

Time-Dependent Density Functional Theory Description of $^{238}\text{U}(\text{n},\text{f})$, $^{240,242}\text{Pu}(\text{n},\text{f})$, and $^{237}\text{Np}(\text{n},\text{f})$ Reactions

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In nuclei with an odd nucleon number the nonvanishing spin number density is the source of a pseudomagnetic field, which favors the splitting of the nucleon Cooper pairs. Such a pseudomagnetic field is generated always in the dynamics of any nucleus, but its effects on Cooper pairs are significantly enhanced in the dynamic evolution of nuclei with an odd number of nucleons. We present for the first time a microscopic study of the induced fission of the odd neutron compound nuclei ^{239}U , $^{241,243}\text{Pu}$, and the odd proton, odd neutron compound nucleus ^{238}Np , performed within the time-dependent density functional theory extended to superfluid fermion systems, without any simplifying assumptions, with controlled numerical approximations, and for a very large number of initial conditions. Because of the presence of the unpaired odd nucleon(s), the time-reversal symmetry of the fission compound nucleus is spontaneously broken, an aspect routinely neglected in the most advanced microscopic approaches of the past. The emerging fission fragment properties are quite similar to the properties of fission fragments of neighboring even-even nuclei. The time from saddle-to-scission is often significantly longer in odd-odd or odd-mass nuclei than for even-even nuclei, since systems with unpaired nucleons are easier to excite and the potential energy surfaces of these nuclei have more structure, often resembling a very complicated obstacle course, rather than a more direct evolution of the nuclear shape from the top of the outer fission barrier to the scission configuration. The Pauli blocking approximation, often invoked in the literature, expected to inhibit the fission of nuclei with unpaired nucleons, is surprisingly strongly violated during the fission dynamics.

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Nuclei with an odd number of neutrons and/or protons have much higher level densities at the same excitation energy than those with even numbers of both nucleon types. This is true not only near the ground state but also along the often-times circuitous path a nucleus takes from a compact shape to fission. This includes navigation through the critical barrier regions and onto ultimate scission. The oddness of the individual nucleon numbers leads to closely spaced potential energy surfaces and thus to an enhanced opportunity to jump from one surface to another. This is similar to the situation known for a long time from chemistry studies [1–3]. For the excitation energies of the compound nuclei in induced fission with low-energy neutrons [4,5], even near the top of the outer fission barrier, the nucleus is “cold” according to 1956 Bohr’s analysis [6], and the number of accessible fission channels is still large in odd-mass or odd-odd nuclei. The theoretical difficulties in dealing with this problem has lead to a dearth of

treatments of the fission of systems with oddness, with simplifications and assumptions not sufficiently well underpinned by theory. The importance of the oddness issue was manifest when the first reactor produced plutonium for the Manhattan Project. While the microscopic amounts of pure ^{239}Pu were produced with a cyclotron, which has negligible spontaneous fission, the reactor produced also ^{240}Pu , which has sufficient spontaneous fission that predetonation became a concern, that required implosion to circumvent. This is not an isolated case; the systematics of spontaneous fission half-lives of even-even (E-E) versus odd-even (O-E), even-odd (E-O), and odd-odd (O-O) compound nuclei is presented in every discussion of fission [6–9]. The spontaneous fission half-lives of even-even nuclei are several orders of magnitude shorter than the spontaneous fission half-lives of E-O, O-E, or O-O nuclei, even though the fission barriers are very similar [6].

In nuclei with odd numbers of neutrons and/or protons, new order parameters appear in the static mean fields, proportional to the spatial spin number density (a quantity similar to a local spin magnetization) and a pseudo-magnetic field emerges $\mathbf{B}(\mathbf{r}) \propto \mathbf{s}(\mathbf{r})$ [10], which impacts the nucleon spins in a manner similar to a magnetic field acting on a spin 1/2-particle $\mathbf{B}(\mathbf{r}) \cdot \boldsymbol{\sigma} \propto \mathbf{s}(\mathbf{r}) \cdot \boldsymbol{\sigma}$. These new

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pseudomagnetic fields are axial vectors, which break both the rotational and time-reversal symmetry. Such pseudomagnetic fields appear also in the dynamic evolution of E-E systems, e.g., in induced fission, which is a highly non-equilibrium and dissipative process [3,11–13]. As we demonstrate in this Letter, in the case of induced fission of compound nuclei with an odd number of neutrons and/or protons, their effect is significantly stronger than in the case of induced fission of E-E compound nuclei. As established by Caldeira and Leggett [14], tunneling times are dramatically affected by dissipation, and it is expected that dissipation will manifest itself differently in nuclei with even and odd nucleons. The time-reversal properties of the many-body wave functions are also crucial in describing β decay of fission fragments (FFs) and P -even and T -odd asymmetries of angular correlations of FFs with light particles emitted in induced fission with cold polarized neutrons [15].

The microscopic treatment of the low-energy structure and dynamics of odd- A and O-O nuclei has always presented significant technical difficulties compared to E-E nuclei. An extra odd nucleon cannot participate in a Cooper pair, and the corresponding single-particle state cannot contribute to the anomalous density (the superfluid order parameter) [16–18], thus inhibiting pairing correlations compared to neighboring even-even nuclei. It has been established microscopically, in full agreement with the experimental findings, that the induced fission dynamics of E-E nuclei has a strongly nonequilibrium character [3,11,12], now accepted by the community [19]. At the same time, pairing correlations play a crucial role in the evolution of the nuclear shape of the compound nucleus [20–25]. In the absence of pairing correlations, fission dynamics is strongly slowed and often brought to a stop, while by artificially increasing their strength, the dynamics is accelerated by at least an order of magnitude and the dynamics is mostly adiabatic [12].

In the literature, two different approximations are typically used to treat pairing in nuclei with odd nucleon numbers in static calculations: the Pauli blocking approximation [26] and the equal filling approximation (EFA) [27]. In the presence of pairing correlations, the many-body wave functions of systems with even or odd numbers of protons or neutrons do not have a well-defined particle number in the Hartree-Fock-Bogoliubov (HFB) mean-field approximation. In the Pauli blocking approximation the corresponding quasiparticle level of the odd nucleon does not contribute to the anomalous density, but it does contribute to all the other one-body number densities, and this leads to the spontaneous breaking of the time-reversal and particle number symmetries, and along with them also the rotational symmetry is also often broken. On the other hand, in the EFA the nucleus is treated as an even-even nucleus, with exactly vanishing pseudomagnetic fields $\mathbf{B}(\mathbf{r}) \propto \mathbf{s}(\mathbf{r}) \equiv 0$ and no time-symmetry breaking

terms, but with an average particle number corresponding to the corresponding odd number of nucleons [27]. In the EFA pair correlations are expected to be stronger than in the Pauli blocking approximation, since the odd nucleon(s) contribute to the pairing correlations in the EFA.

The most recent and detailed microscopic descriptions of the fission of odd- A nuclei were obtained within the EFA and were reported in Refs. [28,29], where brief reviews of older and simplified theoretical studies can be found. In Refs. [28,29] odd-nucleon systems have been treated within the density functional theory (DFT) framework as even-even within the EFA approximation, thus with unbroken time-reversal symmetry and even particle parity. One might argue that the inaccuracy of adopting either the Pauli blocking approximation or the EFA is of the order $1/N$ or $1/Z$, respectively, but this is in clear conflict with at least one observable, in particular the fact that spontaneous fission half-lives of even-even on odd-nucleon nuclei differ by orders of magnitude [6]. The single-particle occupation probabilities are double degenerate in odd-nucleon systems in the static calculations, with the exception of the Pauli blocked state. Under time evolution, these doublets are no longer double degenerate, due to the presence of the induced pseudomagnetic fields; see the Supplemental Material [18]. The effect of these induced pseudomagnetic fields is also significantly more pronounced in compound nuclei with odd number of nucleons than in E-E compound systems.

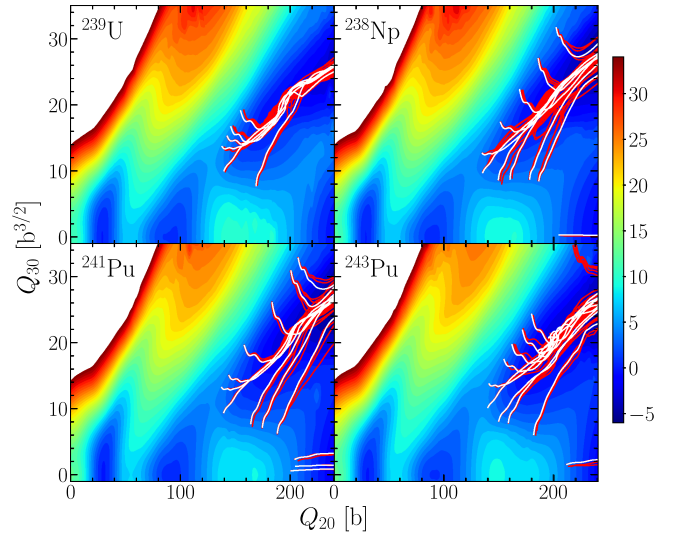


FIG. 1. For the four nuclei with odd number of protons and/or neutrons reported here we show the individual TDSLDA trajectories for the even-even seed nuclei (white) and for the odd-nucleon nuclei (red) on the initial PES of the “seed” even-even nucleus. The symmetric trajectories, for relatively small initial $Q_{30}(t=0)$ extend beyond the limits of each plot and they are shown in Fig. 3. The initial conditions have been chosen along the rim of the outer fission barrier as in previous studies [3,11,12,49–51].

In this Letter we present the induced fission of odd- A compound nuclei ^{239}U (23 trajectories), ^{241}Pu (43 trajectories), ^{243}Pu (35 trajectories), and of odd-odd nucleus ^{238}Np (48 trajectories) within time-dependent density functional theory extended to include pairing correlations, dubbed the time-dependent superfluid local density approximation (TDSLDA) [30,31], without any assumptions or uncontrolled approximations. In total we have simulated 149 trajectories for these nuclei, considering various initial conditions corresponding to using different quasiparticle states for the odd nucleon and various initial quadrupole and octupole deformations (Q_{20} , Q_{30}) of the initial nucleus outside the outer fission barrier [18]. The initial states were generated by considering a “seed” even-even (E-E) nucleus with the average N and Z identical to the corresponding target odd mass (O-E) or odd-odd (O-O) nucleus, and thus the volume, surface, Coulomb, and symmetry energies are essentially identical. The only difference between these nuclei left being the small change(s) in the pairing gap(s) and the particle parity of the corresponding many-body wave functions [18]. In order to contrast the time evolution of even-even nuclei versus those with an odd number of protons and/or neutrons, for each set of initial deformations Q_{20} , Q_{30} of the “seed” nuclei we analyzed the corresponding fission dynamics of even-even seed nuclei

^{239}U (6 trajectories), ^{241}Pu (13 trajectories), ^{243}Pu (10 trajectories), and ^{238}Np (11 trajectories); see Fig. 1 and the Supplemental Material [18]. We used then the prescription outlined in Refs. [26,28,29,32] and flipped various low-energy quasiparticle states with occupation probability $n_k \approx 0.5 \pm 0.2$ to create odd-nucleon systems, with a small number of exceptions; see the Supplemental Material [18], which contains also Refs. [13,33–48].

The energy density functional (EDF) SeaLL1 [41] used here is a typical modern accurate EDF, constructed to describe overwhelmingly even-even nuclei only; see Ref. [52] for one of the most recent studies and for earlier references. Most of the time-reversal symmetry contributions to an EDF, however, arise from the presence of an odd number of nucleons and their magnitudes are known only from a limited number of studies [10,41,53]. Thus, one can expect that the actual role of time-reversal symmetry-breaking terms in induced fission in particular could be significantly larger than what we find here.

When comparing the FF properties for $^{238}\text{U}(n, f)$, $^{240,242}\text{Pu}(n, f)$, and $^{237}\text{Np}(n, f)$ reactions to those emerging from fission of E-E compound nuclei, remarkable differences can be observed in Table I. O-E and O-O systems have total kinetic energy (TKE) distributions about twice as broad as those of E-E nuclei ^{236}U , ^{240}Pu or seed

TABLE I. The TKE, the FF proton and mass numbers, and the number of runs performed in each case and their standard deviations in parentheses. We categorize the runs as “symmetric” and “asymmetric” depending on the initial value of the octupole deformation of the compound nucleus Q_{30} being smaller or larger than $\approx 2.5 \text{ b}^{3/2}$. In case of $^{235}\text{U}(n, f)$ and $^{239}\text{Pu}(n, f)$ we have added the corresponding TKE and the FF proton and mass numbers obtained in our previous papers [50,51,54], but never reported until now. Our results for TKE are within 1 MeV from Madland systematics for $^{235,238}\text{U}(n, f)$ and $^{239}\text{Pu}(n, f)$ [55]. Average properties of FFs can be compared against CGMF results [18], which in turn are based on available experimental data [56]. A direct comparison with experimental data for all observables is challenging given the corrections that enter into the experimental analysis to extract the preneutron emission information.

	Treatment	Type	TKE	Z_H	Z_L	A_H	A_L	No. Runs
$^{235}\text{U}(n, f)$	E – E	Asymmetric	168.71(4.03)	52.07(0.84)	39.94(0.84)	134.85(2.24)	101.16(2.24)	22
$^{235}\text{U}(n, f)$	E – E	Symmetric	142.35(3.05)	47.36(1.35)	44.64(1.35)	121.60(3.58)	114.40(3.58)	2
$^{237}\text{Np}(n, f)$	O – O	Asymmetric	174.68(4.34)	52.34(0.56)	40.66(0.53)	135.11(1.45)	102.90(1.35)	46
$^{237}\text{Np}(n, f)$	E – E	Asymmetric	175.36(2.75)	52.18(0.27)	40.84(0.27)	134.52(0.73)	103.50(0.72)	10
$^{237}\text{Np}(n, f)$	O – O	Symmetric	151.04(1.30)	49.53(0.05)	43.50(0.05)	127.37(0.05)	110.69(0.02)	2
$^{237}\text{Np}(n, f)$	E – E	Symmetric	148.24	49.90	43.10	128.36	109.66	1
$^{238}\text{U}(n, f)$	O – E	Asymmetric	169.08(3.56)	51.85(0.49)	40.15(0.49)	135.43(1.29)	103.54(1.32)	23
$^{238}\text{U}(n, f)$	E – E	Asymmetric	169.40(1.36)	51.60(0.32)	40.40(0.32)	134.79(0.80)	104.21(0.80)	6
$^{239}\text{Pu}(n, f)$	E – E	Asymmetric	176.85(1.00)	53.09(0.21)	40.91(0.21)	136.49(0.43)	103.51(0.43)	5
$^{239}\text{Pu}(n, f)$	E – E	Symmetric	150.03(1.16)	49.87(1.02)	44.13(1.02)	128.46(2.57)	111.54(2.57)	2
$^{240}\text{Pu}(n, f)$	O – E	Asymmetric	173.88(4.18)	52.71(0.75)	41.29(0.75)	136.03(1.79)	104.93(1.80)	36
$^{240}\text{Pu}(n, f)$	E – E	Asymmetric	173.98(3.00)	52.79(0.75)	41.21(0.75)	136.15(1.66)	104.85(1.66)	10
$^{240}\text{Pu}(n, f)$	O – E	Symmetric	151.58(3.59)	50.70(0.76)	43.19(0.75)	131.11(1.92)	109.69(1.76)	7
$^{240}\text{Pu}(n, f)$	E – E	Symmetric	151.26(2.64)	51.00(0.42)	42.88(0.47)	131.48(1.23)	109.41(1.27)	3
$^{242}\text{Pu}(n, f)$	O – E	Asymmetric	171.50(4.96)	52.51(0.61)	41.49(0.61)	136.62(1.41)	106.40(1.42)	32
$^{242}\text{Pu}(n, f)$	E – E	Asymmetric	170.04(2.99)	52.58(0.59)	41.42(0.60)	136.78(1.40)	106.21(1.42)	9
$^{242}\text{Pu}(n, f)$	O – E	Symmetric	151.41(1.38)	50.78(0.34)	43.22(0.34)	132.54(0.83)	110.48(1.00)	3
$^{242}\text{Pu}(n, f)$	E – E	Symmetric	151.62	50.72	43.28	131.95	111.05	1

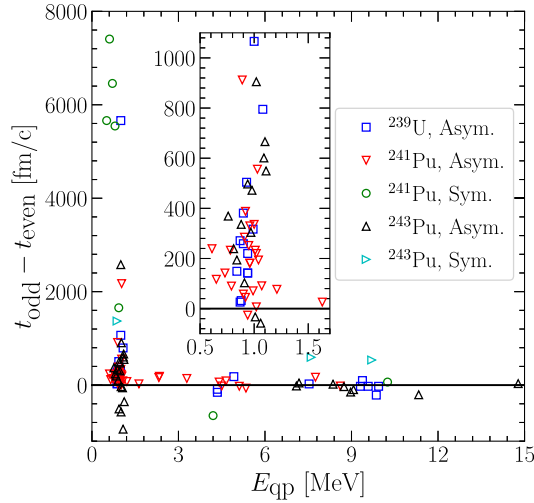


FIG. 2. Differences in evolution times between the odd-nucleon systems and the corresponding even-even “seed” nuclei. States with initial blocked nucleon(s) with $E_{qp} < 1$ MeV often show distinctly longer evolution times, in agreement with the trajectories illustrated in Fig. 3. See also the detailed tables in the Supplemental Material [18], which also includes a similar figure for ^{237}Np .

E-E nuclei ^{239}U , $^{241,243}\text{Pu}$, or ^{238}Np . Similar differences can also be observed for the average neutron and proton number distributions in the fission fragments. Thus, a correct treatment of the time-reversal symmetry breaking is essential to obtain the correct distribution width.

When considering ^{239}U , $^{241,243}\text{Pu}$, and ^{238}Np nuclei we have chosen a wide range of quasiparticle states in the seed nuclei, resulting in a wide distribution of the initial energy of the corresponding odd-nucleon nuclei. The largest

variations in the TKE is between initial states with either rather small or large initial octupole deformations $Q_{30}(0)$. These choices of $Q_{30}(0)$ lead to predominantly symmetric-like FF distributions in the case of small initial $Q_{30}(0)$ with smaller TKE, similar to what we have observed in the induced fission of even-even nuclei [12]. In particular, the trajectories originating from initial states with low $Q_{30}(0)$ often appear to be “confused,” as the system might wander around both positive and negative values of Q_{30} . If the initial value of $Q_{30}(0)$ [$Q_{30}(t) \approx \langle \Phi(t) | z(5z^2 - 3r^2) | \Phi(t) \rangle > 0.0$] is relatively large and positive, the heavy fission fragment typically ends up on the left [$Q_{30}(t_{\text{final}}) > 0$] of the fission Oz axis in the intrinsic reference frame of the fissioning compound nucleus. For low initial values of $Q_{30}(0)$ close to zero the system might take some time, before deciding that the HFF should go to the left or right (namely to $z < 0$ or $z > 0$, z being the fission axis), and that usually leads to longer saddle-to-scission evolution times, particularly in the case of relatively low quasiparticle energies; see Figs. 2 and 3 and the Supplemental Material [18]. In case of low-energy quasiparticle states chosen for the odd system, the fissioning nucleus has a relatively low intrinsic initial energy, and that also corresponds to a significantly large total level density of the nucleus with one or two odd nucleons when compared to an even-even system at comparable energies at and above the outer fission barrier. This leads to a significantly larger number of potential energy surfaces (PESs) per unit energy, and to an increased role of dissipative effects manifested by more frequent jumps between these more closely spaced PESs as the trajectory departs from the initial state. This behavior has been observed in Ref. [57], where the TDSLDA has been

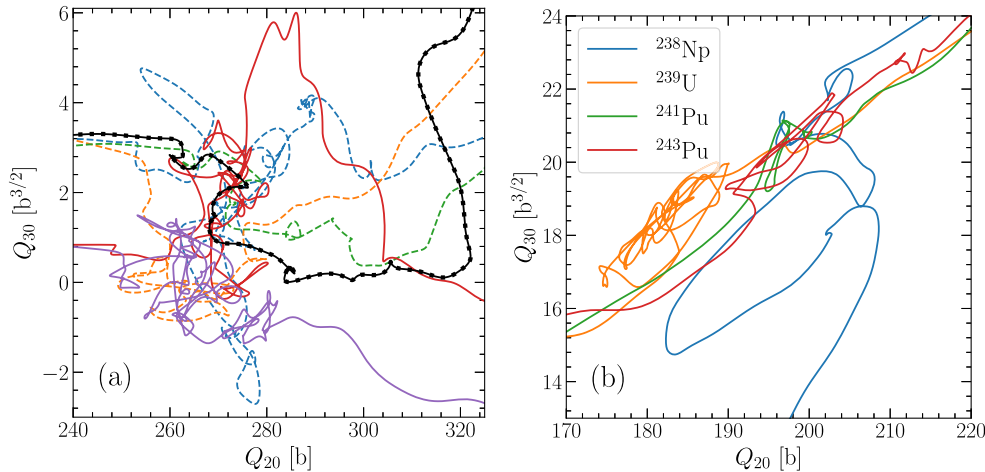


FIG. 3. Selected TDSLDA trajectories with long saddle-to-scission time from Fig. 1. (a) Data for ^{241}Pu only. These trajectories have low initial octupole deformations Q_{30} and, as a rule, low final TKE of the FFs. The trajectory for one “seed” E-E nucleus ^{241}Pu is shown with a black-dotted line. (b) Trajectories where the initial Q_{30} is not small. In all cases where the trajectory in Q_{20} and Q_{30} is “convoluted,” the saddle-to-scission time is significantly longer than for “smoother” trajectories with similar initial deformations. The time from scission until full FFs separation is always very short $\mathcal{O}(100)$ fm/c.

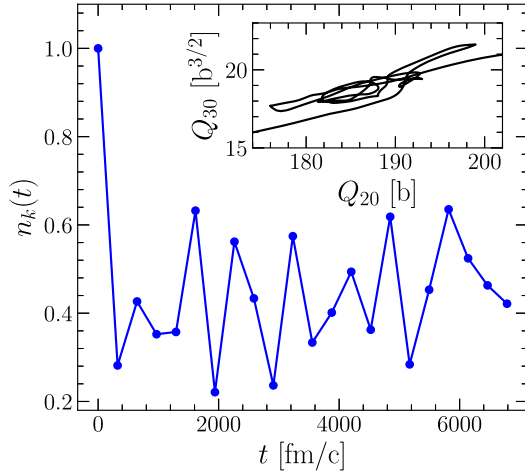


FIG. 4. The occupation probability $n_k(t) = \langle v_k(t) | v_k(t) \rangle$ for a chosen Pauli blocked quasiparticle state in ^{239}U . In this simulation we have used canonical quasiparticle wave functions initially; see Refs. [17,18]. Under time evolution, however, the quasiparticle wave functions cease to be canonical. The inset displays the shape evolution of the compound nucleus in the time interval 280–5000 fm/c.

augmented to include the otherwise absent stochastic elements. Otherwise, the evolution times from saddle-to-scission are very similar for both E-E and odd-nucleon compound nuclei, see Figs. 2 and 3 in the Supplemental Material [18].

A salient aspect of our results is illustrated in Fig. 4, demonstrating that the Pauli blocking approximation is strongly violated. Because of the emergence of pseudomagnetic effective fields, a nucleus with odd nucleons experiences repopulations of the quasiparticle states and the

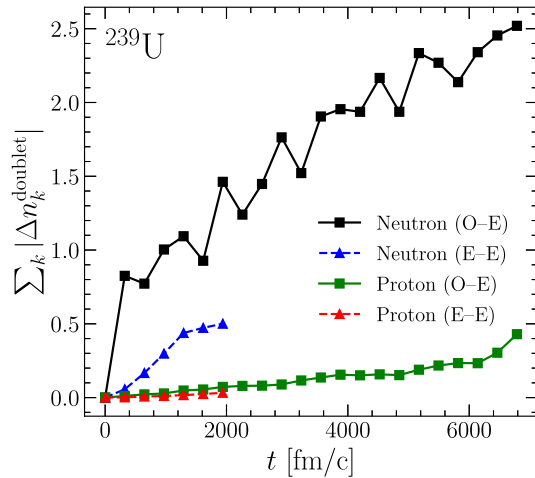


FIG. 5. The sum of the absolute difference of the nucleon occupation probabilities between “Kramers doublets” as a function of time for the system treated as O-E, O-O, and E-E, due to the role of time-odd fields. The run starts in the canonical basis and the neutron level with $n_k(0) \equiv 1$ is excluded.

trajectory is quite long, similar to those illustrated in Fig. 3. We have constructed the canonical states as described in Refs. [17,58] many-body wave functions at all times. Only one quasiparticle state at any time $t \neq 0$ has unit occupancy and is nondegenerate in the case of one odd neutron in ^{239}U , $^{242,243}\text{Pu}$ and for both nucleon types in ^{238}Np . The quasiparticle occupation probabilities emerging from static DFT calculations are different from the canonical occupation probabilities, and only the latter display the expected behavior of a Pauli blocked state; see Refs. [17,18,58] for details. Even if one chooses initial canonical wave functions, their canonical character is lost for all times $t > 0$. Only in static DFT calculations the generalized density matrix commutes with the generalized mean field and then for each quasiparticle state one can assign a quasiparticle energy E_k and a corresponding occupation probability $n_k = \langle v_k | v_k \rangle$. We illustrate in Fig. 5 the significantly enhanced role of the dynamically generated pseudomagnetic field(s), which break(s) time-reversal symmetry during the fission dynamics of nuclei with an odd-nucleon number, as compared to the case of an E-E nucleus, which is the real cause the Pauli blocking approximation is violated and why also the EFA is also expected to be inaccurate.

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Data availability—The data are not publicly available. The data are available from the authors upon reasonable request.

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