Fission of Metallic Clusters within the Shell Correction Method

Armando Vieira and Carlos Fiolhais

Departamento de Física, Universidade de Coimbra, P-3000 Coimbra, Portugal

Abstract. We used the Shell Correction Method to evaluate systematically fission barriers for charged metallic clusters of sodium and aluminum. Shell effects are responsible for the deviation of the fission barrier height from the value predicted by the Liquid Drop Model. Fragment deformations are essential degrees of freedom. The sizes for which fission competes with evaporation are correctly predicted for doubly charged clusters. We also present calculations of fission barrier heights for triply charged potassium clusters.

INTRODUCTION

The Liquid Drop Model (LDM) is useful to understand the main trends of metal cluster fission. In this simple model, fission is controlled by the fissibility x, which is half the ratio between the Coulomb and the surface energy of the spherical cluster [1]. However, quantal corrections to the LDM energy are essential for obtaining realistic heats of reaction and good barrier heights [2]. The Shell Correction Method (SCM) is adequate to study complex quantal systems where more rigorous methods are difficult to apply, such as large or deformed nuclei and clusters, and has been systematically applied to nuclear fission with great success. In the SCM, shell corrections are evaluated replacing the effective potential felt by the valence electrons by a simpler, non-self-consistent potential [3,4]. The total energy is obtained by adding these shell fluctuations to the smooth LDM energy.

One advantage of the SCM is that it may be used, with little increase of computational cost, to systems with arbitrary size and charge. In this work we study fission barriers of doubly charged clusters of sodium and aluminum containing up to 30 atoms (see also [5]). The barriers are calculated for all possible decay channels including independent ellipsoidal deformations in both fragments. We analyze the role of mass asymmetry and fragment deformations in the fission barriers. Finally we analyze triply charged clusters of potassium.

The LDM energy, E_{LDM}, is the sum of surface, curvature (evaluated in the

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Stabilized Jellium Model [6]) and Coulomb contributions (considering that the charge is distributed on the surface) [1]. The shell correction $\Delta E = \sum \epsilon_i - \tilde{E}$, where $\tilde{E}$ is a conveniently defined average energy, was calculated using the single-particle energies levels ϵ_i of the Asymmetric Deformed Two-Center Shell Model [7]. The potential energy is the sum of the LDM term and the shell correction: $E_{SCM} = E_{LDM} + \Delta E$.

RESULTS

In Fig. 1 we present the barriers heights for the fission of Na_N^{+}, with $6 \leq N \leq 30$, for the most favourable channel, as a function of the number of atoms, considering spherical (SCM-sph) and deformed fragments (SCM-def). In the second case (SCM-def) we obtained the fission path fixing the distance between the fragments and the asymmetry parameter, and then selecting the deformation of each fragment that minimizes the total energy.

For $N \leq 10$, the LDM barrier vanishes and the total barrier is due exclusively to electronic shell effects. The most favourable channel has, in general, a magic, or near-magic piece ($p = 3$, 9 or 21), in agreement with the heat of reaction analysis [8]. However, the channel with $p = 3$ is the most favoured only for a small number of systems. The very asymmetric channel $p = 2$, not predicted on the basis of the heats of reaction, is the most favoured for small clusters in the SCM-sph method.

In both spherical and deformed versions of SCM, initial clusters with a magic number of electrons, like $N = 10$ and 22, are particularly stable against fission. SCM-def gives rise, in general, to higher fission barriers than SCM-sph, especially for $N \leq 16$ and $20 \leq N \leq 23$. This is due to the deformation of non-magic clusters, whose ground-state energies may be much lower than those of magical ones. For $N = 28$, 29 and 30 the barrier is only slightly higher than the evaporation energy for spherical fragments. This agrees with experimental data from Bréchignac *et al.* [9], who detected a strong competition between fission and evaporation in that size range. The so-called appearance size for these clusters is $N_c = 27$ (SCM-def) and 28 (SCM-sph), in excellent agreement with the experimental value $N_c = 27 \pm 1$ [10].

Fig. 2 shows the barrier heights for doubly charged aluminum clusters. Systems with 7, 13 and 18 atoms are particularly stable, while clusters with 6, 9, 14, 15 and 16 atoms have a small barrier. In contrast to sodium clusters, and with the exceptions of $N = 7$, 17 and 18, the SCM-def and SCM-sph barriers are very similar. Shell effects are more pronounced for aluminum than for sodium clusters. These strong effects are responsible for the following features: i), the SCM critical number is much lower than the LDM one; ii) the favourable decay channels predicted in the heat of reaction analysis [11] are not those with the lowest barrier, excepting $N = 5$, 7, 8, 10 and 11; iii) small clusters ($N < 14$) have significative barriers, between 1 and 2 eV, while

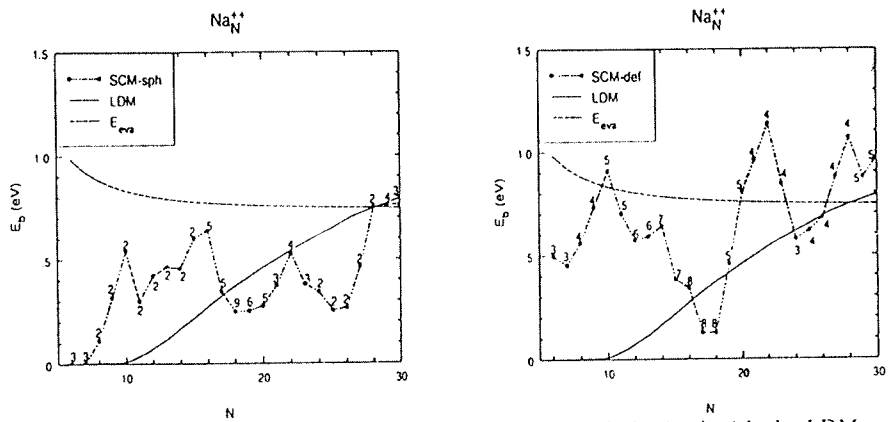

FIGURE 1. Barrier heights for the most favourable channel obtained with the LDM and the SCM with spherical (SCM-sph) (left) and deformed fragments (SCM-def) (right). The LDM curve corresponds to the $p = 3$ channel. The numbers indicate the respective channel.

for $N < 5$ the LDM shows no barrier. The cluster Al_2^{++} breaks spontaneously in two magic fragments. The appearance size is $N_c = 21$ and 20, for SCM-sph and SCM-def, respectively, in reasonable agreement with the experimental value of $N_c = 17 \pm 1$.

Let us now consider the decay of triply charged potassium clusters: $K_N^{3+} \rightarrow K_{N-p}^{++} + K_p^+$. Fig. 3 shows the fission barrier height within K_N^{3+}, with $40 \leq N \leq 80$. For $40 < N < 55$ and $70 < N < 76$ we find that the most favourable channel is around $p = 6$, while for $55 < N < 70$, we get $p = 2$ and 3. These findings agree partially with recent experimental results obtained by Bréchignac *et al.* [13]. These authors found that the observed decay channel increases from about 7, for $N \approx 50$, to 20, for $N \approx 100$.

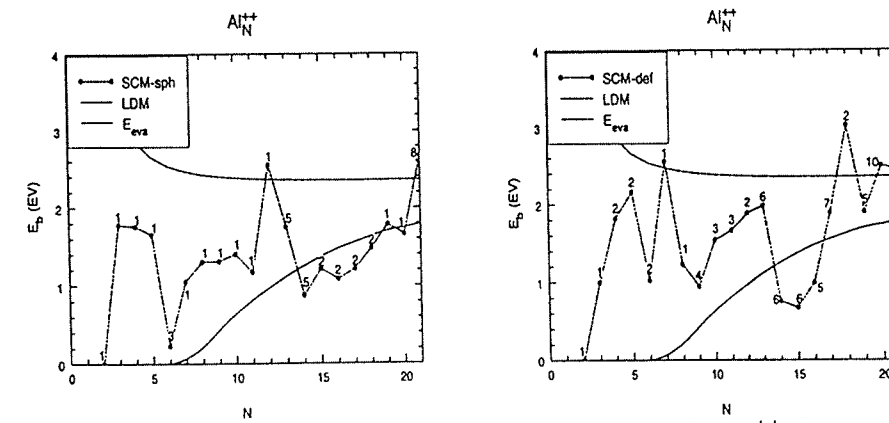

FIGURE 2. Barrier heights for the most favourable decay channel of Al_N^{++} clusters. See caption of Fig. 1.

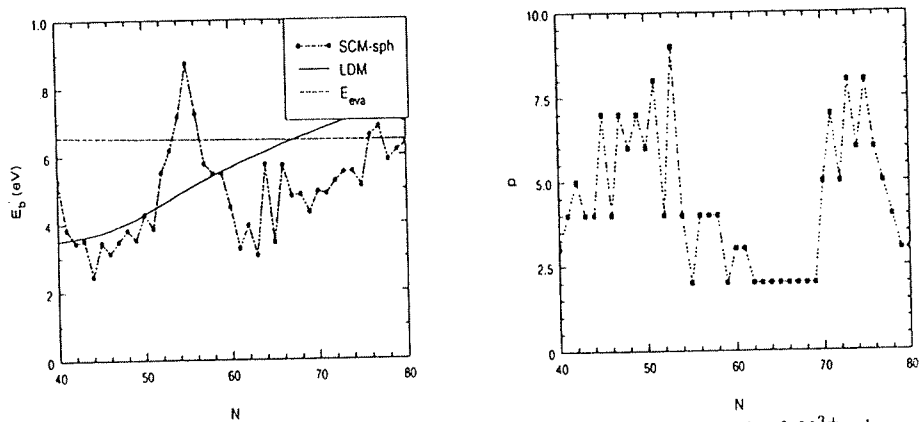

FIGURE 3. Left: Barriers heights for the most favourable channel of K_N^{3+} clusters. Right: Most favoured channel as a function of N.

In summary, we reproduced well the appearance sizes N_c for doubly charged sodium and aluminum clusters. We have also presented first results for triply charged potassium clusters.

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