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Microscopic description of quadrupole-octupole coupling in actinides with the Gogny-D1M energy density functional

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The interplay between quadrupole and octupole degrees of freedom is discussed in a series of U, Pu, Cm and Cf isotopes both at the mean-field level and beyond. In addition to the static Hartree-Fock-Bogoliubov approach, dynamical beyond-mean-field correlations are taken into account via both parity restoration and symmetry-conserving Generator Coordinate Method calculations based on the parametrization D1M of the Gogny energy density functional. Physical properties such as correlation energies, negative-parity excitation energies as well as reduced transition probabilities $B(E1)$ and $B(E3)$ are discussed in detail and compared with the available experimental data. It is shown that, for the studied nuclei, the quadrupole-octupole coupling is weak and to a large extent the properties of negative parity states can be reasonably well described in terms of the octupole degree of freedom alone.

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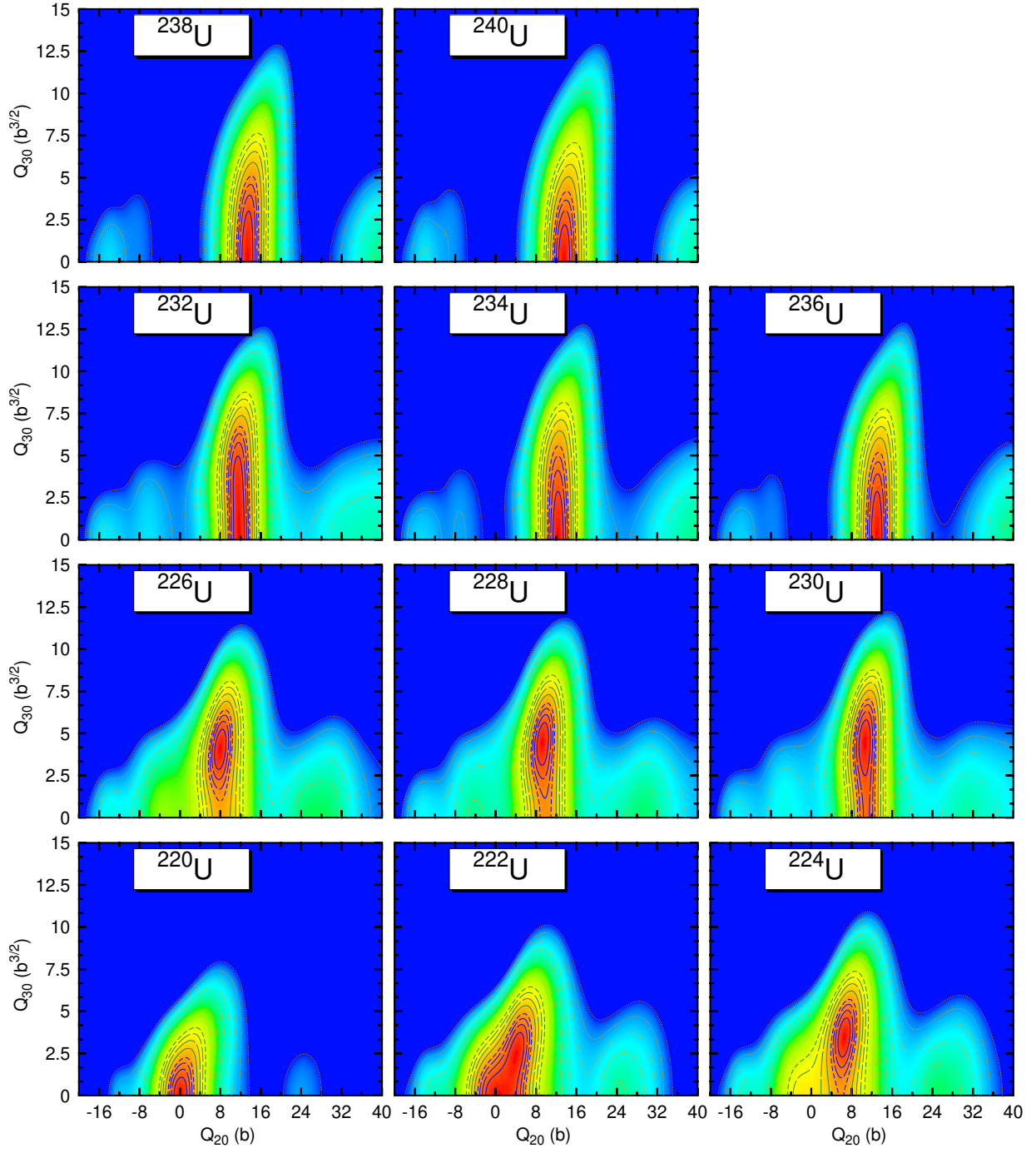


FIG. 1: (Color online) MFPEs computed with the Gogny-D1M EDF for the isotopes $^{220-240}\text{U}$. Taking the lowest mean-field energy as a reference, solid and dashed contour lines extend from 0.25 MeV up to 1 MeV in steps of 0.25 MeV. Solid and dashed contours are then drawn in steps of 0.5 MeV up to 3 MeV and from there up dotted lines are drawn in steps of 1 MeV. The intrinsic HFB energies are symmetric under the exchange $Q_{30} \rightarrow -Q_{30}$. For $A = 230$, the conversion factor from barn to β_2 values is 0.0212 and the one from $b^{3/2}$ to β_3 values is 0.0342. For additional details, see the main text.

a recent state-of-the-art quadrupole-octupole symmetry-projected configuration mixing study for ^{144}Ba [43].

Given the experimental interest in studying octupole properties of nuclei heavier than Th, we consider in the present work the dynamical interplay between quadrupole and octupole degrees of freedom in a selected set of even-even actinides, i.e., $^{220-240}\text{U}$, $^{222-242}\text{Pu}$, $^{222-242}\text{Cm}$ and $^{222-242}\text{Cf}$. These nuclei have Z values away from $Z = 88$ (Ra) which is considered to be a “magic number” for the existence of permanent octupole deformation [4]. The study of the dynamical quadrupole-octupole coupling in the selected actinide nuclei allows us to examine the role of the corresponding zero-point quantum fluctuations on the systematic of the 1^- excitation energies, transition strengths and correlation energies around the $N = 134$ (a neutron octupole magic number) isotones ^{226}U , ^{228}Pu , ^{230}Cm and ^{232}Cf .

As in our previous study [55], we consider three levels of approximation for each of the studied nuclei. The constrained Gogny-HFB scheme is used to obtain MFPEs as functions of both the quadrupole and octupole moments. As discussed later, those MFPEs can be rather soft along the octupole direction. Some of the considered nuclei also exhibit transitional features along the quadrupole direction. In this case the HFB approximation can only be considered as a starting point and beyond-mean-field correlations should be taken into account. First, parity projection is carried out in order to build the corresponding parity-projected potential energy surfaces (PPESs). Next, both symmetry restoration as well as fluctuations in the collective quadrupole and octupole coordinates are taken into account within the 2D-GCM framework. Although reflection symmetry is also restored by our GCM ansatz (see, Sec. IIC), the parity-projected results allow us to disentangle the relative contribution to the total correlation that has to be associated with the restoration of the reflection symmetry.

All the results discussed in this paper have been obtained with the Gogny-D1M EDF [47]. Among the members of the D1 family of parametrizations of the Gogny-EDF, D1S [45] has already built a strong reputation among practitioners, given its ability to reproduce a wealth of low-energy nuclear data all over the nuclear chart both at the mean-field level and beyond (see, for example, Ref. [58] and references therein). Nevertheless, the parametrization D1M, specially tailored to better describe nuclear masses, has already provided a reasonable description of nuclear properties in different regions of the nuclear chart (see, for example, Refs. [59–62] and references therein). In particular, previous studies [44, 53, 55, 57] have shown that the parametrization D1M essentially keeps the same predictive power as D1S when applied to the description of octupole properties.

The paper is organized as follows. The different approaches employed in this work are briefly outlined in Secs. IIA, IIB and IIC. In each section the results obtained with the corresponding approaches are discussed.

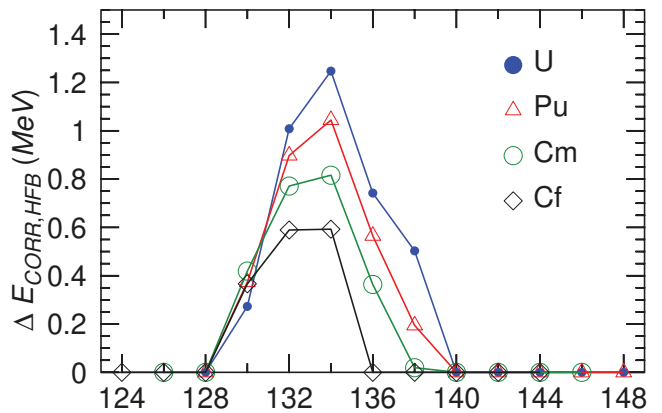


FIG. 2: (Color online) The mean-field octupole correlation energies Eq.(3) are plotted as functions of the neutron number. Results have been obtained with the Gogny-D1M EDF. For more details, see the main text.

Mean-field results are presented in Sec. IIA. We then turn our attention to beyond-mean-field properties, i.e., parity restoration and configuration mixing in Secs. IIB and IIC. Special attention is paid in Sec. IIC to 1^- energy splittings, reduced transition probabilities, correlation energies and their comparison with the available experimental data [63]. Finally, Sec. III is devoted to the concluding remarks.

II. RESULTS

The aim of this work is to study the quadrupole-octupole dynamics in a selected set of actinide nuclei. Three levels of approximation have been considered: the HFB approach [56] with constraints on the (axially symmetric) quadrupole and octupole operators, parity projection and the 2D-GCM. In what follows, we outline those approaches [55, 57], based on the Gogny-D1M EDF, and discuss the results obtained with each of them.

A. Mean-field

To obtain the MFPEs, the HFB equation with constraints on the axially symmetric quadrupole

$$\hat{Q}_{20} = z^2 - \frac{1}{2}(x^2 + y^2) \quad (1)$$

and octupole operator

$$\hat{Q}_{30} = z^3 - \frac{3}{2}(x^2 + y^2)z \quad (2)$$

is solved. The mean value with the HFB intrinsic state $|\Phi\rangle$ of the two operators define the quadrupole and octupole deformation parameters Q_{20} and Q_{30} . From them

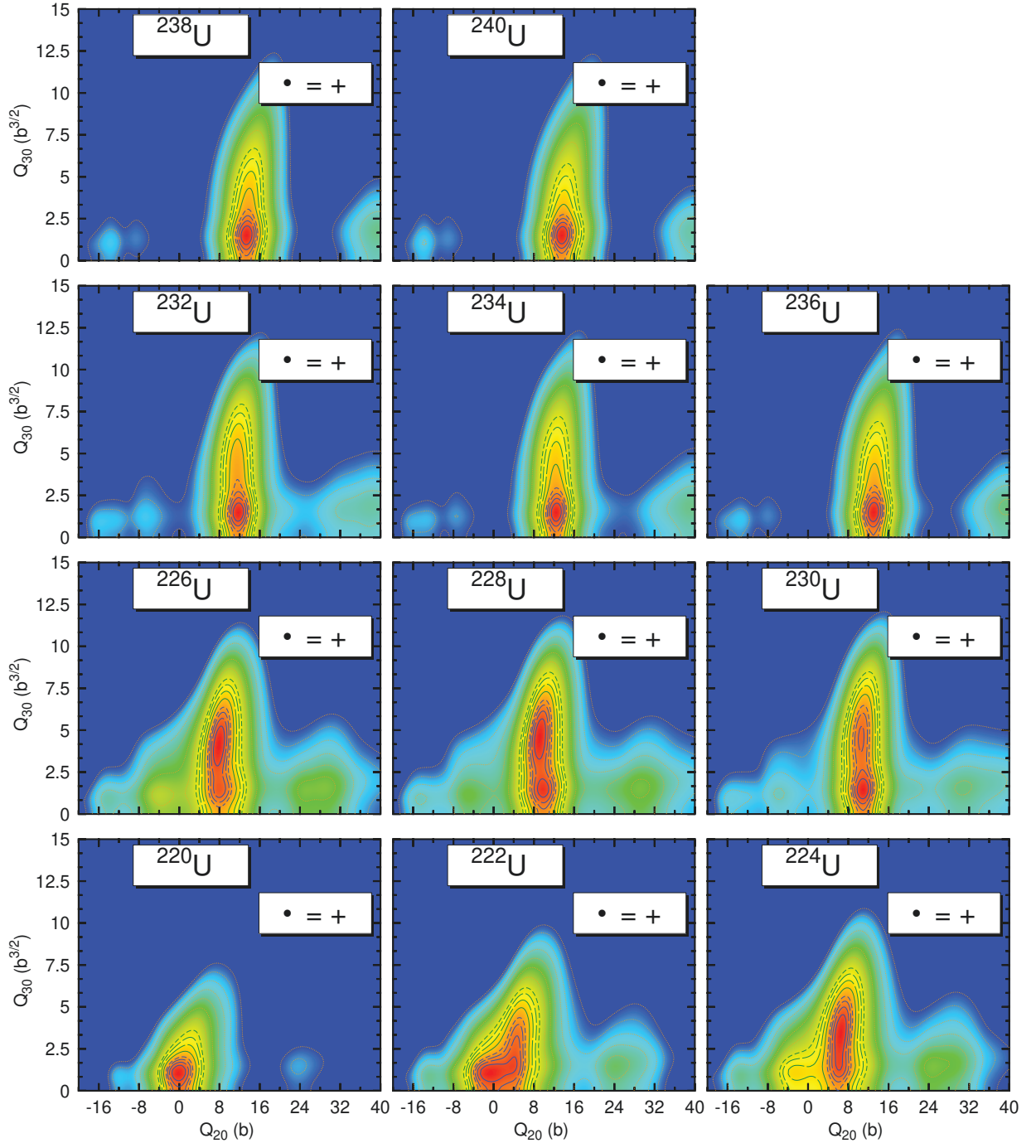


FIG. 3: (Color online) Positive $\pi = +1$ parity-projected potential energy surfaces (PPPEs) computed with the Gogny-D1M EDF for the isotopes $^{220-240}\text{U}$. See, caption of Fig. 1 for the contour-line patterns.

one can compute [35] the standard deformation parameters $\beta_l = \sqrt{4\pi(2l+1)/(3R_0^l A)} Q_{l0}$ with $R_0 = 1.2A^{1/3}$

¹ In order to alleviate the already substantial com-

putational effort, both axial and time-reversal symmetries have been kept as self-consistent symmetries. The HFB equation is solved using a performing, approximate second-order gradient method [64]. The center of mass is fixed at the origin to avoid spurious effects associated with its motion [55, 57]. The HFB quasiparticle operators [56] have been expanded in a deformed (axially symmetric) harmonic oscillator (HO) basis containing 16 major

¹ For $A = 230$ a value of $Q_{20} = 1000 \text{ fm}^2$ is equivalent to $\beta_2 = 0.212$ and a value of $Q_{30} = 1000 \text{ fm}^3$ is equivalent to $\beta_3 = 0.034$.

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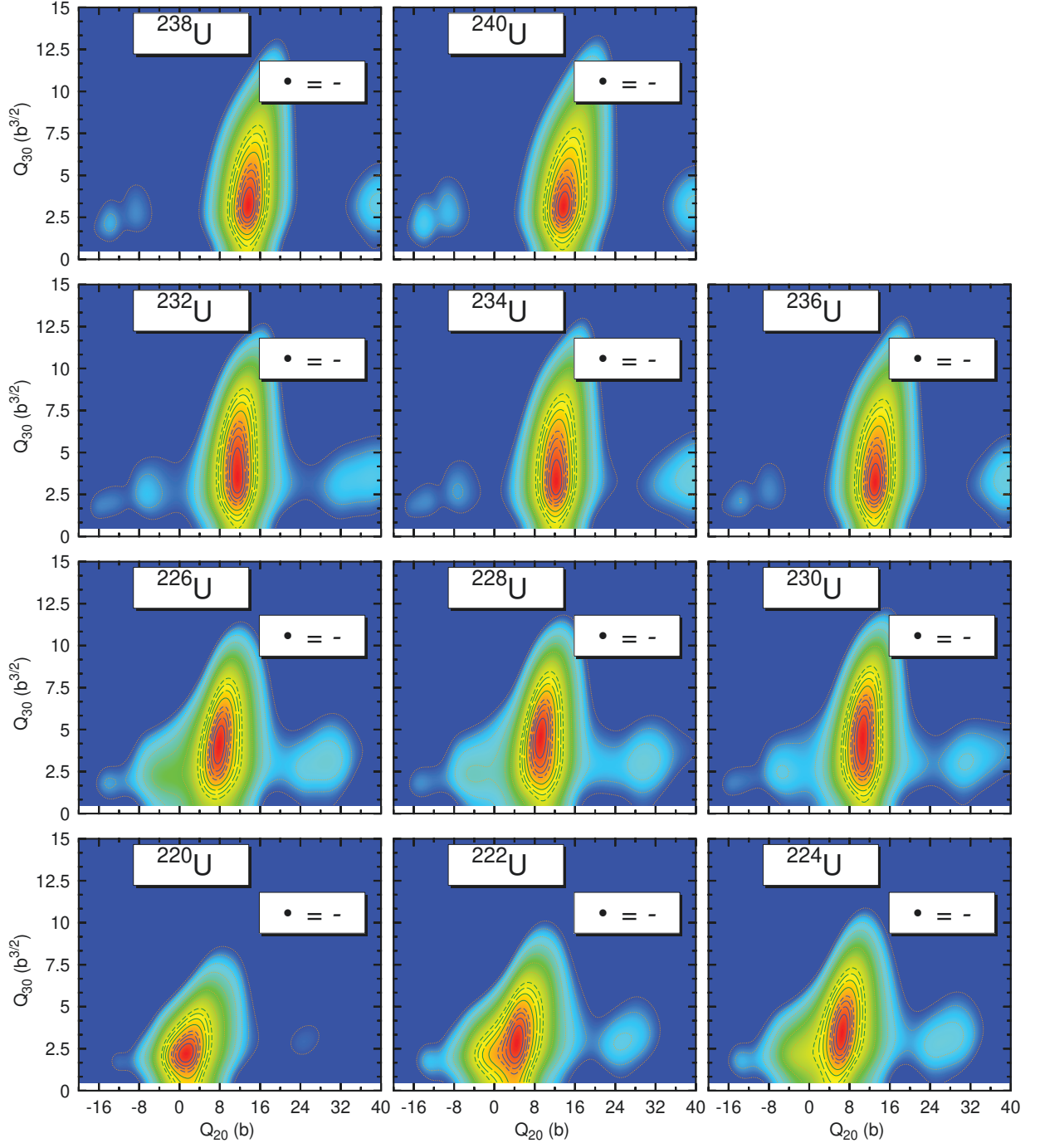


FIG. 4: (Color online) Negative $\pi = -1$ parity-projected potential energy surfaces (PPPEs) computed with the Gogny-D1M EDF for the isotopes $^{220-240}\text{U}$. See, caption of Fig. 1 for the contour-line patterns.

The $\pi = +1$ and $\pi = -1$ PPPEs obtained for the isotopes $^{220-240}\text{U}$ are depicted in Figs. 3 and 4 as illustrative examples. Along the $Q_{30} = 0$ axis, the projection onto positive parity is unnecessary as the corresponding quadrupole deformed even-even intrinsic states are already pure $\pi = +1$ states. On the other hand, in the case of negative parity, the evaluation of the projected energy along the $Q_{30} = 0$ axis requires to resolve

a "zero-over-zero" indeterminacy [32, 55]. However, the $\pi = -1$ projected energy increases rapidly when approaching $Q_{30} = 0$ (see, Fig. 5) and its limiting value does not play a significant role in the discussion of the PPPEs. We have then omitted this quantity along the $Q_{30} = 0$ axis in Fig. 4.

The absolute minima of the $\pi = +1$ and $\pi = -1$ PPPEs are located at quadrupole deformations close

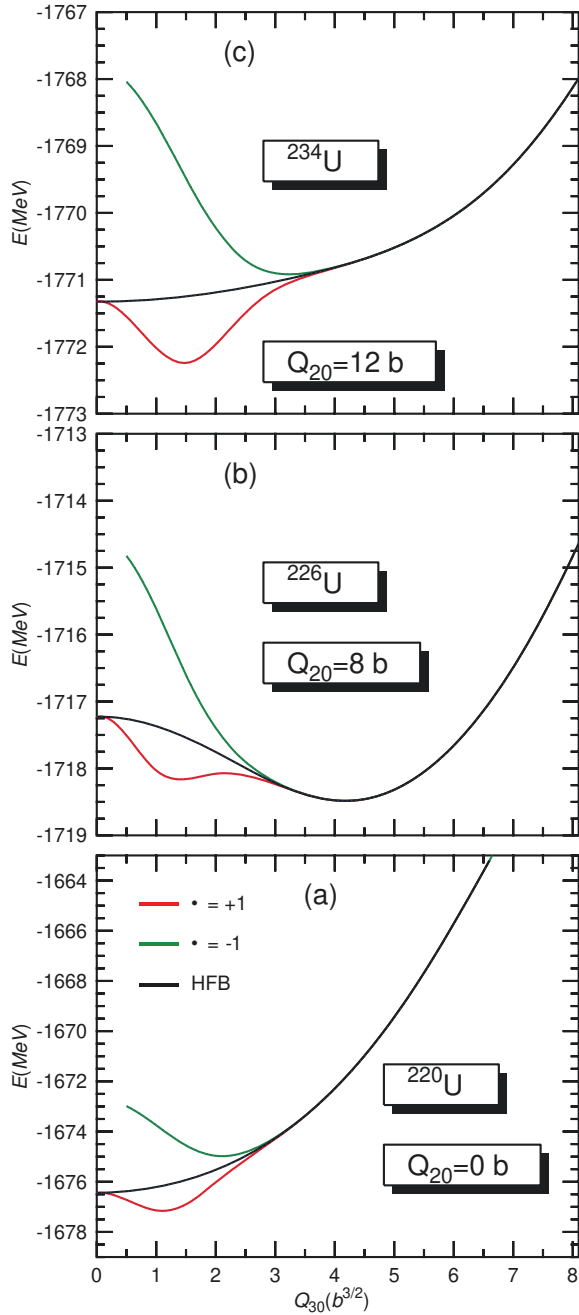


FIG. 5: (Color online) The $\pi = +1$ (red) and $\pi = -1$ (green) parity-projected energies are depicted as functions of the octupole moment Q_{30} for fixed values of the quadrupole moment Q_{20} in the nuclei ^{220}U , ^{226}U and ^{234}U . The corresponding HFB energies are also included in the plots. Results have been obtained with the Gogny-D1M EDF.

to the HFB values discussed in Sec. II A. In the case of the $\pi = +1$ PPPEs, depicted in Fig. 3, a characteristic pocket develops with a minimum at $Q_{30} = 1.0 - 1.5b^{3/2}$. In the case of nuclei with a reflection-symmetric HFB ground state, such a minimum is the global one. This is illustrated in panels (a) and (c) of Fig. 5 where the $\pi = +1$ parity-projected energies obtained for ^{220}U and

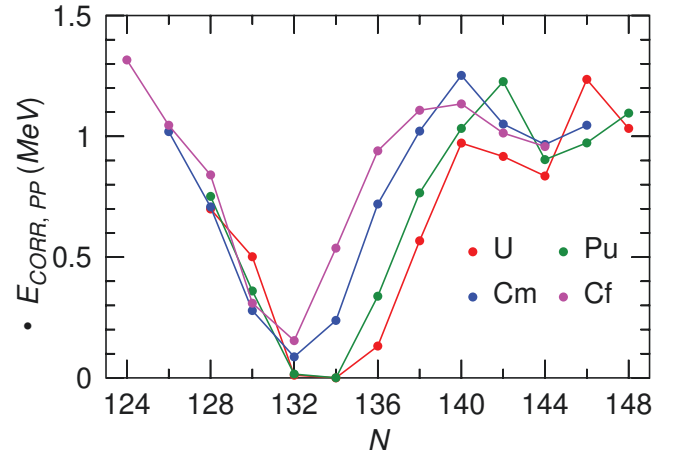


FIG. 6: (Color online) The correlation energies stemming from parity restoration Eq.(8) are plotted as functions of the neutron number. Results have been obtained with the Gogny-D1M EDF. For more details, see the main text.

^{234}U are plotted, as functions of Q_{30} , for fixed values of the quadrupole moment corresponding to the absolute minima of the PESs. On the other hand, for nuclei with a reflection-asymmetric mean-field ground state, there is a pronounced competition with a second minimum at $Q_{30} = 3.0 - 4.5b^{3/2}$ as illustrated in panel (b) of Fig. 5 for ^{226}U . In the case of ^{226}U , the global $\pi = +1$ minimum at $Q_{30} = 4.0b^{3/2}$ is only 500 KeV deeper than the one at $Q_{30} = 1.0b^{3/2}$. Similar results have been obtained for Pu, Cm and Cf isotopes. For example, the global $\pi = +1$ minima correspond to $Q_{30} = 3.0b^{3/2}$ and $4.0b^{3/2}$ in ^{226}Pu and ^{228}Pu , respectively, while for other Pu isotopes as well as for Cm and Cf nuclei they are located at $Q_{30} = 1.0 - 1.5b^{3/2}$. As can be seen from Figs. 1, 3 and 5 not only the MFPEs but also the $\pi = +1$ PPPEs are rather soft along the Q_{30} -direction.

The $\pi = -1$ PPPEs, depicted in Fig. 4, display well developed absolute minima at $Q_{30} = 2.0 - 4.5b^{3/2}$. In the case of nuclei with a reflection-symmetric HFB ground state, such as ^{220}U and ^{234}U , the absolute $\pi = -1$ minima have larger octupole deformations than the $\pi = +1$ ones [see, panels (a) and (c) of Fig. 5]. On the other hand, for some nuclei with a reflection-asymmetric HFB ground state, such as ^{226}U , the (almost degenerate) $\pi = -1$ and $\pi = +1$ absolute minima have similar octupole deformations [see, panel (b) of Fig. 5]. Similar features have been found for the other isotopic chains. Let us mention, that the complex topography along the Q_{30} -direction as well as the transition to an octupole-deformed regime found in our Gogny-D1M calculations has also been studied, as a function of the strength of the two-body interaction, in Ref. [71] using the parity-projected Lipkin-Meshkov-Glick (LMG) model.

As a measure of the correlations induced by parity symmetry restoration one can use the correlation energy, defined in terms of the difference between the HFB $E_{\text{HFB},GS}$ and parity projected $E_{\pi=+1,GS}$ ground state

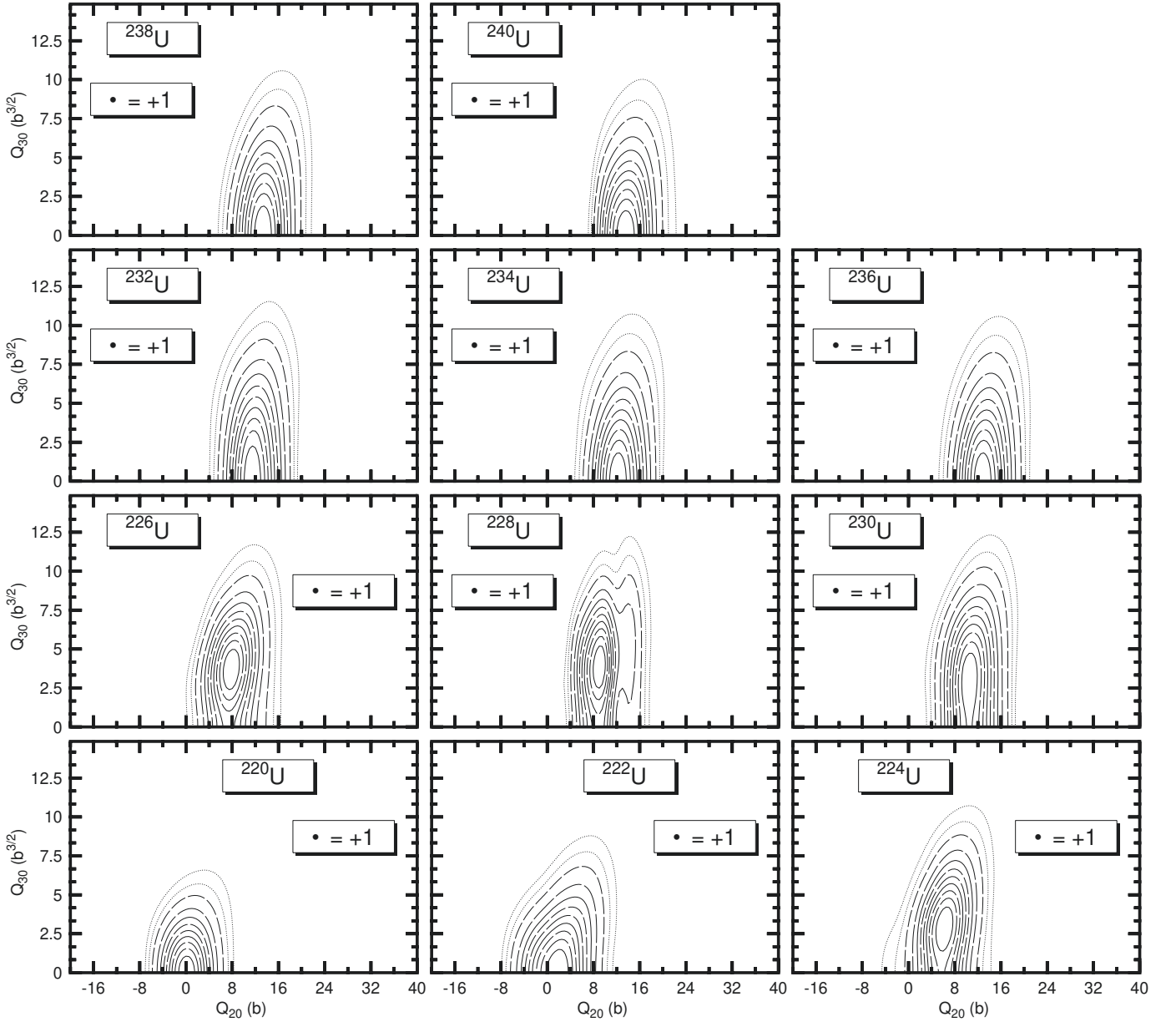


FIG. 7: Collective wave functions Eq.(13) squared for the ground states of the nuclei $^{220-240}\text{U}$. The contour lines (a succession of solid, long dashed and short dashed lines) start at 90% of the maximum value up to 10% of it. The two dotted-line contours correspond to the tail of the amplitude (15% and 1% of the maximum value). Results have been obtained with the Gogny-D1M EDF. For more details, see the main text.

energies

$$\Delta E_{\text{CORR},PP} = E_{\text{HFB},GS} - E_{\pi=+1,GS}. \quad (8)$$

In Fig. 6, we show this quantity for the different isotopes considered. The correlation energy shows a minimum around $N = 132 - 134$ corresponding to strongly octupole-deformed intrinsic states. As shown later on in Sec. II C, the comparison between the correlation energies $E_{\text{CORR},PP}$ and the ones obtained within the symmetry-conserving 2D-GCM framework (see, Fig. 9) reveals the key role played by quantum fluctuations around those neutron numbers.

C. Generator Coordinate Method

We include quantum fluctuations in the quadrupole and octupole degrees of freedom by considering a linear superposition of the HFB states $|\Phi(\mathbf{Q})\rangle$

$$|\Psi_{\sigma}^{\pi}\rangle = \int d\mathbf{Q} f_{\sigma}^{\pi}(\mathbf{Q}) |\Phi(\mathbf{Q})\rangle \quad (9)$$

where, both positive and negative octupole moments Q_{30} are included in the integration domain. In this way the parity of the collective amplitude under the change of sign

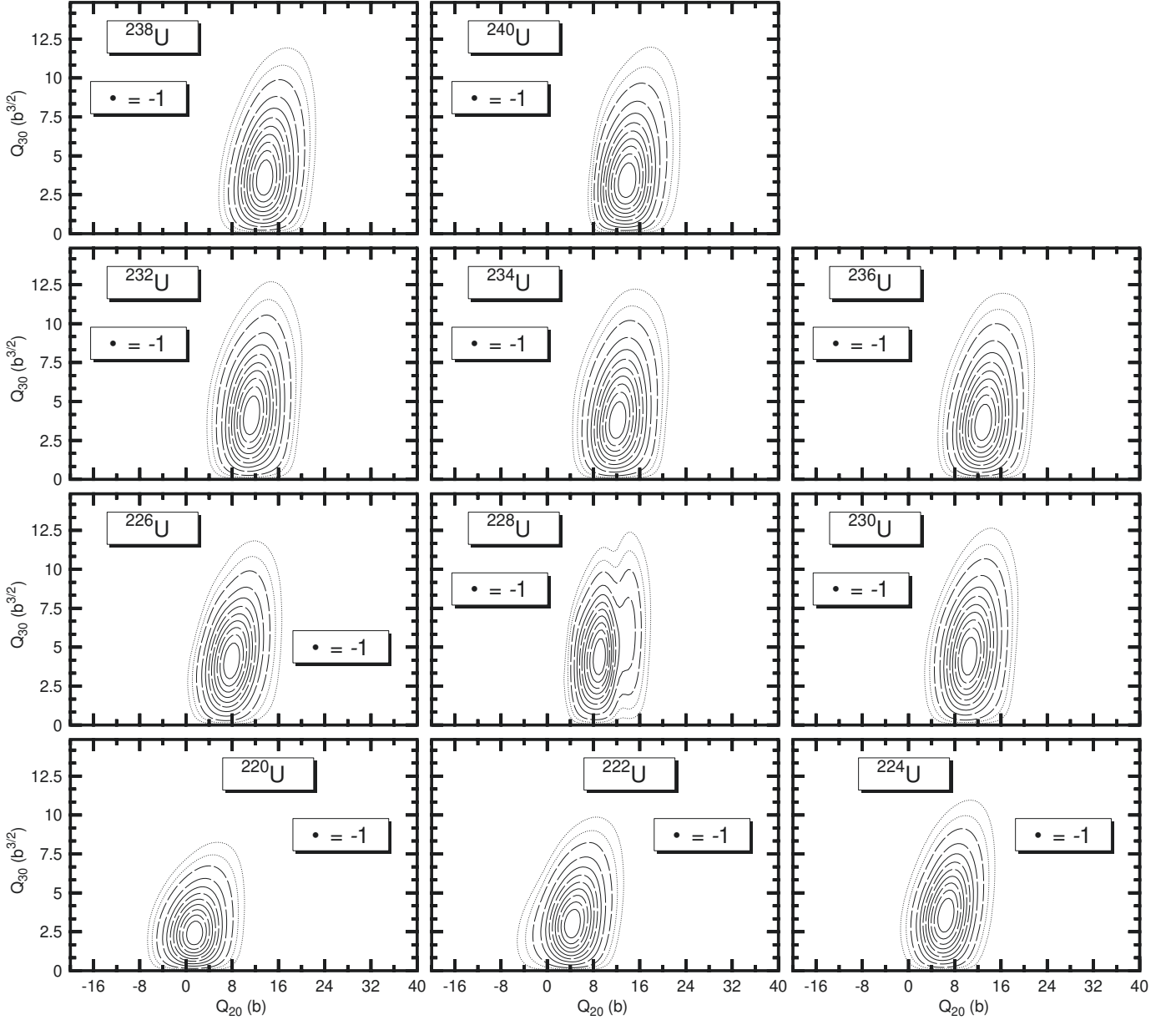


FIG. 8: Collective wave functions Eq.(13) squared for the lowest negative-parity states of the nuclei $^{220-240}\text{U}$. See, caption of Fig. 7 for contour-line patterns. Results have been obtained with the Gogny-D1M EDF. For more details, see the main text.

of Q_3 , namely $f_\sigma^\pi(Q_{20}, -Q_{30}) = \pi f_\sigma^\pi(Q_{20}, Q_{30})$, determines the parity of $|\Psi_\sigma^\pi\rangle$. The property $f_\sigma^\pi(Q_{20}, -Q_{30}) = \pi f_\sigma^\pi(Q_{20}, Q_{30})$ is a direct consequence of the invariance of the interaction under the parity symmetry operation. The index σ in Eq.(9) labels the different GCM solutions.

The amplitudes $f_\sigma^\pi(\mathbf{Q})$ are solutions of the Griffin-Hill-Wheeler (GHW) equation [56]

$$\int d\mathbf{Q}' \left(\mathcal{H}(\mathbf{Q}, \mathbf{Q}') - E_\sigma^\pi \mathcal{N}(\mathbf{Q}, \mathbf{Q}') \right) f_\sigma^\pi(\mathbf{Q}') = 0. \quad (10)$$

with the Hamiltonian and norm kernels defined in the standard way

$$\mathcal{H}(\mathbf{Q}, \mathbf{Q}') = \langle \Phi(\mathbf{Q}) | \hat{H}[\rho^{GCM}(\vec{r})] | \Phi(\mathbf{Q}') \rangle,$$

$$\mathcal{N}(\mathbf{Q}, \mathbf{Q}') = \langle \Phi(\mathbf{Q}) | \Phi(\mathbf{Q}') \rangle \quad (11)$$

In the evaluation of the Hamiltonian kernel $\mathcal{H}(\mathbf{Q}, \mathbf{Q}')$ for the Gogny-EDF, we have employed the *mixed density* prescription

$$\rho^{GCM}(\vec{r}) = \frac{\langle \Phi(\mathbf{Q}) | \hat{\rho}(\vec{r}) | \Phi(\mathbf{Q}') \rangle}{\langle \Phi(\mathbf{Q}) | \Phi(\mathbf{Q}') \rangle}. \quad (12)$$

As in the parity projection case, first-order corrections to take into account deviations in both the proton and neutron numbers [55, 57, 69, 70] are included.

The HFB basis states $|\Phi(\mathbf{Q})\rangle$ are not orthonormal. Therefore, the amplitudes $f_\sigma^\pi(\mathbf{Q})$ cannot be interpreted

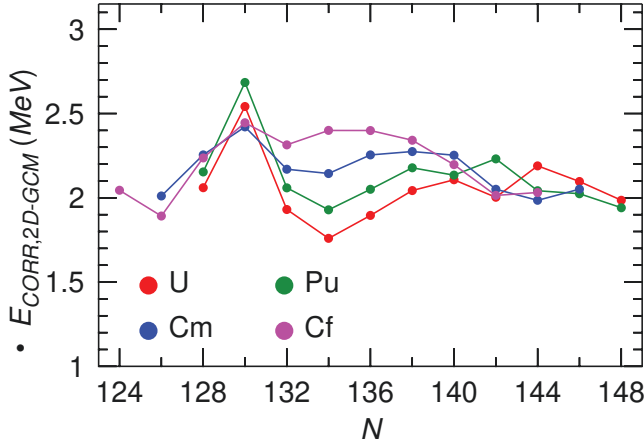


FIG. 9: (Color online) The correlation energies obtained within the 2D-GCM framework Eq.(20) are plotted as functions of the neutron number. Results have been obtained with the Gogny-D1M EDF. For more details, see the main text.

as probability amplitudes. Instead, one considers the so-called collective wave functions

$$G_{\sigma}^{\pi}(\mathbf{Q}) = \int d\mathbf{Q}' \mathcal{N}^{\frac{1}{2}}(\mathbf{Q}, \mathbf{Q}') f_{\sigma}^{\pi}(\mathbf{Q}'), \quad (13)$$

written in terms of the square root operator $\mathcal{N}^{\frac{1}{2}}(\mathbf{Q}, \mathbf{Q}')$ of the norm kernel [55, 56, 65] defined by the property

$$\mathcal{N}(\mathbf{Q}; \mathbf{Q}') = \int d\mathbf{Q}'' \mathcal{N}^{\frac{1}{2}}(\mathbf{Q}; \mathbf{Q}'') \mathcal{N}^{\frac{1}{2}}(\mathbf{Q}''; \mathbf{Q}') \quad (14)$$

The overlap $\langle \Psi_{\sigma}^{\pi} | \hat{O} | \Psi_{\sigma'}^{\pi'} \rangle$ of an operator \hat{O} between two different GCM states Eq.(9) is required in the computation of physical quantities such as, for example, the electromagnetic transition probabilities. It reads

$$\langle \Psi_{\sigma}^{\pi} | \hat{O} | \Psi_{\sigma'}^{\pi'} \rangle = \int d\mathbf{Q} d\mathbf{Q}' G_{\sigma}^{\pi*}(\mathbf{Q}) \mathcal{O}(\mathbf{Q}, \mathbf{Q}') G_{\sigma'}^{\pi'}(\mathbf{Q}') \quad (15)$$

where

$$\begin{aligned} \mathcal{O}(\mathbf{Q}, \mathbf{Q}') &= \int d\mathbf{Q}'' d\mathbf{Q}''' \mathcal{N}^{-\frac{1}{2}}(\mathbf{Q}; \mathbf{Q}'') \langle \mathbf{Q}'' | \hat{O} | \mathbf{Q}''' \rangle \times \\ &\times \mathcal{N}^{-\frac{1}{2}}(\mathbf{Q}'''; \mathbf{Q}') \end{aligned} \quad (16)$$

For the reduced transition probabilities $B(E1, 1^{-} \rightarrow 0^{+})$ and $B(E3, 3^{-} \rightarrow 0^{+})$ the rotational formula for K=0 bands have been used

$$B(E\lambda, \lambda^{-} \rightarrow 0^{+}) = \frac{e^2}{4\pi} \langle \Psi_{\sigma}^{\pi=-1} | \hat{O}_{\lambda} | \Psi_{\sigma'=1}^{\pi'=+1} \rangle^2. \quad (17)$$

For $B(E1)$ and $B(E3)$ transitions σ corresponds to the first excited GCM state with negative parity. The electromagnetic transition operators \hat{O}_1 and \hat{O}_3 are the dipole moment operator and the proton component of the octupole operator, respectively [55].

Some comments are in order here regarding the use of Eq.(17). Previous studies [72, 73] have revealed that the

use of proper angular momentum projected (AMP) wave functions concurs in an enhancement of the $E3$ strengths in spherical and/or weakly quadrupole deformed nuclei as compared to the strength obtained with the rotational formula implicit in Eq.(17). On the other hand, the $E1$ transitions do not show a clear pattern due to their less collective nature. With this in mind, the $E3$ strengths obtained in our calculations for spherical and/or weakly deformed $N \approx 126$ nuclei via Eq.(17), should be viewed as lower bounds.

The collective wave functions Eq.(13) squared corresponding to the ground and lowest negative parity 2D-GCM states in $^{220-240}\text{U}$ are plotted in Figs. 7 and 8, respectively. As can be seen from Fig. 7, the ground state collective amplitudes $|G_{\sigma=1}^{\pi=+1}(Q_{20}, Q_{30})|^2$ reach global maxima for octupole moments different from zero only in $^{224-230}\text{U}$. The same holds for $^{226-232}\text{Pu}$ and $^{228,230}\text{Cm}$ while for other U, Pu and Cf nuclei, the peaks are located around $Q_{30} = 0$. As illustrated in Fig. 7, the spreading of the amplitudes $|G_{\sigma=1}^{\pi=+1}(Q_{20}, Q_{30})|^2$ along the Q_{30} -direction is large, indicating the octupole-soft character of the $\pi = +1$ 2D-GCM ground states. In the case of the $\pi = -1$ amplitudes, depicted in Fig. 8, the maxima are always located at a nonzero octupole moment as could be anticipated from the behavior of the $\pi = -1$ PPPEs (see, Fig. 4).

Using Eq.(15), we have computed the 2D-GCM average quadrupole moments

$$(\bar{Q}_{20})_{\sigma}^{\pi} = \langle \Psi_{\sigma}^{\pi} | \hat{Q}_{20} | \Psi_{\sigma}^{\pi} \rangle. \quad (18)$$

In the case of a negative-parity operator like \hat{Q}_{30} the quantity $\langle \Psi_{\sigma}^{\pi} | \hat{Q}_{30} | \Psi_{\sigma}^{\pi} \rangle$ is zero by construction. Therefore, a meaningful averaged quantity has to be defined [55] by restricting the integration domain \mathcal{D} to positive values of Q_{30} and Q'_{30}

$$(\bar{Q}_{30})_{\sigma}^{\pi} = 4 \int_{\mathcal{D}} d\mathbf{Q} d\mathbf{Q}' G_{\sigma}^{\pi*}(\mathbf{Q}) Q_{30}(\mathbf{Q}, \mathbf{Q}') G_{\sigma}^{\pi}(\mathbf{Q}') \quad (19)$$

In the case of a strongly peaked collective inertia, the average octupole moment \bar{Q}_{30} is a good estimator of the location of the peak.

The ground-state dynamical quadrupole moments $(\bar{Q}_{20})_{\sigma=1}^{\pi=+1}$ increase as more neutrons are added along a given isotopic chain and their values remain close to the ones predicted at the HFB level. On the other hand, at variance with the HFB results, once both $\pi = +1$ symmetry restoration and (Q_{20}, Q_{30}) -fluctuations are considered at the 2D-GCM level, dynamical octupole deformations $0.53b^{3/2} \leq (\bar{Q}_{30})_{\sigma=1}^{\pi=+1} \leq 2.15b^{3/2}$ are found in the ground states of all the studied nuclei with the largest values corresponding to isotopes with neutron numbers $N = 132 - 138$. The quadrupole moments $(\bar{Q}_{20})_{\sigma}^{\pi=-1}$ corresponding to the lowest negative-parity states also increase their values with increasing N . Moreover, the corresponding average octupole moments lie within the range $1.87b^{3/2} \leq (\bar{Q}_{30})_{\sigma}^{\pi=-1} \leq 3.75b^{3/2}$ with their largest values being reached once more for $N = 132 - 138$ isotopes.

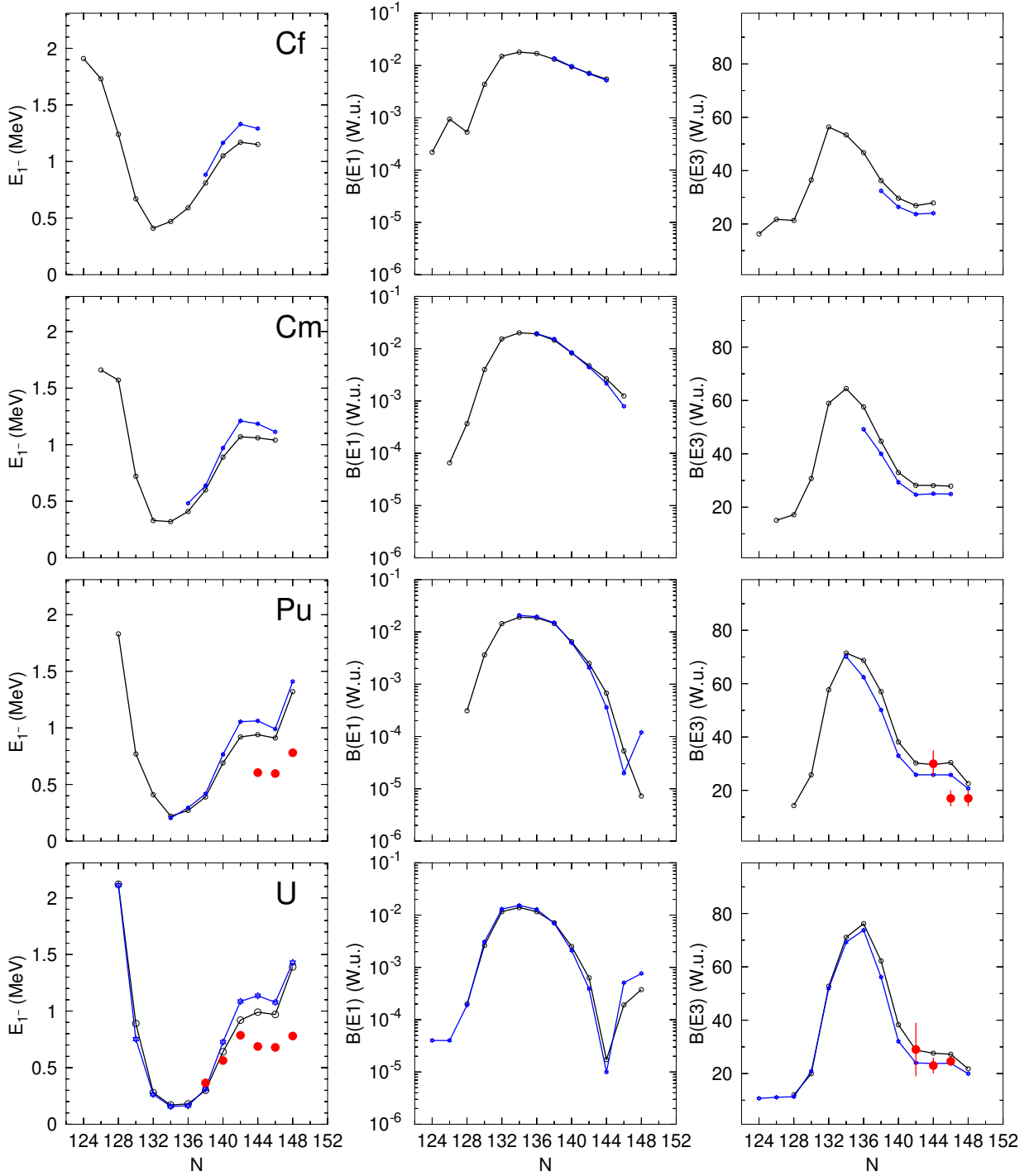


FIG. 10: (Color online) The 2D-GCM E_{1-} energy splittings (left panels) and the reduced transition probabilities $B(E1)$ (middle panels) and $B(E3)$ (right panels) are plotted (in black) as functions of the neutron number for the studied U, Pu, Cm and Cf isotopic chains. The available experimental data (in red) have been taken from Ref. [63]. The E_{1-} , $B(E1)$ and $B(E3)$ values obtained in the framework of the 1D-GCM [53], with the octupole moment as single generating coordinate, have also been included (in blue) in each of the plots. Results have been obtained with the Gogny-D1M EDF. For more details, see the main text.

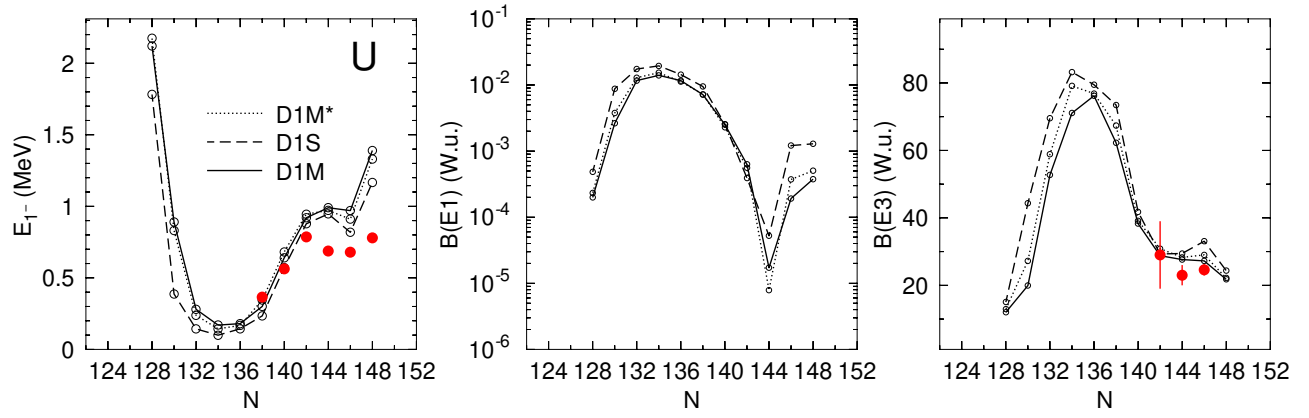


FIG. 11: (Color online) Same as Fig 10 but for different parametrizations of the Gogny force (D1M full line, D1M* dotted line and D1S dashed line).

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