility parameter accounts also for the disruptive action of Coulomb forces in addition to the cohesive action of the surface tension. For small ellipsoidal deformations characterized by a shape parameter α_2, the surface and Coulomb energies, E_S and E_C, are given by

$$E_S = E_S^0\left(1 + \frac{2}{5}\,\alpha_2\right), \qquad E_C = E_C^0\left(1 - \frac{1}{5}\,\alpha_2\right), \tag{10}$$

where E_S^0 and E_C^0 are the respective energies at a spherical shape. In these terms, the fissility parameter is given by

$$\chi_{\text{ld}} = -\frac{\partial E_C}{\partial \alpha_2} \Big/ \frac{\partial E_S}{\partial \alpha_2} = \frac{E_C^0}{2E_S^0}\,. \tag{11}$$

A thermodynamical generalization of Eq. (11) is obtained by replacing the surface energy $E_S^0 = 4\pi r^2 \Lambda_{\text{ld}} A^{2/3}$ by its thermodynamical counterpart $4\pi r^2 \Lambda_{td} A^{2/3}$:

$$\chi_{td} = \frac{E_C^0}{8\pi r^2 \Lambda_{td} A^{2/3}} = \chi_{\text{ld}}\left(1 - \frac{\alpha_S}{\epsilon_S}\,T^2\right)^{-1}. \tag{12}$$

Consequently, a spherical nucleus becomes unstable against ellipsoidal distortions when the thermodynamical fissility approaches $\chi_{td} = 1$, i.e., at a limiting temperature of

$$T_{\text{lim}} = T_T\sqrt{1 - \chi_{\text{ld}}}\,. \tag{13}$$

Here, the quantity T_T is the transition temperature introduced in Eq. (9).

For the present system of $A = 200$, $Z = 80$, Eq. (13) predicts $T_{\text{lim}} \approx 4.9$ MeV, when the liquid drop fissility parameter is approximated by $\chi_{\text{ld}} = Z^2/50A$. This

temperature is consistent with what is seen in the bottom panel in Fig. 1 as necessary or sufficient to cause a transition from the mononuclear to the dinuclear configuration.

In summary, a new surface effect is described that can lead to a loss of macroscopic stability of finite nuclei already at very moderate excitation energies and, hence, to fragmentation. In the constructed schematic microcanonical model of a two-phase system, one observes a second-order phase transition from a mononuclear to a dinuclear configuration. This transition occurs at a temperature that is by more than 1 order of magnitude lower than the change in potential energy associated with this transition. The large magnitude of the discovered effect calls for a further study of its possible implications. In particular, it appears desirable to include this effect in the practical models of nuclear multifragmentation proposed in the literature. It is worth emphasizing that this surface effect further decreases the stability of hot finite nuclei. It thus amplifies the known effects of a thermal expansion or a reduction of the surface energy coefficient at elevated temperatures.

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