Microscopic description of fission in neutron-rich radium isotopes with the Gogny energy density functional

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Abstract. Mean-field calculations, based on the D1S, D1N and D1M parametrizations of the Gogny energy density functional, have been carried out to obtain the potential energy surfaces relevant to fission in several Ra isotopes with the neutron number 144 \( \leq N \leq 176 \). Inner and outer barrier heights as well as first and second isomer excitation energies are given. The existence of a well-developed third minimum along the fission paths of Ra nuclei is analyzed in terms of the energetics of the “fragments” defining such elongated configuration. The masses and charges of the fission fragments are studied as functions of the neutron number in the parent Ra isotope. The comparison between fission and \( \alpha \)-decay half-lives, reveals that the former becomes faster for increasing neutron numbers. Though there exists a strong variance of the results with respect to the parameters used in the computation of the spontaneous fission rate, a change in tendency is observed at \( N = 164 \) with a steady increase that makes heavier neutron-rich Ra isotopes stable against fission, diminishing the importance of fission recycling in the r-process.

1 Introduction

Fission is a very challenging kind of collective motion, whose theoretical description can be addressed in terms of the evolution of a given nuclear system from its ground state to scission. To simplify this view such evolution is described in terms of several intrinsic shapes, labeled by the corresponding deformation parameters \([1–4]\). The precise description of both the fission energy landscape and the associated shell effects remains as a major challenge in nuclear structure physics, with a potential impact on several basic research and technological areas.

A detailed knowledge of the fission mechanism is required, for example, to better understand the very limits of the nuclear stability. As one goes up in atomic number \( Z \), the stability of the nucleus against fission tends to decrease, due to the increasing Coulomb repulsion, and the quantum mechanical shell effects are the only mechanism to increase the chances of survival of a given element \([5–8]\).

Fission properties of neutron-rich heavy nuclei are also relevant in the nucleosynthesis and abundances of elements with mass number greater than 120 due to competing fission recycling in the \( r \)-process in scenarios involving long neutron exposures \([9–11]\). Although induced fission is the process to understand, a preliminary description of the potential energy surfaces and spontaneous fission lifetimes of the involved nuclei can seed some light into the problem. Also, systematic studies of the fission paths and related properties, based on complementary theoretical approaches, are very useful to deepen our knowledge of the different decay channels (fission, \( \alpha \)-decay, \ldots) and their competition in heavy and super-heavy nuclei (see, for example, \([12–16]\) and references therein). On the other hand, observables like the half-lives, fragment mass and kinetic energy distributions are sensitive to the topography of the fission landscape. In turn, an improvement in the computation of those quantities would be very useful to determine the upper end of the nucleosynthesis flow \([17]\).

In addition to the physics already mentioned, nuclear fission remains a topic of high interest for reactor physics, the degradation of radioactive waste, prompt neutron-capture data from weapon tests as well as in the context of the progress made in recent years in the production of super-heavy elements (see, for example, \([1, 3, 18–20]\) and references therein).

With all this in mind, we have carried out fission calculations for the radium isotopes from the stability line to very neutron rich isotopes in order to study the evolution of the potential energy surfaces and spontaneous fission lifetimes. This work can be considered as a complement to our previous fission studies in neutron-rich U and Pu isotopes \([14, 15]\). As a side product of the calculations, several examples of second fission isomers have been found.

From the theoretical point of view several models are used to describe nuclear fission. Among them, both macroscopic-microscopic \([20–26]\) and mean-field \([8, 16, 27–36]\) ones are common choices. The approximation employed...
in this work belongs to the second class, i.e., the Hartree-Fock-Bogoliubov (HFB) [37] framework based on the highly predictive parametrizations D1S [38], D1N [39] and D1M [40] of the Gogny [41] energy density functional (EDF). In this kind of studies, the potential energy surfaces (PES) of the different paths to fission, the associated collective masses and zero-point quantum corrections provided by the mean-field calculations, have been used to compute the spontaneous fission half-lives $t_{SF}$ as well as a first approach to the masses and charges of the fission fragments based on energetics. Special attention has been paid to the uncertainties in the computation of the spontaneous fission half-lives and the impact of pairing correlations on observables. For recent complementary work, based on the Barcelona-Catania-Paris-Madrid (BCPM) EDF [42], the reader is referred to [16,43].

In this paper, we focus on the fission properties of even-even Ra isotopes with neutron numbers $144 \leq N \leq 176$ in order to examine to which extent the main features found for the neutron-rich U and Pu nuclei are still preserved down to $Z = 88$. With the aim to evaluate the robustness of our predictions, HFB calculations based on the D1S, D1N and D1M parametrizations of the Gogny-EDF have been performed. It should be kept in mind, that the D1N [39] and D1M [40] EDFs provide a better description of the nuclear masses, an aspect of relevance to the evaluation of the competing $\alpha$-decay channel. In addition, those EDFs were fitted using information on neutron matter and therefore they are expected to perform well when extrapolating to neutron-rich systems like the ones considered in the present study. On the other hand, the thoroughly tested Gogny-D1S [38] EDF, already successfully applied to fission studies in heavy and super-heavy nuclei [8,27,29], has been taken as a reference in this work. This will allow us to test the performance of the D1N and D1M Gogny-EDFs scarcely used in fission calculations up to now. We implicitly assume that the fission properties are determined by general features of the considered Gogny-EDFs and therefore, no fine tuning has been carried out.

The paper is organized as follows. In sect. 2, we briefly outline our theoretical framework. The results obtained for the fission paths, barrier heights, fission isomers, fragments’ mass and charge in $^{232-234}$Ra as well as the isotopic dependence of the spontaneous fission half-lives and the competition with the $\alpha$-decay mode are discussed in sect. 3. Special attention has been paid, in the same section, to second fission isomers (i.e., third minima) along the fission paths of the considered nuclei. Such minima have been found for several U and Pu nuclei in our recent HFB studies [14-16] and have attracted considerable attention [25-27,32,44-53]. Finally, sect. 4 is devoted to the conclusions and work perspectives.

2 Theoretical framework

In this section, we briefly outline the theoretical framework used in the present study. A detailed account of our methodology can be found in [14-16].

The main ingredient is the HFB approximation [37], that is used with constrains on the axially symmetric quadrupole $Q_{20} = z^2 - \frac{1}{2}(x^2 + y^2)$ and octupole $Q_{30} = z^3 - \frac{3}{2}z(x^2 + y^2)$ operators [14-16,43,54,55] to obtain one-fragment (1F) solutions. We are aware of the role played by triaxiality for configurations around the top of the inner barrier [14,27,34]. However, we have kept axial symmetry, as a self-consistent symmetry, along the whole fission path in order to reduce the already substantial computational effort. The $\gamma$ degree of freedom has also been neglected in the computation of the $t_{SF}$ values (see, eqs. (1) and (2) below) as it has been shown in previous studies [6,56] that its impact is very limited.

For large values of the quadrupole moment $Q_{20}$, two-fragment (2F) solutions have been reached by constraining on the necking operator $\hat{Q}_{\text{Neck}}(20,C_0)$. In computing spontaneous fission lifetimes (see below) the ridge connecting the 1F and 2F curves in the multidimensional space of deformations $(Q_{20},Q_{30},Q_{\text{Neck}},\ldots)$ has been neglected and therefore the 2F curves are considered as really intersecting the 1F ones [14-16].

Aside from the constraints already mentioned, as well as the typical HFB ones on both the proton and neutron numbers [37], a constraint on the operator $\hat{Q}_{18}$ is used to prevent spurious effects associated to the center of mass motion [54,55]. Note, that the average values of higher multipolarity deformations (i.e., $Q_{40},Q_{60},\ldots$) are automatically adjusted during the self-consistent minimization of the HFB energies. Furthermore, as a result of projecting multi-dimensional paths into one-dimensional plots (see, figs. 1 and 2) kinks and multiple branches may appear in this type of calculations [14-16,57].

The HFB quasiparticle operators are expanded in a deformed axially symmetric harmonic oscillator (HO) basis with quantum numbers restricted by the condition $2n_1 + |m| + \frac{1}{2}n_z \leq M_z$, where $M_z = 17$ and $q = 1.5$ for the whole interval of quadrupole moments. This amounts to consider states with $J_z$ quantum numbers up to $35/2$ and up to $26$ quanta in the $z$-direction. For a neutron-rich nucleus like $^{260}$Pu this basis provides an error (with respect to a larger basis with $M_z = 18$) smaller than 0.8 MeV all over the fission path [15]. In addition, for each of the considered $(Q_{20},Q_{30},Q_{\text{Neck}},\ldots)$ configurations, the HO lengths $b_1$ and $b_2$ have been optimized for each quadrupole moment so as to minimize the total HFB energy. This guarantees a much better convergence for relative energies (see, fig. 2).

For the solution of the constrained mean-field equations we have employed an approximate second order gradient method based on the parametrization of a given HFB vacuum with the help of the Thouless theorem [54,55,58]. The two-body kinetic energy correction has been fully taken into account in the Ritz-variational [37] procedure while for the Coulomb exchange term we have considered the Slater approximation. The spin-orbit contribution to the pairing field has been neglected.

We have computed the spontaneous fission half-life using the Wentzel-Kramers-Brillouin (WKB) formalism [59,60]

$$t_{SF} = 2.86 \times 10^{-21} \times (1 + e^{2S}),$$

(1)
where the action $S$ along the (minimal energy one-dimensional projected) fission path reads

$$S = \int_a^b dQ_20 \sqrt{2B(Q_20)} (V(Q_20) - (E_{GS} + E_0)) \, .$$

(2)

In the above expression, $B(Q_20)$ and $V(Q_20)$ represent the collective mass and potential for the collective variable $Q_20$. The potential energy is given by the HFB energy of the corresponding constrained state corrected by the quantum zero point vibrational $\Delta E_{vib}(Q_{20})$ and rotational $\Delta E_{\text{ROT}}(Q_{20})$ energies. Both the inertia $B(Q_{20})$ and $\Delta E_{\text{vib}}(Q_{20})$ have been computed using the cranking approximation to the Adiabatic Time Dependent HFB (ATDHF) approach [61–64] and the Gaussian Overlap Approximation (GOA) to the Generator Coordinate Method (GCM) [14–16,37] while $\Delta E_{\text{ROT}}(Q_{20})$ has been computed in terms of the Yoccoz moment of inertia $[37,65–67]$. The integration limits $a$ and $b$ in eq. (2) are the classical turning points [68] corresponding to the energy $E_{GS} + E_0$. The main effect of the free parameter $E_0$, is to determine the integration limits in the integral of the action in eq. (2) and the zero of the potential energy in the integrand. This parameter can be associated with the zero point energy of the potential well, as a function of the quadrupole moment, and defining the ground state $[4,33]$. It can be estimated as $E_0 = \sqrt{K/M}/2$ where $K$ is the curvature of the PES around the ground state and $M$ is the collective quadrupole inertia. Typical values (see below) are around 1 MeV. However, and given its impact on lifetimes we have considered four values for this parameter $E_0 = 0.5, 1.0, 1.5$ and 2.0 MeV in order to estimate its impact on lifetimes. This is particularly relevant, in the case of neutron-rich Ra isotopes which display high and wide fission barriers. The pre-factor in front of the exponential of the action in eq. (1) is given by $\ln(2)/\nu$ where $\nu$ is the assault frequency. This quantity is usually related to the angular frequency of the quadrupole motion $\nu = \omega/(2\pi)$ with $\hbar \omega = 1$ MeV. Given the connection between $\hbar \omega$ and $E_0$, this pre-factor should change accordingly to the value of $E_0$. However, this change is usually overlooked (as in the present study) given the larger uncertainties in the estimation of $t_{3\text{F}}$ coming from other sources.

Finally, in order to study the competition between the spontaneous fission and $\alpha$-decay modes, we have computed the corresponding $t_{\alpha}$ values using the Viola-Seaborg formula,

$$\log_{10} t_{\alpha} = \frac{AZ + B}{\sqrt{Q_\alpha}} + CZ + D,$$

(3)

with the parameters $A = 1.64062$, $B = -8.54399$, $C = -0.19430$ and $D = -33.9054$ as given in [13]. The $Q_\alpha$ value is obtained from the calculated binding energies for Ra and Rn nuclei. Obviously, other types of decay (for example, $\beta$-decay) may play a role in the case of heavy neutron-rich nuclei. However, their study lies outside the scope of this work.

Fig. 1. (Color online) The HFB plus the zero point rotational energies obtained with the D1S, D1N and D1M Gogny-EDFs are plotted in panel (a) as functions of the quadrupole moment $Q_{20}$ for the nucleus $^{240}$Ra. For each EDF, both the one (1F) and two-fragment (2F) solutions are included in the plot. The pairing interaction energies are depicted in panel (b) for protons (dashed lines) and neutrons (full lines). The octupole and hexadecapole moments corresponding to the 1F and 2F solutions are given in panel (c). The collective masses obtained within the ATDHF approximation are plotted in panel (d). For more details, see the main text.
Fig. 2. (Color online) The one-fragment (full lines) and two-fragment (dashed lines) HFB plus the zero point rotational energies obtained with the D1S (black), D1N (blue) and D1M (red) parametrizations of the Gogny-EDF are plotted for the nuclei $^{232}$Ra (panel (a)), $^{240}$Ra (panel (b)), $^{248}$Ra (panel (c)), $^{254}$Ra (panel (d)), $^{260}$Ra (panel (e)) and $^{264}$Ra (panel (f)) as functions of the quadrupole moment $Q_{20}$. All the relative energies are referred to the absolute minima of the corresponding one-fragment curves. For more details, see the main text.

3 Discussion of the results

An illustrative outcome of our calculations is presented in fig. 1 for the nucleus $^{240}$Ra. A similar analysis, as described below, has been carried out for each of the studied Ra isotopes. In panel (a), we have plotted the HFB energies plus the rotational corrections $E_{\text{HFB}} + \Delta E_{\text{ROT}}$ as functions of the quadrupole moment. The zero point vibrational energies are not included in the plot, as they are rather constant as functions of $Q_{20}$. However, they are always included in the computation of the spontaneous fission half-lives.

As can be seen, the Gogny-D1S EDF provides a pronounced under-binding as compared to the D1N and D1M ones. This reflects a well known deficiency of the former in heavier nuclei [14,15,69] as a result of which both the D1N and D1M parametrizations have been specially tailored in an effort to build an accurate mass table based on the
Gogny-EDF [39,40]. A similar behavior has been found in our symmetry-projected configuration mixing study of the quadrupole collectivity across the $N=126$ neutron shell closure [70] as well as in [71]. Nevertheless, it is satisfying to observe that the 1F curves provided by all the functionals are rather similar with the ground state at $Q=14$ b.

The first fission isomer appears at $Q=48$, 44 and 40 b with the D1S, D1N and D1M parametrizations. Their excitation energies are 1.74, 3.23 and 3.86 MeV, respectively. They are separated from the ground state by inner barriers with heights (without triaxiality) of 8.41, 8.44 and 8.48 MeV, respectively. Second fission isomers ($Q\approx 82$ b) are also apparent from panel (a). They lie 5.36, 5.87 and 8.26 MeV above the ground state while the heights of the second barriers ($Q\approx 64$ b) are 9.53, 11.03 and 12.67 MeV with the D1S, D1N and D1M parametrizations. Reflectio-nasymmetric second fission isomers, have already been found in previous studies [14,16,25–27,32,47–53]. As will be shown later on (see, fig. 2) they also emerge along the fission paths of several Ra isotopes.

The proton (dashed lines) and neutron (full lines) pairing correlation energies $E_{\text{pair}} = \frac{1}{2} \text{Tr}(\Delta \epsilon^s)$ are shown in panel (b). Minima are observed for the neutron pairing energies at the spherical configuration, the top of the inner and second barriers as well as for $Q=100$ b. In panel (c), we have plotted the octupole $Q_{20}$ and hexadecapole $Q_{40}$ moments corresponding to the 1F (i.e., $Q_{20}(1F)$ and $Q_{40}(1F)$) and 2F (i.e., $Q_{20}(2F)$ and $Q_{40}(2F)$) paths which are clearly separated in the multidimensional (collective) deformation space. All the multipole moments are almost superimposed for all the parametrizations considered.

The ATDHF collective masses are displayed in panel (d). Their behavior is well correlated with the one for the pairing energies shown in panel (b) and the inverse dependence of the collective mass with the square of the pairing gap [68,72]. The GCM collective masses (not shown in the figure) display a similar pattern though they are smaller than the ATDHF ones. For $^{240}$Ra and $E_0 = 1.0$ MeV, for example, this leads to pronounced differences of 10, 12 and 13 orders of magnitude in the $t_{\text{SF}}$ values predicted in the two schemes with the D1S, D1N and D1M parameter sets, respectively. These large uncertainties, are one of the main reasons driving our choice of both schemes in the computation of the spontaneous fission half-lives. In all the computations of the $t_{\text{SF}}$ values, the wiggles in the collective masses have been softened by means of a three point filter [14].

Let us also mention that, for large quadrupole moments, the 2F solutions in $^{240}$Ra correspond to a spherical $^{130}$Cd and an oblate ($\beta_2 = -0.21$) and slightly octupole deformed ($\beta_3 = 0.03$) $^{110}$Zr fragment. With the D1S, D1N and D1M Gogny-EDFs, the oblate $^{110}$Zr fragment minimizes Coulomb repulsion energies of 166.31, 166.34 and 166.55 MeV, respectively. Oblate fragments have also been found by fissioning other Ra (see, below), U and Pu [14–16] nuclei. Here, we would like to stress that fragment’s masses are the result of the minimization of the HFB energy at a given distance between the fragments and therefore should only be taken as rough approximations to the peaks of the broad fragments’ mass distribution [73]. The heavier fragment, $^{130}$Cd, is a consequence of its neutron number $N = 82$ being a magic number [15,74–76]. A more realistic description of fragments’ mass distribution [77, 78] has to take into account the dynamics of the system around the loosely defined scission configuration [14,15,73,79].

In fig. 2 we have plotted the energies $E_{\text{HFB}} + \Delta E_{\text{ROT}}$, as functions of the quadrupole moment $Q_{20}$, for $^{232}$Ra, $^{240}$Ra, $^{248}$Ra, $^{254}$Ra, $^{260}$Ra and $^{264}$Ra as a representative sample of the considered isotopes. Both the 1F (full lines) and 2F (dashed lines) paths are shown in the plots. For each isotope the relative energies are always referred to the absolute minima of the 1F curves in order to accommodate all the paths, obtained with the D1S, D1N and D1M Gogny-EDFs, in a single plot. Results for $^{240}$Ra are also included in the figure for the sake of completeness.

The ground state deformation decreases with increasing neutron number $N$ reaching its minimal value $Q_{20} = 2$ b for $^{264}$Ra. Using the corresponding ground state energies, we have computed the two-neutron separation energies $S_{2N}$ shown in fig. 3. Regardless of the considered functional, the $S_{2N}$ values exhibit a clear decreasing trend with increasing neutron number. In the case of $^{234}$Ra we have obtained $S_{2N} = 8.59, 9.42$ and 9.66 MeV with the D1S, D1N and D1M parametrizations while for $^{264}$Ra the corresponding two-neutron separation energies are $S_{2N} = 4.36, 5.48$ and 5.36 MeV, respectively. The smaller $S_{2N}$ values obtained with the D1S as compared with the D1N and D1M parametrizations (typically around 1 MeV) are due to the well known under-binding of the D1S parametrization.

The excitation energies $E_1$ of the first fission isomers and the inner barrier heights $B_1$ (without triaxiality) are summarized in fig. 4. With all the functionals, the $E_1$ values increase almost linearly as functions of the neutron number exception made of $^{252}$Ra (4.02, 3.70 and 4.71 MeV).

![Figure 3](image_url) (Color online) Two neutron separation energies $S_{2N}$ as functions of the neutron number.
with the D1S, D1N and D1M Gogny-EDFs) for which a change in tendency is observed. The barrier heights $B_I$ reach their minimal values ($4.51$, $3.85$ and $4.86$ MeV with the D1S, D1N and D1M Gogny-EDFs) for the $N = 164$ nucleus $^{252}$Ra and increase for heavier isotopes. Such an increase agrees well with the HFB predictions for neutron-rich U and Pu nuclei [14–16] and previous Extended Thomas-Fermi (ETF) results [80]. An increase is also visible in the recently reported macroscopic-microscopic $B_I$ values for even-even Ra isotopes with $166 \leq N \leq 182$ [20]. Larger $B_I$ values, together with the widening of the 1F curves, leads to larger spontaneous fission half-lives as one moves towards more neutron-rich systems (see, fig. 9).

We are aware of the reduction, by a few MeV, of the inner barrier heights $B_I$ once triaxiality is taken into account (see, for example, [34]). In fact, such a reduction has already been found in our previous Gogny-D1M HFB calculations for a set of U, Pu, Cm and Cf nuclei (see table I and fig. 4 of [14]). Though we have mainly kept calculations for a set of U, Pu, Cm and Cf nuclei (see has already been found in our previous Gogny-D1M HFB count (see, for example, [34]). In fact, such a reduction comes together with a steady increase in heavier isotopes. Note, that for neutron numbers $N \geq 164$, the predicted D1S and D1N $B_{II}$ values are rather close (see, fig. 2). The largest $E_{II}$ energies are also the ones obtained with the Gogny-D1M EDF. Here, one observes two minima (one at $N = 156$ and the other at $N = 164$ regardless of the EDF employed) and a steady increase in heavier isotopes for which the corresponding D1S and D1N $E_{II}$ values are, once more, rather close (see, fig. 2).

The previous results for $E_I$, $E_{II}$, $B_I$ and $B_{II}$ agree well with the ones obtained for U and Pu nuclei [14,15]. In particular, one sees that, regardless of the employed functional, third minima along the fission paths represent a robust feature within our Gogny-HFB framework. They are also visible in the 1F curves of the isotopes $^{232}$–$^{254}$Th for which preliminary calculations have been carried out. The same conclusions can be extracted from recent BCPM [16], Skyrme [32] and relativistic mean-field [53] studies. Therefore, the shell effects leading to second fission isomers (i.e., third minima) in this region of the nuclear chart are systematically present in all the mean-field approximations already mentioned. However, further studies are required in order to clarify the relation between the mean-field predictions and the available experimental data.

In order to further understand the origin of the extra stability leading to second fission isomers, we have performed a similar analysis to the one of [32] where the spatial density for that second isomer is compared to the densities of two nuclei summing up the same number of protons and neutrons as the parent nucleus. In order to choose the $N$ and $Z$ values used, we plot the number of

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**Fig. 4.** (Color online) Excitation energies $E_I$ of the first fission isomers and inner barrier heights $B_I$ as function of the neutron number in $^{232}$–$^{254}$Ra.

**Fig. 5.** (Color online) Excitation energies $E_{II}$ of the second fission isomers and second barrier heights $B_{II}$ as function of the neutron number in $^{232}$–$^{254}$Ra.
particles $N(z) = 2\pi \int_{-\infty}^{z} dz' \int d\mathbf{r}_\perp \rho(\mathbf{r}_\perp, z')$ up to a distance $z$. This quantity has several regions where it behaves quadratically with $z$ and a central region (corresponding to the neck) where it behaves almost linearly. By locating the mid point of that region of linear growing, we determine the $Z$ and $N$ values mentioned above. A subsequent HFB calculation for the ground state or an excited configuration (see below) allows to obtain the corresponding densities that are then shifted as to match the tips of the density of the parent nucleus. In all the cases, the densities overlap at around $z = 3$ fm where the neck is located.

In fig. 6, we have plotted the density (left-hand side of each panel) corresponding to the second fission isomer in $^{232}$Ra, $^{240}$Ra and $^{248}$Ra and the densities (right-hand side of each panel) of the two nuclei as described above. In each panel the $Z$ and $N$ values of the two nuclei are given in the lower right side. The densities computed with the D1S, D1N and D1M Gogny-EDFs are quite similar and therefore, we will focus on the results obtained with the D1S parameter set. The densities of the three nuclei show some differences as a consequence of the different number of protons and neutrons as well as the different quadrupole moment but qualitatively they look rather similar pointing to a weak dependence with neutron number. With respect to the “fragments”, the heavier one is always a rare earth nucleus with a prolate deformation in its ground state. The light “fragment” is characterized by its proton number close or equal to the magic $Z = 28$ one, that leads to a spherical ground state. In spite of being spherical, the three light “fragments” have a quadrupole potential energy surface showing a shoulder at $\beta_2 \approx 0.25–0.35$ with an excitation energy of just a couple MeV. The density of that “excited configuration” matches quite well the density of the second isomer and therefore the existence of the third minimum could be linked to shell effects in the light system associated to the above mentioned shoulder. This finding has to be confirmed by a similar analysis in other Th, U and Pu nuclei showing a second fission isomer.

Let us turn our attention, to the proton $(Z_1, Z_2)$, neutron $(N_1, N_2)$ and mass $(A_1, A_2)$ numbers of the fragments obtained at the HFB level in $^{232-264}$Ra which are plotted in fig. 7. As can be seen the overall trend is quite reminiscent of the one in U and Pu nuclei [14, 15]. The key role played by the magic neutron and proton numbers in the fragments’ masses and charges is also apparent from the figure. For example, exception made of the isotopes $^{262,264}$Ra with the parametrizations D1S and D1N, the neutron number in one of the fragments is always rather
close or equal to 82. Moreover, for $^{232-244}$Ra, the proton number in one of the fragments is close to 50 which compares well with available experimental data for this region of the nuclear chart \cite{82}. We stress, however, that such a comparison with the experiment should be taken with care because, as already discussed above for the case of $^{240}$Ra, in our calculations the masses and charges of the fragments are obtained from HFB 2F solutions minimizing the energy at the largest quadrupole deformations.

In fig. 8 we have plotted the 2F density contours for the nuclei $^{236}$Ra, $^{252}$Ra and $^{264}$Ra at $Q_{20} = 150$, 170 and 180\,b, respectively. Results are shown for the Gogny-D1M EDF but similar ones are obtained with the D1S and D1N parametrizations. The lighter and heavier fragments in $^{236}$Ra and $^{264}$Ra turn out to be oblate ($\beta_2 \approx -0.23$ and $-0.14$, respectively). In fact, our calculations predict, for example, oblate ($-0.26 \leq \beta_2 \leq -0.17$) and slightly octupole ($\beta_3 \approx 0.03$) deformed lighter fragments for the isotopes $^{232-244}$Ra. Such oblate fragments have also been found in previous HFB calculations based on both the Gogny and BCPM EDFs \cite{14–16}. A better understanding of the shell effects leading to them is required as only prolate deformations are usually assumed for the fission fragments \cite{21, 22}. On the other hand, a symmetric splitting into two $^{126}$Ru nuclei ($\beta_2 = -0.02, \beta_3 \approx 0.01$) is predicted for $^{252}$Ra. The same holds for the $N = 164$ nuclei $^{256}$U \cite{14} and $^{258}$Pu \cite{15} (two $^{128}$Pd and $^{129}$Ag fragments, respectively). Moreover, our calculations for $^{254}$Th suggest a splitting into two $^{127}$Rh fragments.

Finally, in fig. 9, we have depicted the spontaneous fission half-lives obtained for $^{232-264}$Ra as functions of the neutron number. Both the GCM and ATDHFB schemes have been employed. For each isotope we have considered four values of $E_0$ (i.e., $E_0 = 0.5, 1.0, 1.5$ and $2.0$\,MeV) and, as can be seen from the figure, an increase of this parameter leads to a decrease in the $t_{SF}$ values by several orders of magnitude. On the other hand, the ATDHFB half-lives are always larger than the GCM ones for a given $E_0$. For example, for $^{234}$Ra ($^{252}$Ra) the GCM values are $5.492 \times 10^{25}$ ($2.243 \times 10^{34}$), $2.951 \times 10^{31}$ ($4.578 \times 10^{41}$) and $5.247 \times 10^{39}$ ($2.755 \times 10^{50}$)\,s while ATDHFB ones are $5.884 \times 10^{36}$ ($2.283 \times 10^{45}$), $2.513 \times 10^{33}$ ($1.992 \times 10^{43}$) and $8.511 \times 10^{32}$ ($3.094 \times 10^{42}$)\,s with the D1S, D1N and D1M Gogny-EDFs and $E_0 = 1.0$\,MeV. In panels (d), (e) and (f) an estimation of the parameter $E_0$ based on the quadrupole curvature and inertia of the ground state (see discussion following eq. (2)) is given as a function of neutron number for the three Gogny parametrizations. The estimated values lie within the range of $E_0$ values considered. We also observe how sensitive those estimated values are to the collective inertia considered. The $t_{SF}$ values obtained with these $E_0$ values are also given, in panels (a), (b) and (c). Coming back to the differences between the results obtained with the two schemes used to compute the inertias, we notice that for the isotopes with $N \leq 166$, we have found differences of up to 11, 14 and 15 orders of magnitude between the two schemes. We observe a steady increase of the $t_{SF}$ values in heavier isotopes, with the differences between the GCM and ATDHFB predictions reaching 20, 24 and 22 orders of magnitude in the case of $^{264}$Ra with the D1S, D1N and D1M parameter sets ($E_0 = 1.0$\,MeV). The conclusion is that $t_{SF}$ values strongly depend upon the details of the calculation and the functional used with “error bars” of up to 20 orders of magnitude. However, a global trend is observed in all the cases, i.e., the slight increase of $t_{SF}$ as a function of neutron number up to $N = 166$ which is followed by a steeper increase that continues up to the largest neutron number considered, ruling out the possibility of fission recycling in the r-process. In order to examine the competition between the spontaneous fission and $\alpha$-decay modes, in fig. 9 we have also included the $\alpha$-decay half-lives eq. (3). Though the precise transition

**Fig. 7.** (Color online) The proton ($Z_1, Z_2$), neutron ($N_1, N_2$) and mass ($A_1, A_2$) numbers of the two fragments obtained in our HFB calculations for the isotopes $^{232-264}$Ra are shown as functions of the neutron number in the parent nucleus. Results have been obtained with the Gogny-D1S (panel (a)), Gogny-D1N (panel (b)) and Gogny-D1M (panel (c)) EDFs. The magic proton $Z = 50$ and neutron $N = 82$ numbers are highlighted with dashed horizontal lines to guide the eye.
Fig. 8. (Color online) Density contour plots for the nuclei $^{236}$Ra (panel (a)), $^{252}$Ra (panel b)) and $^{264}$Ra (panel (c)). The density profiles correspond to 2F solutions at the quadrupole deformations $Q_{20} = 150, 170$ and 180 b, respectively. Results are shown for the parametrization D1M of the Gogny-EDF. The density is in units of $\text{fm}^{-3}$ and contour lines are drawn at 0.01, 0.05, 0.10 and 0.15 $\text{fm}^{-3}$.

point depends of the selected Gogny-EDF, we conclude that with increasing neutron number fission turns out to be faster than $\alpha$-decay.

As expected, pairing correlations have a strong impact on the spontaneous fission half-lives obtained for the considered Ra nuclei. We have tested that, similar to previous Gogny and/or BCPM results [14–16], an increase of the pairing field by only 5 or 10% leads to a significant decrease in the predicted $t_{\text{SF}}$ values. However, even in such a case, the previous conclusion (i.e., fission dominates over $\alpha$-decay for increasing $N$) remains valid regardless of the Gogny-EDF used in the calculations.

4 Conclusions

In the present work, we have studied the fission properties of even-even Ra isotopes with neutron numbers $144 \leq N \leq 176$ within a mean-field framework [37]. With the aim to test the robustness of our HFB predictions with respect to the particular version of the Gogny-EDF [41] employed, calculations have been carried out with the parameter sets D1S [38], D1N [39] and D1M [40]. The fission paths (i.e., the 1F and 2F solutions) have been determined, for each Ra isotope, with the help of constraints on the proton $\hat{Z}$ and neutron $\hat{N}$ number operators as well as on the axially symmetric quadruple $\hat{Q}_{20}$, octupole $\hat{Q}_{30}$, $\hat{Q}_{10}$ and necking $\hat{Q}_{\text{Neck}}(z_0, C_0)$ operators. For each of the considered ($Q_{20}, Q_{30}, Q_{\text{Neck}}, \ldots$) configurations we have also performed an optimization of the lengths of the (deformed) axially symmetric HO single-particle basis in order to improve the convergence of the relative energies [14–16]. The mean-field equations have been solved using an approximate second-order gradient method that allows to handle several constraints efficiently [54, 55, 58]. Zero point vibrational and rotational corrections have always been added to both the 1F and 2F HFB energies.

Regardless of the considered Gogny-EDF, the 1F curves of the studied Ra nuclei exhibit a similar rich topology consisting of, for example, the ground-state minimum, first and second fission isomers as well as first and second barriers. A change in tendency is observed for the excitation energies of the first $E_I$ and second $E_{II}$ isomers as well as for the heights of the first $B_I$ and second $B_{II}$
bars at the neutron number $N = 164$. For some selected Ra isotopes, we have also corroborated the expected reduction of the inner barrier heights $B_I$ [14, 15, 34] once the $\gamma$ degree of freedom is taken into account. Moreover, in order to better understand the structure of the third minima along the 1F curves, we have studied their density profiles. Ours agree well with previous results [32] and suggest that those third minima in Ra nuclei could be linked to shell effects associated with an excited (deformed) configuration in a light (spherical) system with $Z \approx 28$. A more detailed study, including other Th, U and Pu nuclei showing a second fission isomer, will be presented elsewhere.

We have obtained the masses and charges of the fission fragments, from (variational) 2F solutions at the largest quadrupole moments. A symmetric splitting into two $^{126}$Ru nuclei is predicted for $^{252}$Ra as well as oblate and slightly octupole deformed fission fragments for several other Ra isotopes. Though the predicted overall trend for the fragments’ masses and charges agrees reasonably well with available data [82] for this region of the nuclear chart, we stress that our procedure tends to overestimate [14, 15] the role of the proton $Z = 50$ and neutron $N = 82$ magic numbers [74–76]. Therefore, a more sophisticated [73] approach, taking into account the quantum dynamics around the scission configurations [79], is still required.

The spontaneous fission half-lives $t_{SF}$ have been computed within the standard WKB approximation [59, 60]. Both the ATDHFB [61–64] and the GCM [14–16, 37] schemes have been employed to obtain the collective masses and the zero point vibrational corrections while for the rotational energies we have resorted to an approximate variation-after-projection (VAP) [37] in terms of the Yoccoz moment of inertia [65–67]. In spite of the large uncertainties in the predicted $t_{SF}$ values, mainly related to the details of the calculations (including the strength of pairing correlations and the Gogny-EDF used) a robust global trend is observed, i.e., the slight increase of the $t_{SF}$ values up to $N = 166$ followed by a steady increase up to $^{264}$Ra that correlates well with the one of the barrier heights and the widening of the 1F curves in heavier Ra isotopes. As a result, heavier neutron-rich Ra nuclei become stable against spontaneous fission, diminishing the importance of fission recycling in the r-process [9–11]. Furthermore, our calculations indicate that with increasing neutron number fission turns out to be faster than $\alpha$-decay.

From the results discussed in this paper we conclude that the fission properties found for neutron-rich U and

**Fig. 9.** (Color online) The spontaneous fission half-lives $t_{SF}$, predicted within the GCM and ATDHFB schemes, for the isotopes $^{232–264}$Ra are depicted as functions of the neutron number. Results have been obtained with the Gogny-D1S (panel (a)), Gogny-D1N (panel (b)) and Gogny-D1M (panel (c)) EDFs. For each parametrization, calculations have been carried out with $E_0 = 0.5$, 1.0, 1.5 and 2.0 MeV, respectively. The $\alpha$-decay half-lives are plotted with short dashed lines. In the upper panels ((d), (e) and (f)) an estimation of the parameter $E_0$, denoted $E_{0}^{est}$, is given as a function of neutron number. The $t_{SF}$ values obtained with this estimation are also included in panels (a), (b) and (c). For more details see the main text.
Pu [14–16] are preserved down to neutron-rich Ra nuclei. This motivates further explorations in this region of the nuclear chart using the Gogny-HFB framework. Within this context, a long list of tasks should be undertaken. Among them, the following two appear as our next plausible steps. First, a minimal action, instead of minimal energy, description of fission including pairing fluctuations [43] in addition to multipole moments [83] should receive more attention. However, a more realistic treatment of the vibrational mass parameters is still required within the framework presented in [43]. Here, we also refer the reader to the recent study [84] where coherent and time-feasible calculations of the vibrational masses are performed. The reader is referred to the recent study [84] where coherent and time-feasible calculations of the vibrational masses have been envisioned using the Gogny-HFB plus the quasiparticle random-phase approximation (RPA). Second, the study of the fission properties in odd-A nuclei will provide valuable information on the predictive power of our Gogny-HFB approach to account for the larger $t_{SF}$ values in those nuclear systems compared with the corresponding even-A neighbors (i.e., the hindrance factors) [2]. Here, the Gogny-HFB equal filling approximation (EFA) [85–89], represents a reasonable and computationally feasible starting point. Work along these lines is in progress.

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