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EXCITATION ENERGY DEPENDENCE OF SHELL EFFECTS ON NUCLEAR LEVEL DENSITIES AND FISSION FRAGMENT ANISOTROPIES

V. S. Ramamurthy, S. S. Kapoor, and S. K. Kataria
Bhabha Atomic Research Centre, Trombay, Bombay-85, India
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It is shown that the nuclear shell effects on the level density disappear even at medium excitation energies of the order of 40 MeV. Fission-fragment anisotropy data for actinide nuclei having double-humped fission barriers are shown to contain evidence for this effect. The implication of this effect on the production of superheavy nuclei is pointed out.

The inclusion of a shell correction to the liquid-drop model (LDM) deformation energy of a nucleus, obtained from the deviation of the distribution of single-particle states in the nucleus from a uniform distribution, has led to the prediction of a pronounced double-humped fission barrier[1] for nuclei in the actinide region. Experimental support[2] for these concepts has recently come from several features of low-energy fission, such as the discovery of a large number of spontaneously fissioning isomers, sub-barrier resonance groups in slow-neutron–induced fission, and systematics of near-threshold fragment anisotropies. However, an important question arises as to how the ground-state nuclear shell corrections influence the observables such as the fragment anisotropies and fission excitation functions in the case of a "hot" nucleus, having an excitation energy much above the fission threshold. It is known[3-5] that the fission probability and the fragment angular distributions depend on the properties of the transition-state nucleus, which, by definition, corresponds to the deformation where the nuclear entropy S is minimum. If one uses the Fermi gas expression $S=2(aE_x)^{1/2}$, the point of minimum entropy becomes identical with the point of minimum excitation energy E_x and the transition state corresponds to the nuclear shape at the top of the fission barrier as assumed in all previous work. This expression, however, which is valid only for a system having a uniform spacing of single-particle levels, should be suitably modified to include the nuclear shell effects, now known to be present even at large deformation, and the transition state should be redetermined from considerations of nuclear entropy. In other words, the interpretation of fission data in the statistical region is closely related to the question as to how the shell effects on nuclear entropy (or level density) vary with excitation energy. It is shown in this note that the shell effects on nuclear entropy disappear even at medium excitation energies of the order of 40 MeV, and as an evidence for this the fragment anisotropy data in medium-energy fission are shown to be consistent with the above conclusion.

A brief description of the present calculations of nuclear entropy S is given below. For a system of noninteracting Fermions, containing N particles with total energy E, the following relations hold:

$$N=\sum n_k \tag{1}$$

$$E=\sum n_k \epsilon_k \tag{2}$$

where ϵ_k is the energy of the kth single-particle state. The Fermi-Dirac distribution function n_k and the entropy S are given by

$$n_k=\frac{1}{1+\exp[(\epsilon_k-\mu)/T)]} \tag{3}$$

and

$$S=-\sum[n_k \ln n_k+(1-n_k)\ln(1-n_k)], \tag{4}$$

μ and T being the chemical potential and the temperature of the system, respectively. The entropy and the total energy of a nucleus can be obtained by adding the individual entropies and total energies of the protons and neutrons. The corresponding excitation energy E_x can be determined by subtracting from the total energy E the ground-state energy E_g, obtained by summing the energies of the lowest N and Z single-particle states of neutrons and protons. In the present calculations the spectrum of single-particle states was first obtained for a modified spherical harmonic-oscillator potential with the potential parameters of Seeger.[6] Numerical calculations of S vs E_x were then carried out with Eqs. (1)-(4) for several nuclear systems. Here we present the results of these calculations for two typical cases: (i) for the doubly magic nucleus, Pb^{208}, where the shell correction to the LDM mass is negative, and (ii) for the spherical shape of Pu^{242} where the shell correction is positive.

In order to bring out clearly the deviation of the present results from the predictions of the Fermi gas model we have shown in Fig. 1 a plot of S^2 vs E_x for these two cases. It is seen that the curve deviates considerably from a straight line of the form $S^2=4aE_x$, expected from the Fermi gas model. However at excitation energies $E_x > 30$-40 MeV an asymptotic behavior of the form

$$S^2=4a(E_x \pm \Delta E_x) \tag{5}$$

is apparent where ΔE_x represents the magnitudes of the intercepts on the energy axis of the asymptotic straight lines, and ± signs refer to the two cases of positive and negative shell corrections, respectively. In order to invesitgate any possible relation between the asymptotic values of a and ΔE_x and the ground-state shell correction Δ_{shell} to the LDM mass, the magnitude of the ground-state shell corrections was calculated by Strutinsky's procedure[1] by generating a uniform level sequence corresponding to the single-particle level sequence ϵ_k, and were found to be −9.2

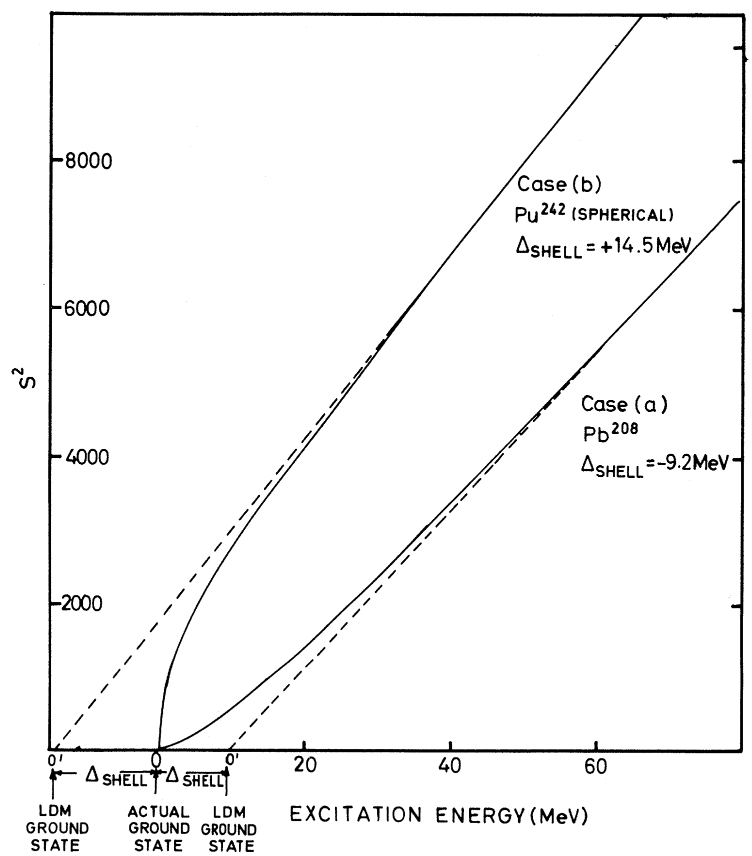

FIG. 1. Plot of S^2 vs E_x, for the cases of the doubly magic nucleus Pb^{208} and the nucleus Pu^{242} (spherical shape). The dashed curves in each case represent the asymptotic behavior at high excitation energies.

MeV for Pb^{208} and +14.5 MeV for the spherical shape of Pu^{242}. It is seen that these values of the shell corrections are numerically equal to the energy intercept OO' ($=\Delta E_x$) of the asymptotic straight lines in both the cases within 0.2 MeV, implying that the energy point O' corresponds to the LDM ground state. Calculations of the entropy versus the excitation energy were also carried out for the uniform level scheme using equations analogous to Eqs. (1)-(4) with the summations replaced by integrations. It was found that in this case plots of S^2 vs E_x exactly coincide with the asymptotic dotted lines shown in Fig. 1, remembering that the zero excitation energy in this case corresponds to the LDM ground state O'. The slope of the asymptotic straight line corresponded to a value of $a=A/8$ in both cases.

A general relationship for the entropy S can, therefore, be written as

$$S^2=4[a\pm\Delta_a(E_x)][E_x\pm\Delta_E(E_x)], \qquad (6)$$

where Δ_a and Δ_E are energy dependent, with the + and − sign referring to systems with positive and negative ground-state shell corrections, respectively. Further, the energy dependence of Δ_a and Δ_E are such that as the excitation energy increases, Δ_a decreases approaching an asymptotic value of zero, while Δ_E increases approaching an asymptotic value equal to Δ_{shell}, and it can be seen that these asymptotic values are reached at $E_x \geqslant 30$ to 40 MeV. The important conclusion emerging from the above analysis is, therefore, that a relation of the form $S=2(a'E_x')^{1/2}$ can be used for the calculation of S with the following qualifications: (i) The effective excitation energy E_x' is to be measured from a reference energy surface which coincides with the actual ground state at low E_x values and with the LDM ground state at $E_x > 30$-40 MeV; (ii) for low E_x values, a' is to be taken as an energy-dependent parameter which asymptotically becomes equal to the energy-independent Fermi gas parameter a, as $E_x > 30$-40 MeV. It can therefore be concluded that at excitation energies exceeding 30-40 MeV the shell effects are not manifesting themselves on the nuclear entropy (or level density).

The conclusion drawn above regarding the excitation energy dependence of nuclear entropy for nuclei having either a negative or a positive shell correction can be regarded as general and applicable to systems having any other single-particle level scheme. Applying these ideas to a nucleus having a double-humped fission barrier, it follows that although for low values of E_x, points of minimum entropy would correspond to the shapes on the top of the barriers I and II, at medium excitation energies ($E_x > 30$-40 MeV) the point of minimum entropy would correspond to the LDM barrier shapes. It has been shown[2] that in near-threshold fission, the fragment angular distributions are in fact characterized by the distribution of the K values of the open channels on the top of the barrier II only. On the basis of our analysis it can, therefore, be predicted that for nuclei having a double-humped fission barrier the shape of the transition-state nucleus should change from that of barrier II to that of the LDM barrier with increasing excitation energy in the region of 0 to 30 MeV.

In what follows it can be seen that in the fragment anisotropy data, evidence for this new effect does exist. Figure 2 shows the values of the effective moment of inertia J_{eff} of the transition state nucleus, derived from the experimental data[7] on the fragment anisotropies for 42.8-MeV helium-ion–induced fission in various target nuclei as a function of the value of Z^2/A of the fissioning nuclei on the basis of Halpern-Strutinsky statistical theory[5] and a recent expression given by Huizenga, Behkami, and Moretto.[8] We have also shown in the figure the expected variation of J_0/J_{eff} for shapes corresponding to barriers I and II, and for the LDM barrier shapes corrected[9] for the curvature correction to the surface tension. The nuclear deformations corresponding to barriers I and II for various nuclei were taken from the recent calculations of Nilsson et al.[10] Referring only to the actinide region for which a pronounced double-humped fission barrier is predicted, it can be seen from Fig. 2 that the experimental values of J_0/J_{eff} are not characteristic of the shapes at the barrier II but are closer to those expected for the LDM barrier shapes. Rather unreasonable assumptions about the level density parameter a will be needed if the values of J_0/J_{eff} derived from the anisotropy data are forced to coincide with the theoretical curves for barrier II or for barrier I. Noting that the experimental values of J_0/J_{eff} for actinide fissioning nuclei refer to those determined from anisotropy data at $E_x \approx 30$ MeV, one can conclude that at these excitation energies the fragment anisotropies are indeed characteristic of the LDM barrier alone. For nuclei lighter than thorium, the excitation energy of the fissioning nucleus is only about 15 MeV for the data of Fig. 1 and therefore one can expect shell effects

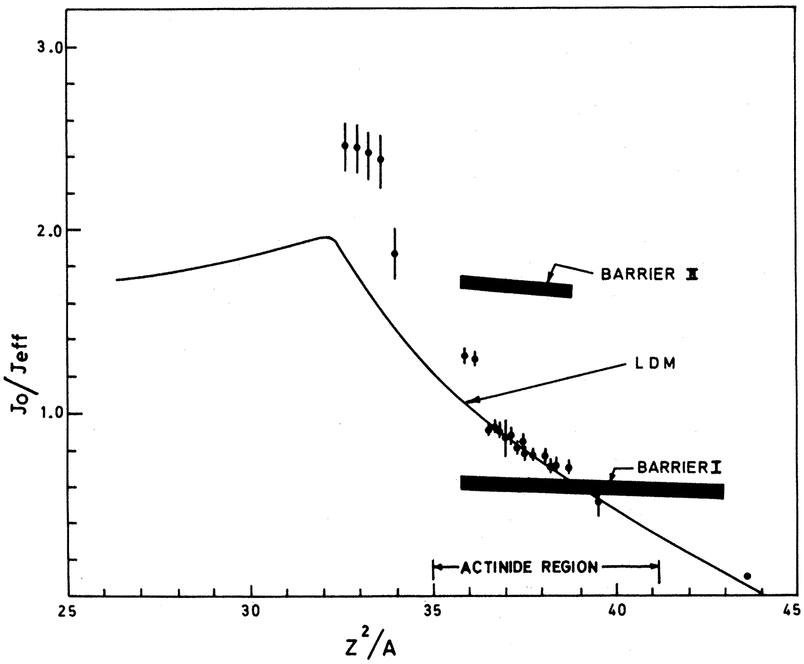

FIG. 2. Variation of J_0/J_{eff} with Z^2/A. The experimental points are derived (Ref. 8) from the fragment anisotropy data (Ref. 7) in helium-ion–induced fission of various target nuclei. For the calculation of the spherical moment of inertia J_0 a value of $r_0 = 1.21$ fm was used. The continuous curve gives the calculated (Ref. 9) variation for LDM barrier shapes and the patches for the shapes calculated by Nilsson et al. (Ref. 10) corresponding to barriers I and II.

on the barrier shapes and a deviation of J_{eff} from the rigid-body value, giving rise to the observed deviation of the experimental J_0/J_{eff} from the LDM prediction. In order to bring out the excitation energy dependence of the shape of the transition state nucleus, we have shown in Fig. 3 the experimental values[11] of K_0^2 versus the excitation energy for a typical case of alpha-induced fission of U^{238}, together with the theoretical curves calculated for the shapes corresponding to barrier I, barrier II, and the LDM barrier. In these theoretical curves, the shell and pairing effects on J_{eff} for specified shapes have already been incorporated on the basis of the results of Damgaard et al.[12] It is evident from Fig. 3 that the different slopes at low and medium excitation energies of the experimental curve of K_0^2 vs E_x arise from a change of the shape of the transition-state nucleus from that of barrier II to that of the LDM barrier in the energy range of about 4 to 30 MeV. It may be pointed out that these different slopes were earlier attributed[13] to the presence of a large pairing-energy gap at the barrier, but the recent analysis[14] of fission data does not support this explanation.

Further confirmation of the validity of our calculations of nuclear entropy has come from the good fits to the experimental values[15] of Γ_f/Γ_n for Tl^{201} which have been obtained with the present results without the use of any free parameters in the level-density expression. These

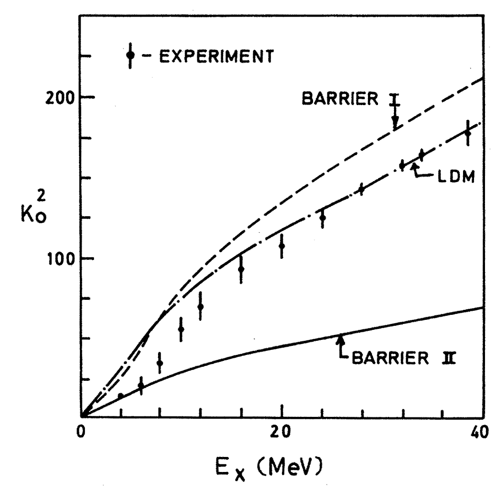

FIG. 3. Variation of K_0^2 with the excitation energy of the transition-state nucleus Pu^{242}. The experimental points are taken from Ref. 11. The calculated variations for the critical shapes corresponding to the LDM barrier, barrier I, and barrier II are shown by the different curves. The deviation of each of the calculated curves at low excitation energies from a parabola arises due to the inclusion (Ref. 12) of shell and pairing effects on the effective moment of inertia for specified shapes of the nucleus.

calculations on Γ_f/Γ_n are now being extended to other nuclei and these results will be published elsewhere.

Another interesting consequence of the present results is as follows: In the attempts currently being made to produce superheavy nuclei around the doubly magic nucleus $_{114}X^{296}$ by heavy-ion bombardment, the compound nucleus is always formed with an excitation energy exceeding a few tens of MeV. The calculations of nuclear entropy for nuclei in this region showed that the conclusions regarding the rapid washing out of shell effects on nuclear entropy with excitation energy are also valid for these nuclei. Therefore even at medium excitation energies of the order of 30-40 MeV, the nuclear entropy of these nuclei at any deformation would be as if the nucleus encounters the LDM potential energy surface and not the actual shell-dependent potential energy curve. Consequently, if the fissioning nucleus is "hot" ($E_x \geq 30$-40 MeV), the existence of a shell fission barrier in the ground state of these nuclei will not decrease the otherwise very large relative fission probability Γ_f/Γ_n, expected for the case of zero LDM fission barrier. This would then make the fraction of the compound nuclei surviving fission and reaching the ground state after a cascade of neutron emission vanishingly small. It is therefore imperative that the compound nucleus should be formed at a much lower excitation energy in order to produce these superheavy nuclei and the reactions involving heavy-ion bombardments may not be suitable for producing these nuclei. Further quantitative calculations concerning the production probability of these superheavy nuclei are in progress and will be reported elsewhere.

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