International Journal of Modern Physics E

Further microscopic studies of the fission barriers of heavy nuclei

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Received (received date)
Revised (revised date)

Two systematic sources of error in most current microscopic evaluations of fission-barrier heights are studied. They are concerned with an approximate treatment of the Coulomb exchange terms (known as the Slater approximation) in the self-consistent mean fields and the projection on good parity states (e.g., of positive parity for the spontaneous fission of an even-even nucleus) of left-right reflection asymmetric intrinsic solutions (e.g., around the second barrier). Approximate or unprojected solutions are shown to lead each to an underestimation of the barrier heights by a few hundred keV.

1. Introduction

In most microscopic calculations of nuclear binding energies using effective nucleon-nucleon interaction or their Energy Density Functional (EDF) avatars, data are reproduced in the best cases with an accuracy which hardly goes below a couple of MeV. When evaluating relative energies, one may hope for a partial cancellation of such errors but this is by far not granted in general. A particular case of choice of such relative energies is constituted by the so-called fission barrier heights.

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Being a rough one-dimensional description of the very complex many-body fission process, their "experimental" values result in most cases from a model dependent -or partial- analysis of some data. Nevertheless, they provide undoubtedly a much wanted energy scale on which microscopic calculations adjust their phenomenological parameters (e.g. this has been in particular the case for the fit of the SkM* Skyrme interaction [1]). As an example of the accuracy of these barrier calculations we may quote the results of Ref. [2] where an average accuracy of about 1 MeV for the first and second barriers of actinide nuclei (as well as fission isomer excitation energies) is found upon making a very rough estimate of the rotational energy correction.

Such a range of error is very significant in physical terms. It is generally estimated that a variation of 1 MeV roughly corresponds to a change of spontaneous fission half lives by 4 orders of magnitude. It is therefore of paramount importance to pinpoint systematical errors inherent to a given theoretical (phenomenological) approach which could lead to wrong estimates of fission properties or conversely, when making a fit of the interaction taking care of fission barriers, which could have adverse effects on theoretical estimates of other quantities.

Without aiming at any exhaustibility in chasing such errors, two examples will be studied here: i) the approximate treatment of the Coulomb exchange terms known as the Slater approximation in the self-consistent mean fields, ii) the projection on good parity states (e.g. of positive parity for the spontaneous fission of an even-even nucleus) of left-right reflection asymmetric intrinsic solutions (e.g. around the second barrier).

2. On the effect of the Slater approximation of Coulomb exchange terms

Most microscopic calculations use the infinite matter Pauli correlation function (within a local density approximation) to compute the Coulomb exchange expectation value in a Slater determinant producing an EDF piece involving only the local proton density

$$E^{\rm Slater}_{\rm Coul.exch.} = -\frac{3}{4}e^2\left(\frac{3}{\pi}\right)^{\frac{1}{3}}\int d^3r\,\rho_p^{4/3}(\vec{r})\,. \tag{1}$$

The above yields, within a variational approach, the following mean field piece

$$V^{\rm Slater}_{\rm Coul.exch.}(\vec{r}) = -\left(\frac{3}{\pi}\right)^{\frac{1}{3}}e^2\rho_p^{1/3}(\vec{r})\,. \tag{2}$$

This approach is usually refered to as the Slater approximation [3]. It has been tested many years ago [4] by incorporating an exact Coulomb exchange treatment in Hartree-Fock calculations of the ground state (spherical or deformed) solutions of 8 light nuclei (from ^{16}O to ^{56}Ni). In this early work, the Skyrme SIII interaction [5] had been used.

Such a study has been recently revisited by J. Skalski [11] for the ground-state Hartree-Fock solutions of 9 spherical subshell closed nuclei, from nuclei as light as ^{16}O to superheavies such as ^{310}126. The SkP [6] and SkM* Skyrme interactions have been used.

From these two studies, one has concluded that the errors found (namely relative errors, consistently in all this Section) for the Coulomb exchange energies, are relatively small, amazingly interaction-independent (at least for the three interactions under consideration, namely SIII, SkM* and SkP) and decreasing with the total particle number A (as expected in view of the infinite matter origin of the Slater approximation).

Even though no quantitative account nor general argument, has been given there, as far as fission barrier heights are concerned, one should mention the calculations of Ref. [7]. The authors have included in a HFB framework full Coulomb exchange terms in various nuclear systems , including the fission barriers of ^{254}No.

In the following we have performed self-consistent (Hartree-Fock or Hartree-Fock-plus-BCS calculations) using only the SIII Skyrme effective interaction (with a seniority interaction for the BCS part). The Coulomb energy and mean-field contributions have been calculated as proposed in Ref. [8] by evaluating the Coulomb matrix elements upon using a gaussian integral representation of a Yukawa-type two-body interaction as

$$\frac{e^{-\mu|\vec{r}_1-\vec{r}_2|}}{|\vec{r}_1-\vec{r}_2|} = \sqrt{\frac{2}{\pi}}\int_0^\infty e^{-\mu^2\sigma^2/2} e^{-\mu|\vec{r}_1-\vec{r}_2|^2/2\sigma^2}\frac{d\sigma}{\sigma^2}\,. \tag{3}$$

First, we have extended our study of the above spherical subshell closed nuclei beyond the Hartree-Fock approximation in use and allowed for pairing correlations à la BCS with a seniority force. For two nuclei (^{90}Zr and ^{298}114) where pair correlated solutions have been found to occur, the error has been dramatically reduced (by much more than half as seen in Fig. 1). This appears thus as a hint that the error found so far is due to the particular filling of those nuclei which had been considered.

We have then calculated the error for six isotonic series ($N = 28, 64, 78, 106, 126, 146$) of nuclei constrained to be spherically symmetric (allowing pairing correlations to be included). As seen in Fig. 1 the trend of the error is totally unambiguous. Away from proton magic numbers ($Z = 20$, 28, 50, 82) the quality of the Slater approximation improves very rapidly. To confirm the role of the proton single-particle level scheme, we have performed similar calculations for three short isotopic series ($Z = 72, 82, 92$). The error is found, see Fig. 1, to be remarkably stable within a given isotopic series.

Then, we have explored changes in the proton single-particle level density $\overline{\rho_p}$ at the Fermi surface, not upon changing the proton number but instead the axial quadrupole deformation for the same nucleus. As examplified in Fig. 2 reporting the results of calculations for 5 nuclei (^{24}Mg, ^{48}Cr, ^{106}Mo, ^{178}Hf, ^{238}U) the error is always–and often very significantly–larger for their deformed ground-state solutions

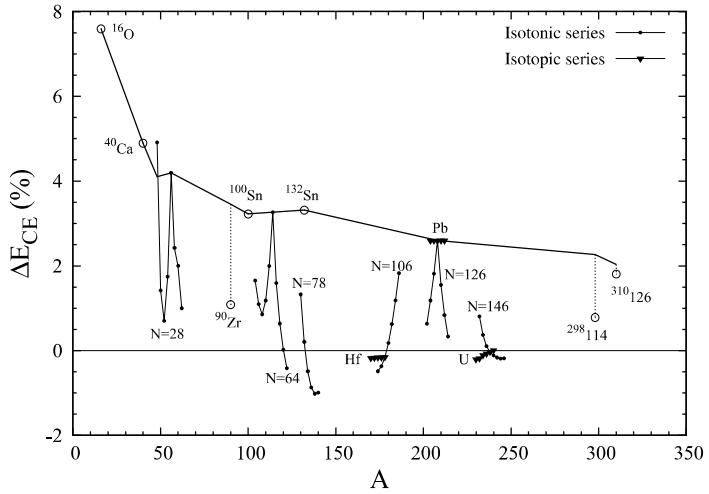

Fig. 1. Relative error $\Delta E_{\rm CE}$ associated with open proton shell nuclei, where $E_{\rm CE}$ is the expectation value of the exchange part of the Coulomb interaction. The closed proton shell nuclei are reported as open circles linked by the solid line. Some isotonic series are represented in dash lines, whereas three isotopic series corresponding to Hf, Pb, and U isotopes are plotted with black triangles. For the ^{90}Zr and ^{298}114 nuclei, the plotted solutions correspond to BCS solutions.

than for the solutions constrained to have a vanishing quadrupole moment. In the

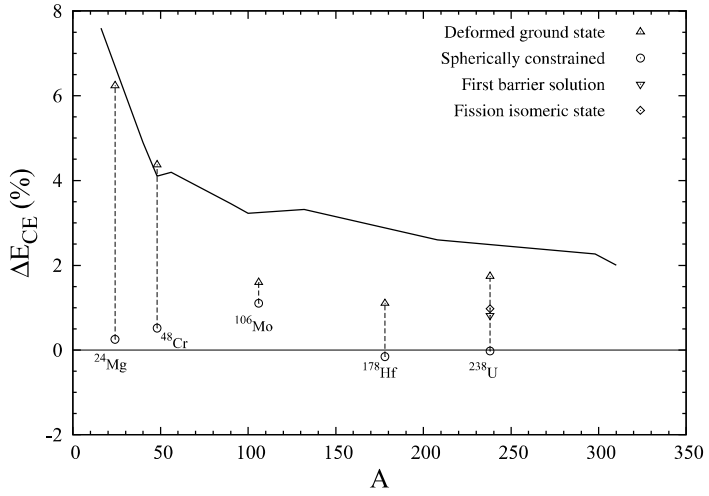

Fig. 2. Relative errors $\Delta E_{\rm CE}$ in the ground-state solution of deformed nuclei and in their spherically constrained solution. The solid line, shown for comparison, corresponds to the closed proton shell nuclei appearing in Fig. 1.

case of ^{238}U, we have also reported the errors corresponding to the top of the first barrier and the fission isomeric state. These errors lie between those obtained for the spherical and deformed solutions. To confirm these findings we have compared the error made on the Coulomb exchange energies (middle panel of Fig. 3) with the proton BCS pairing gap, taken as an index of the proton single-particle level density at the Fermi surface (lower panel of Fig. 3) along the beginning of the calculated fission barrier of the ^{70}Se nucleus imposing a vanishing axial octupole moment (upper panel of Fig. 3). As seen from Fig. 3 the error and the gap are very

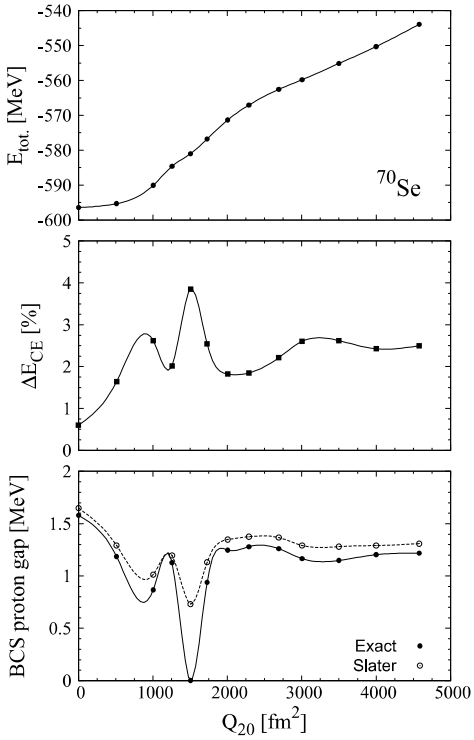

Fig. 3. (Top panel) Deformation-energy curve of the ^{70}Se as a function of the mass quadrupole moment Q_{20} in fm^{2}2. (Middle panel) Relative error $\Delta E_{\rm CE}$ as a function of Q_{20}. (Bottom panel) Variation with Q_{20} of proton pairing gaps calculated with the exact treatment of Coulomb exchange terms in solid line (with full circles) and using the Slater approximation in dashed line (with open circles).

well anti-correlated functions of the deformation.

We can therefore conclude that the quality of the Slater approximation will be less good for low values of $\overline{\rho_p}$ than for densities corresponding to mid-shell situations.

Since it is well known that systematically $\bar{\rho}$ will be higher at the top of the fission barrier than at the ground state deformation (for a direct evidence of that

fact in the context of fission-barrier calculations within the Hartree-Fock-plus-BCS framework, see Ref. [9]), we can predict that the correction to be made to account for the error brought in by the Slater approximation will be lower in the former case than in the latter. As a result, using the Slater approximation will systematically lead to an underestimation of the fission-barrier heights. In the calculations already discussed (see Fig. 2) of the ^{238}U fission barrier [10], one should add beyond the Slater approximation values about 310 keV to the first fission barrier height and about 280 keV to the fission isomeric energy, quantities which are not without consequences on spontaneous fission halflives as already mentioned. Even though it is hard to get more than a tendency on the l.h.s part of the Figure 12 of Ref. [7] their results (for ^{254}No) are consistent with our above conclusions.

Before closing this Section, two facts deserve a short notice.

In the above discussion of the impact of $\overline{\rho_p}$ on the quality of the Slater approximation, we have only considered the errors on the Coulomb exchange energies. Of course, the quantities whose errors should have been compared (and actually which have been discussed just above in the ^{238}U fission study) are total energies (the sum of kinetic, nuclear, direct and exchange Coulomb energies). However, it has been demonstrated [10] that, in most cases, the errors on such total energies and Coulomb exchange energies are equal up to some tens of keV.

As found out already in the results of Ref. [4] and emphasized in Ref. [11], it has been systematically found that all hole states, in Hartree-Fock parlance, are pushed up by the Slater approximation from their location in exact calculations, while conversely, all particle states are pushed down. As a result the proton single particle level density at the Fermi surface is unduly enhanced by the Slater approximation. The effect of such an error on proton particle-hole energies as far as correlation properties are concerned, remain to be specifically studied. Indeed, if the residual interaction is to be considered as a perturbation, it is easy to see that such a trend could affect significantly the configuration mixing.

3. On the effect of a projection on good parity states

For a very long time, it is known [12] that the second fission barriers of actinides are unstable with respect to the left-right asymmetry. It has been then recognized that this phenomenon is driven by the heavy fragment shell effects explaining thus already at this early stage of the fission process, the asymmetric pattern of the fragment mass yields (at low compound-nucleus energy) which had been observed in this region long before.

In most microscopic or macro-microscopic calculations in the U, Pu region, upon increasing the elongation after the fission isomeric state, the intrinsic equilibrium solution becomes unstable with respect to the left-right reflection symmetry and acquires rapidly a larger and larger octupole deformation which stabilizes at a value corresponding, as we just have noted, to the most probable fragmentation. This fact, as we will see, plays an important role for our purpose here.

Yet, even though the intrinsic parity may be broken for some microscopic solutions under consideration, the parity of the physical solution must be conserved during the fission process. For instance, if one describes the spontaneous fission of an even-even nucleus as ^{240}Pu, one should evaluate the fission barrier obtained upon imposing the positive parity to the solutions. This may be obtained by projecting intrinsic solutions on the desired parity. This corresponds thus to a projection after variation, amounting to a mixing of two configurations $|\Psi\rangle$ and $|\widetilde{\Psi}\rangle = \hat{\Pi}|\Psi\rangle$, where $\hat{\Pi}$ is the parity operator.

Recently such a projection of correlated microscopic solutions à la HTDA (Highly Truncated Diagonalization Approach) [13] has been extensively studied as the PhD thesis work of one of the authors [14]. We will thus simply outline here, the major points of this approach and discuss the results which have been obtained when applying it to the second fission positive-parity barrier of ^{240}Pu.

The HTDA calculations are designed to produce realistic correlated wave functions in an approach which conserve explicitly the particle number and do not violate the Pauli principle. They consist in performing intrinsic shell-model like calculations using single-particle states deduced from a mean-field potential $\hat{U}$. This field is a priori arbitrary. Yet, its realistic character, given a state-of-the-art microscopic effective Hamiltonian $\hat{H}$, is essential to limit the size of our n-particle n-hole many-body basis. The mean field $\hat{U}$ will be taken here as the one-body reduction of the nucleon-nucleon effective interaction $\hat{V}$ included in $\hat{H}$ for a corresponding approximate (e.g., à la BCS) microscopic correlated wave function.

The HTDA Hamiltonian is written as

$$\hat{H} = \hat{K} + \hat{V} = \hat{H}_{\rm MF} + \hat{V}_{\rm res} \tag{4}$$

where $\hat{K}$ is the kinetic energy while the mean field (one body) Hamiltonian $\hat{H}_{\rm MF}$ is defined by

$$\hat{H}_{\rm MF} = \hat{K} + \hat{U} - C \tag{5}$$

and the residual interaction $\hat{V}_{\rm res}$ by

$$\hat{V}_{\rm res} = \hat{V} - \hat{U} + C \tag{6}$$

The particle-hole vacuum $|\Phi_0\rangle$ is defined as an eigensolution of $\hat{H}_{\rm MF}$. Here we have chosen this vacuum as the ground state of $\hat{H}_{\rm MF}$. The constant C appearing in the above definitions is chosen in such a way that the expectation value of the residual interaction is vanishing for $|\Phi_0\rangle$.

In the present calculations, we have restricted the particle and hole single-particle states to lie within 6 Mev below and above the Fermi energy (defined as half the sum of the energies of the last occupied and first unoccupied state in $|\Phi_0\rangle$). The many body basis is comprised, beyond the vacuum, of only one-pair-transfer states (pairs meaning here Cooper pairs of time-reversal conjugate single-particle states). We have chosen the Skyrme SkM* for $\hat{V}$. As usually done in practical HTDA calculations, we have replaced $\hat{V}$ by a density-independent zero-range interaction to

define the residual interaction. The correlated wave function $|\Psi\rangle$ is obtained by a diagonalization of the Hamiltonian $\hat{H}$ through a Lanczös algorithm process.

Through a judicious choice of the mean field $\hat{U}$ (see Ref. [14]) the expression for the energy of the projection of the state $|\Psi\rangle$ on a state of good parity $p = \pm 1$ may be simplified as

$$E_p = \frac{\langle\Psi|\hat{H}|\Psi\rangle + p\langle\Psi|\hat{H}|\widetilde{\Psi}\rangle}{1 + p\langle\Psi|\widetilde{\Psi}\rangle} \,. \tag{7}$$

The non-diagonal overlaps are calculated using methods due to Löwdin [15] (The use of the heavy Balian-Brézin [16] generalized Wick theorem is not necessary in our case since we deal only with Slater determinants).

The results of our projection (after variation) calculations for the fission barrier of ^{240}Pu for a positive parity, are summarized in Figure 4. In the lower panel, it appears that the projection has the following effects: i) it yields a small stable octupole deformation around the fission isomeric state, defined by an elongation $Q_{20}^{(0)}$, for an elongation much smaller than the one ($Q_{20}^{(1)}$) where an instability of the HTDA unprojected solution is observed, ii) before reaching the second (asymmetric) fission barrier at an elongation $Q_{20}^{(2)}$, the projection does not yield any significant effect on the energies.

This can be explained in the following way which is illustrated in the upper panel of Fig. 4. Much before $Q_{20}^{(0)}$, at $Q_{20} = 64$ b for example, the unprojected solution is stable with respect to the axial Q_{30} mode with a stiffness large enough to only allow for the creation, by the projection on positive parity, of a shoulder on the $E(Q_{30})$ curve at a fixed elongation. Around the superdeformed solution $Q_{20}^{(0)} \sim 85$ b the stiffness parameter is no longer sufficiently large to prevent the projection from creating a stable equilibrium deformation (such a weak stiffness had already been found in Hartree-Fock-Bogoliubov calculations in the super-deformation region of Hg and Pb isotopes [17]).

At a quadrupole deformation around $Q_{20} \sim 110$ b, the non-diagonal overlaps (for the identity and Hamiltonian operators) become small enough so that the projected energies are not significantly different from the unprojected ones, a property which will be, of course, all the more verified than the octupole deformation will increase. For even larger elongations, e.g., $Q_{20} \sim 130$ b, the equilibrium octupole deformation parameter will get larger to stabilize, as we have recalled, at a value typical of the most probable asymmetric fission fragmentation and thus yielding a minimum in the $E(Q_{30})$ curve exactly at the same place than in the unprojected case.

As a result the height of the second fission barrier when projected on the positive parity will be enhanced from its unprojected value. This effect is only due to the relatively soft character of the octupole deformation energy curve near the fission isomeric state and not to the projection at the top of the second barrier where the octupole deformation is too large to yield any projection effect.

It is our contention that these two properties (for the isomeric state and at the top of the second fission barrier) are quite general in the actinide region. Therefore,

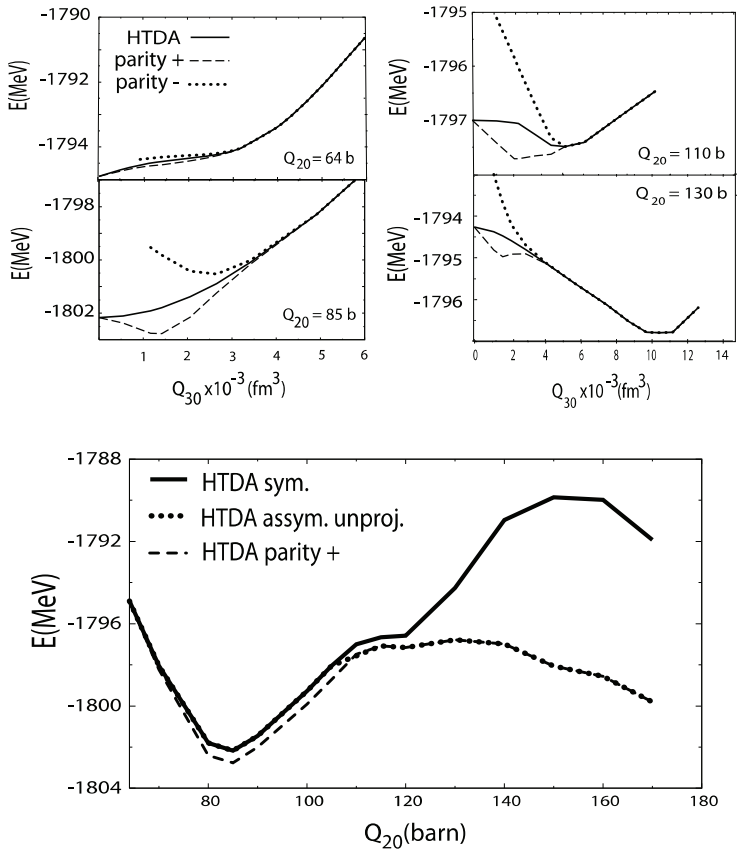

Fig. 4. Deformation energy curves for the ^{240}Pu nucleus in the fission isomeric state and second-barrier region. Upper panel: energy curves as functions of the axial octupole moments for fixed values of the axial quadrupole moment. Lower panel: resulting fission barriers.

we deem that the underestimation of such fission-barrier heights is probably a systematic effect. In our current calculations, which are somewhat approximate, this enhancement of the fission-barrier height is of about 350 keV.

4. Conclusions

We have discussed here two systematic effects leading to the conclusion that usual microscopic calculations of actinide fission-barrier heights could be underestimated by a few hundred keV for each.

One effect is related to the correlation existing between the quality of the so-called Slater approximation for the Coulomb exchange energy and the value of the proton single-particle level density. En passant, we note that this defect (being related to a purely quantal property) could not be possibly cured by a fit of EDF parameters. Moreover it induces a systematic lowering of the proton particle-hole

energy which might play a substantial role in the correlations built upon using such approximate single-particle states.

The second effect has been established from parity-restoration calculations from HTDA correlated solutions. Beyond the systematic conclusion they seem to indicate, they correspond to the first configuration-mixing calculations of such HTDA states where the advantage of their Slater determinantal character has been fully exploited.

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