



Induced Fission of ^{240}Pu within a Real-Time Microscopic Framework

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We describe the fissioning dynamics of ^{240}Pu from a configuration in the proximity of the outer fission barrier to full scission and the formation of the fragments within an implementation of density functional theory extended to superfluid systems and real-time dynamics. The fission fragments emerge with properties similar to those determined experimentally, while the fission dynamics appears to be quite complex, with many excited shape and pairing modes. The evolution is found to be much slower than previously expected, and the ultimate role of the collective inertia is found to be negligible in this fully nonadiabatic treatment of nuclear dynamics, where all collective degrees of freedom (CDOF) are included (unlike adiabatic treatments with a small number of CDOF).

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Nuclear fission has almost reached the venerable age of 80 years [1,2], and it still lacks an understanding in terms of a fully quantum microscopic approach. This is in sharp contrast to the theory of superconductivity, another remarkable quantum many-body phenomenon, which required less than half a century from its discovery in 1911 [3] until the unraveling of its microscopic mechanism in 1957 [4]. Bohr [5–8] realized that the impinging low-energy neutrons on uranium targets leading to the nuclear fission proceed through the formation of a very complex quantum state, the compound nucleus, which has a very long lifetime. In a compound state the initial simple wave function of the impinging neutron is fragmented into a wave function of the nucleon + nucleus system with approximately one million components, as level density suggests [9]. In this respect this is similar to a particle in a box with a very small opening, consistent with the long lifetime of a compound nucleus state. Eventually, because of the interplay of the Coulomb repulsion between the protons and the nuclear surface tension, the nuclear shape evolves like a liquid charged drop and the compound nucleus reaches the scission configuration, leading predominantly to two emerging daughter nuclei. It was a great surprise when, in the 1960s, it was realized that the independent particle model proved to play a major role in the fission dynamics. At that time it became clear that the independent particle motion of nucleons and shell effects play a remarkable role and lead to a very complex structure of the fission barrier [10,11] and to a potential energy surface much more complicated than that suggested by a liquid drop model considered until then. On its way to the scission configuration a nucleus has to overcome not one, but two—the double-humped fission barrier—and sometimes even three potential barriers [10,11]. As in low-energy neutron induced fission, the excitation energy of the mother nucleus

is relatively small, the compound nucleus has a very slow shape evolution, and it was reasonable to assume that the shape evolution is either damped or overdamped. Since the presence of shape isomers has been unequivocally demonstrated, experimentally and theoretically, the dominant phenomenological approach to fission dynamics based on compound nucleus ideas, liquid drop, shell corrections, and the role of fluctuations described within Langevin and statistical approaches [12–21] has been born.

It became clear over the years that the fermion pairing and superfluidity play a critical role in nuclear fission, though in a vastly different manner than in the case of superconductivity [22,23]. Pairing correlations (either vibrations or rotations) are ubiquitous in nuclei [24], and they are expected to play a leading role in the nuclear shape dynamics [22,23,25,26]. The shape evolution of nuclei appears somewhat surprising at first sight since, typically, a nucleus is stiffer for small deformations and rather soft for large deformations. Hill and Wheeler [7] had the first insight into the origin of this aspect of nuclear large amplitude collective motion: the jumping from one diabatic potential energy surface to another and the role of Landau-Zener transitions. The most efficient microscopic mechanism for shape changes is related to the pairing interaction. The difficulty of making a nucleus fission in the absence of superfluidity was illustrated within an imaginary time-dependent Hartree-Fock approach treatment (an instanton, in quantum field theory parlance) of the fission of ^{32}S into two ^{16}O nuclei [27]. The initial and final states have an obvious axial symmetry, with occupied single-particle m -quantum states $\pm 1/2^5$, $\pm 3/2^2$, $\pm 5/2^1$ and $\pm 1/2^6$, $\pm 3/2^2$ for protons and neutrons, respectively, in the mother and daughter nuclei, where the superscript indicates the number of particles with the corresponding m -quantum number. In the absence of short-range pairing interactions, particularly

effective at connecting time-reversed nucleon pair states $(m, -m)$ with $(m', -m')$, and particularly the transition $(5/2, -5/2) \rightarrow (1/2, -1/2)$ in ^{32}S , fission is possible only if an axially broken symmetry intermediate state is allowed.

Since the late 1970s [26]—and particularly during the last decade—an alternative approach in the theoretical treatment of fission dynamics started gaining ground with the implementation of the philosophy of the density functional theory (DFT) [28–36] and its various modifications [37–49]. DFT is viewed as an alternative to solving the Schrödinger equation, in which the role of the many-body wave function is replaced by the one-body density matrix. DFT, however, does not provide a constructive recipe for determining the underlying functional. Application to nuclear physics requires a generalization of the most successful DFT implementation: the Kohn-Sham local density approximation (LDA) [29] to fermionic superfluid and time-dependent phenomena—the superfluid LDA (SLDA) and its time-dependent (TD) extension, a formalism based on local mean-field and pairing potentials. DFT is formulated by construction to appear as the Hartree-Fock or Hartree-Fock-Bogoliubov approximation (sometimes referred to—improperly, in our opinion—as HF or HFB), but it is, in principle, though not in practice, exact. With the use of quantum Monte Carlo results for cold atoms and phenomenological input for nuclear systems, (TD)SLDA has been validated against a wide range of experimental results [50–65].

The structure of the nuclear energy density functional (NEDF) is still largely based on phenomenology [66], and our approach here is based on the popular Skyrme parametrization SLy4 [67] and the SLDA treatment of the pairing correlations [53]. The numerical aspects of our approach have been described in great detail in Refs. [58,65,68,69] and the results presented below have been obtained by solving the TDSLDA equations for a ^{240}Pu nucleus in a simulation box $22.5^2 \times 40 \text{ fm}^3$, with a lattice constant corresponding to a relatively high momentum cutoff $p_c \approx 500 \text{ MeV}/c$, and with no spatial restrictions. The time step used was $0.119 \text{ fm}/c$ for up to 120 000 time steps, using ≈ 1760 graphics processing units (GPUS), for a total wall time of 550 min. The TDSLDA equations, which amounted to $\approx 56\,000$ complex coupled nonlinear TD 3D partial differential equations, were solved using a highly efficient parallelized GPU code [62,63,65] on Titan [70].

A ^{239}Pu nucleus bombarded with low-energy neutrons needs a very long time to evolve from its initial ground state shape until it reaches the outer fission barrier. In a constrained self-consistent calculation, we bring the nucleus to a shape and an energy in the immediate proximity of the outer fission barrier (at zero temperature). Starting from this configuration, we follow the nuclear dynamics within the TDSLDA approach until the two fragments are clearly separated; see Fig. 1. A summary of our results is presented

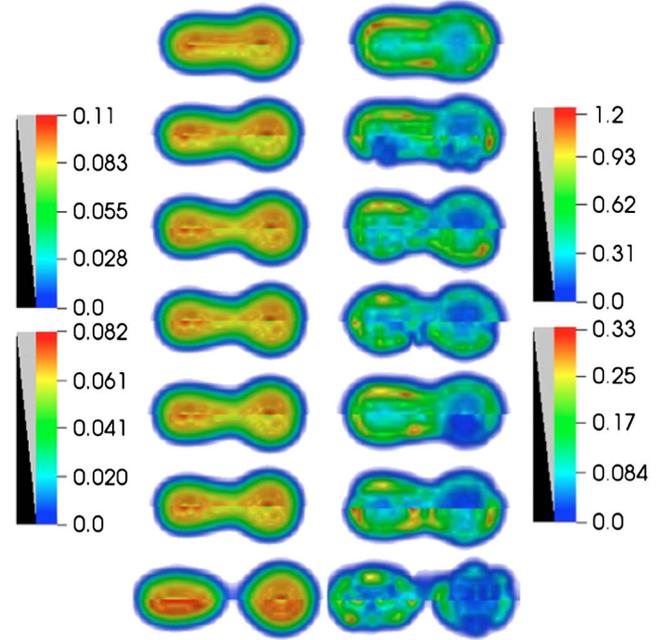


FIG. 1. The left column shows the neutron (proton) densities in the top (bottom) half of each frame. In the right column the pairing field for the neutron (proton) systems are displayed in the top (bottom) of each frame. The time difference between frames is $\Delta t = 1600 \text{ fm}/c$. The range of values is $(0, 0.1)$ and $(0, 0.07) \text{ fm}^{-3}$ for $\rho_{n,p}(\mathbf{r})$ and $(0, 0.9)$ and $(0, 0.7) \text{ MeV}$ for $\Delta_{n,p}(\mathbf{r})$, respectively, with color bars on the left (right) for densities (pairing gaps), and with upper (lower) ones for neutrons (protons). These frames are equally spaced in time for the case of the simulation S1; see Table I.

in Table I and is complemented by movies of the real-time simulations [76]. The main difference between the various simulations is in the character of the pairing correlations. Over the years several distinct parametrizations of the pairing coupling constant(s) have been suggested [77], basically various mixtures of the so-called volume and surface pairing, as compelling *ab initio* information is still lacking. The isospin symmetric density-dependent pairing coupling constant is $g_{\text{eff}}(\mathbf{r}) = g\{1 - \eta[\rho(\mathbf{r})/\rho_0]\}$, where $\rho(\mathbf{r})$ and ρ_0 are the total and saturation nuclear densities. The extensive phenomenological information gathered so far for ground states of nuclei fails to point to a well-defined value of the parameter η [53,77]. The dynamics, as we demonstrate, depends strongly and nonmonotonically on the parameter η . Fission dynamics requires a very efficient mechanism for the shape evolution, which is directly linked to transitions of the type $(m, -m) \rightarrow (m', -m')$, for which the pairing interaction is particularly effective [22,23,25]. The frozen occupation probabilities approximation [41,42] used in the past and a naive TDHF treatment [43] fail in this respect, as they do not allow the needed transitions, from levels with high m values in the mother nucleus to levels with low m values in the daughter nuclei, to take place [25], and a nucleus very often fails to fission or requires an inordinate amount of push

TABLE I. The simulation number, the pairing parameter η , the excitation energy (E^*) of $^{240}\text{Pu}_{146}$ and of the fission fragments [$E_{H,L}^* = E_{H,L}(t_{\text{SS}}) - E_{\text{gs}}(N_{H,L}, Z_{H,L})$], the equivalent neutron incident energy (E_n), the scaled initial mass moments $q_{20}(0)$ and $q_{30}(0)$, the “saddle-to-scission” time t_{SS} , TKE evaluated as in Ref. [71], TKE, atomic (A_L^{sys}), neutron (N_L^{sys}), and proton (Z_L^{sys}) extracted from data [72] using Wahl’s charge systematics [73] and the corresponding numbers obtained in simulations, and the number of postscission neutrons for the heavy and light fragments ($\nu_{H,L}$), estimated using a Hauser-Feshbach approach and experimental neutron separation energies [8,74,75]. Units are in MeV, fm², fm³, fm/c as appropriate.

S no.	η	E^*	E_n	q_{zz}	q_{zzz}	t_{SS}	TKE ^{sys}	TKE	A_L^{sys}	A_L	N_L^{sys}	N_L	Z_L^{sys}	Z_L	E_H^*	E_L^*	ν_H	ν_L
S1	0.75	8.05	1.52	1.78	-0.742	14 419	177.27	182	100.55	104.0	61.10	62.8	39.45	41.2	5.26	17.78	0	1.9
S2	0.5	7.91	1.38	1.78	-0.737	4360	177.32	183	100.56	106.3	60.78	64.0	39.78	42.3	9.94	11.57	1	1
S3	0	8.08	1.55	1.78	-0.737	14 010	177.26	180	100.55	105.5	60.69	63.6	39.81	41.9	3.35	29.73	0	2.9
S4	0	6.17	-0.36	2.05	-0.956	12 751	177.92	181		103.9		62.6		41.3	7.85	9.59	1	1

[37,40–42]. In some cases the axial symmetry beyond the outer barrier could be broken (see, e.g., Ref. [39]), and a suitable valley exists in the potential energy surface and fission can proceed. The approximate treatment of the pairing correlations within the TDBCS approximation [18,39] violates the continuity equation [78]. There is no question that a smooth transfer of the nuclear matter from the waist of the mother nucleus, which allows the nucleus to elongate and eventually to lead to the neck formation, is expected for any approach to fission dynamics. The TDSLDA is, so far, the only theoretical framework with a NEDF that satisfies all expected symmetries and theoretical constraints. At the same time, in SLDA solutions all symmetries can be broken, a situation similar to ferromagnets described by the Heisenberg Hamiltonian. In TDSLDA the evolution is, by default, smooth, and various contributions to the energy (often referred to in the literature as collective potential and kinetic energies) are always continuous as a function of the nuclear shape, unlike traditional approaches [46,49]. TDSLDA eschews the need to evaluate the inertia tensor, to introduce or guess the collective coordinates, or to invoke the adiabaticity of the evolution. The pairing field often reaches very small values, a situation also encountered in the study of the Higgs pairing mode [79,80] in other systems [57,81–84], when the pairing field can attain even exponentially small values for long periods of time, only to revive again. As in the case of density oscillations [64,65] (studied within the random phase amplitude limit), TDSLDA correctly describes the pairing vibrations in the case of a vanishing static pairing.

In all of the simulations performed by us so far, the heavy fragment has emerged basically spherical and with rather small excitation energy, while the light fragment is highly deformed and also has a higher excitation energy. Consequently, the excitation energy of the fragments does not follow from thermal equilibrium, as often has been assumed in the past in phenomenological studies (see the discussion in Refs. [85–87]), or as a Langevin approach (which implies thermal equilibrium throughout the entire system) might suggest. The heavy fragment has neutron and proton numbers very close to magic numbers and, naturally, a very weak pairing field as well. The large

deformation energy of the light fragment is eventually converted into a significant amount of internal excitation energy, which is released by neutron emission and gamma rays. The fact that the excitation energy of the heavy fragment is significantly smaller than the excitation energy of the light fragment correlates with the fact that the heavy fragment emerges as an almost magic nucleus with strong shell effects [85–87]. We did not observe any significant neutron emission at scission, a conclusion confirmed by the density profiles and the current flow we observe; see the Supplemental Material for movies [76]. The total kinetic energy (TKE) of the fission fragments is determined predominantly by the elongation of the fission system at scission [88]. In order to extract the TKE and the fragment excitation energies, we have assumed that, after scission, the internal excitation energies do not change. When compared to the existing evaluated experimental data [71] in the case of $^{239}\text{Pu}(n, f)$, the systematics, which follow the trend $\text{TKE} = 177.80 - 0.3489E_n \approx 177.3$ for S1–S3 (in MeV), we note that our estimated TKEs slightly overestimate the observed values by, at most, $\approx 3\%$; see Table I. This is indicative of the fact that, in our simulations, the system scissions a bit too early. The fission fragment mass and charge can be extracted from the data [72] (which have a resolution of about four to five mass units); see Table I. The evaluated average number of emitted neutrons [71] in this case is close to 3 (see Ref. [89]), which is higher than the values we estimate; see Table I. If the system would scission at a larger elongation, the light fragment would emerge with more excitation energy and the number of emitted neutrons would be larger.

Apart from the fact that a heavy nucleus fissions without any restrictions on the nuclear shape, TDSLDA supplies an additional big surprise. The time it takes a nucleus to descend from the saddle to the scission configuration is very long. A hydrodynamic approach [66] and the Langevin dynamics with various types of viscosities [12,26], along with approximate TD mean-field treatments lead to time scales of about 1000 fm/c or less. TDSLDA however, which incorporates naturally one-body dissipation, both wall and window mechanisms [90,91], points to time scales an order of magnitude larger than predicted in

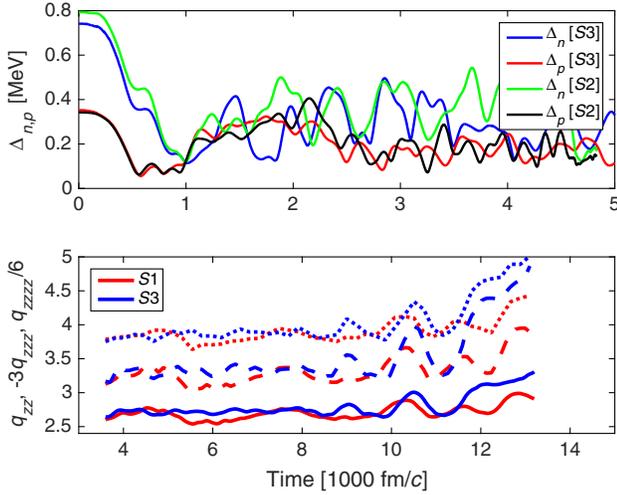


FIG. 2. The time dependence of spatially averaged $|\Delta_{n,p}(\mathbf{r}, \mathbf{t})|$ for S2 (mixed pairing) and S3 (volume pairing) in the upper panel and, in the lower panel, the scaled mass moments $q_{20}(t) = \int d^3z (3z^2 - r^2)/A^{5/3} \rho(\mathbf{r}, t)$, $q_{30}(t) = \int d^3z (5z^2 - 3r^2) \rho(\mathbf{r}, t)/A^2$, $q_{40}(t) = \int d^3z (35z^4 - 30z^2 r^2 + 3r^4) \rho(\mathbf{r}, t)/A^{7/3}$, with solid, dotted, and dashed lines, respectively, for S1 (red) and S3 (blue) [fm^L]; see Table I.

the literature. The nuclear system superficially behaves like an extremely viscous system, but the collective motion, at the same time, is not overdamped. There is a significant amount of collective flow, which is not dissipated and transformed into heat. The slide of the nucleus down from the saddle to the scission is not a monotonic one, but it is accompanied by a significant amount of collective shape and pairing field excitations in “transverse directions”; see Fig. 2. The long saddle-to-scission time t_{SS} can be attributed in part to the weak proton pairing gap in the starting configuration. In cases where the system starts initially with a relatively weak pairing proton gap, during the slide the proton pairing gap shows large temporal and spatial fluctuations [76]. In contradistinction to the TDHF + BCS approximation, spatial fluctuations are absent, the phase of the pairing field can be eliminated by a trivial gauge transformation, and fission does not happen without a boost from configurations near the outer saddle [39,40]. These large pairing gap fluctuations facilitate the shape evolution and the formation of the neck and the eventual scission of the nucleus. The two-body dissipation effects might affect these conclusions. A similar increase of the evolution time was demonstrated by Caldeira and Leggett [92,93], when coupling a simple quantum system with a “thermal bath”; see also Refs. [94,95] and the “bearing balls” video (the Drude model for electrons) [76]. Phenomenologically [14,15], the fission fragment distribution is reconstructed from (over-)damped dynamics and thus on very long time scales, which, superficially, is in agreement with a time averaging of our microscopic dynamics and with the apparent significantly

reduced role of collective inertia in the dynamics in a reduced collective space.

We have explored only axial symmetric configurations with broken left-right or parity symmetry ($q_{zzz} \neq 0$). Most authors agree that axial symmetry is hardly ever broken beyond the outer saddle. The system has spontaneously chosen such an initial deformation after we have imposed a slight pinch slightly off the middle of the mother nucleus. There is collective matter flow from one side to the other of the nucleus before scission, and the system dynamically determines its final fragment sizes; see the movie in the Supplemental Material [76]. This is indicative of the character of the potential energy surface, which shows softness in this collective variable, which was observed in previous studies [44–47]. The axial symmetry can be broken either spontaneously initially (not observed by us) or by quantum fluctuations (not studied here) during the evolution.

The quality of the agreement with experimental observations surprised us in its accuracy since we have made no effort to reproduce any measured data. We have merely used a rather randomly chosen NEDF, with rather decent properties, but far from perfect. However, since this NEDF encodes gross nuclear properties reasonably well, it does not come as a great surprise that gross properties of nuclear fission emerge so close to what one might have hoped for. Clearly, the details of the energy density functional at large deformations and the details of the pairing interaction will have to be pinpointed with greater accuracy. Induced nuclear fission presents us with a unique opportunity, in this respect, as in the study of the ground and weakly excited states, and even in the case of spontaneous fission [46], one can explore only rather small nuclear deformations. The nature of the dynamics of a fissioning nucleus appears to be quite surprising—the overall rolling down the hill is much slower than was ever expected—but not because of a particularly large viscosity. Rather, a large number of collective degrees of freedom (CDOF) are excited, both shape and pairing modes, which is clearly demonstrated by the real-time movies in the Supplemental Material [76]. The strong energy exchange between a large number of CDOF appears to be at the root of the slowness of this unexpected dynamics. There are experimental indications that fission times can be extremely long [96–98].

Even though, in this first study of its kind, we did not obtain perfect agreement with the experiment, our results clearly demonstrate that rather complex calculations of the real-time fission dynamics without any restrictions are feasible, and further improvements in the quality of the NEDF—and especially in its dynamic properties—can lead to a theoretical microscopic framework with great predictive power where experiments are not feasible, particularly in astrophysical environments. Extension of the present approach to two-body observables (fission fragment mass, charge, angular momenta, and excitation energy distribution widths) are rather straightforward to implement [99–101], and more

detailed information could eventually be inferred by introducing the stochasticity of the mean field [102,103].

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