Shape evolution and collective dynamics of quasifission in TDHF

A.S. Umar,1 V.E. Oberacker,1 and C. Simenel2

1Department of Physics and Astronomy, Vanderbilt University, Nashville, Tennessee 37235, USA
2Department of Nuclear Physics, RSPE, Australian National University, Canberra, ACT 0200, Australia

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I. INTRODUCTION

The creation of new elements is one of the most novel and challenging research areas of nuclear physics [1–4]. The search for a region of the nuclear chart that can sustain the so called superheavy elements (SHE) has led to intense experimental activity resulting in the discovery and confirmation of elements with atomic numbers as large as Z = 117 [5–7]. The theoretically predicted island of stability in the SHE region of the nuclear chart is the result of new proton and neutron shell-closures, whose location is not precisely known [8–10]. The experiments to discover these new elements are notoriously difficult, with fusion evaporation residue (ER) cross-sections to discover these new elements are notoriously difficult, with fusion evaporation residue (ER) cross-sections, whose location is not precisely known [8–10]. The experiments to discover these new elements are notoriously difficult, with fusion evaporation residue (ER) cross-sections, whose location is not precisely known [8–10]. The search for a region of the nuclear chart that can sustain the so called superheavy elements (SHE) has led to intense experimental activity resulting in the discovery and confirmation of elements with atomic numbers as large as Z = 117 [5–7]. The theoretically predicted island of stability in the SHE region of the nuclear chart is the result of new proton and neutron shell-closures, whose location is not precisely known [8–10]. The experiments to discover these new elements are notoriously difficult, with fusion evaporation residue (ER) cross-sections, whose location is not precisely known [8–10]. The search for a region of the nuclear chart that can sustain the so called superheavy elements (SHE) has led to intense experimental activity resulting in the discovery and confirmation of elements with atomic numbers as large as Z = 117 [5–7]. The theoretically predicted island of stability in the SHE region of the nuclear chart is the result of new proton and neutron shell-closures, whose location is not precisely known [8–10]. The experiments to discover these new elements are notoriously difficult, with fusion evaporation residue (ER) cross-sections, whose location is not precisely known [8–10]. The search for a region of the nuclear chart that can sustain the so called superheavy elements (SHE) has led to intense experimental activity resulting in the discovery and confirmation of elements with atomic numbers as large as Z = 117 [5–7].

The fusion process implies a transition from a dinuclear configuration – accompanied by particle exchange during the dynamical process – to a single-center compound-like configuration of the composite system. Most dynamical models [13–21] argue that, for heavy systems, a dinuclear complex is formed initially and the barrier structure and the excitation energy of this precompound system determine its survival to breaking up via quasi-fission. Furthermore, if the nucleus survives this initial state and evolves to a compound system, it can still fission due to its excitation.

Among the three stages towards ER given in Eq. (1), the determination of \( P_{CN} \) contains the most uncertainty which can be as much as 1–2 orders of magnitude [22, 23]. Experimentally, \( P_{CN} \) can be extracted from the measurement of fusion-evaporation residue cross-sections [24, 25]. However, these cross-sections become very small for heavy systems and the extraction of \( P_{CN} \) is uncertain for such systems. Information on \( P_{CN} \) can also be obtained by comparing the width of fragment mass distribution with the width expected in the case of pure fusion-fission [26, 27]. This approach, however, only provides an upper limit for \( P_{CN} \) as it assumes that only fusion-fission produces symmetric fragments [26].

An alternative approach involves the analysis of the angular distribution of symmetric fragments [23, 28, 29]. Although this approach has been widely used, it could have limitations due to the fact...
that quasifission in not a statistical decay but a dynamical process, and that fusion-fission could be asymmetric due to, e.g., late-chance fusion-fission at low excitation energies [30] as well as shell structure of pre-scission configurations [31]. Correlations between mass and scattering angle of the fragments have also been measured extensively [27, 32–35]. In particular, they can be used to disentangle fast quasi-fission processes (few zeptoseconds) to longer reaction mechanisms associated with contact times between the fragments exceeding 10 – 20 zs (i.e., long-time quasifission and fusion-fission). However, the analysis of such fragment mass-angle distributions [32, 33], as well as statistical descriptions of fragment angular distributions [28], require external parameters such as moment of inertia and temperature which are chosen somewhat arbitrarily [23, 28, 36]. It is therefore important to provide realistic evaluation of these parameters, in particular for the quasifission mechanism which requires a description of the complex nuclear dynamics.

Dynamical microscopic approaches are a standard tool to extract macroscopic properties in heavy-ion collisions [37–41]. In particular, the time-dependent Hartree-Fock theory [42], has been recognized for its realistic description of several low-energy nuclear reaction mechanisms [43, 44]. It has been recently utilized for studying the dynamics of quasifission [27, 35, 45, 46] and scission [47–49]. The study of quasifission is showing a great promise to provide insight based on very favorable comparisons with experimental data [27, 35]. As a dynamical microscopic theory, TDHF provides us with the complete shape evolution of the nuclear densities which can be used to compute the time evolution of deformation and inertia parameters. Using an extension to TDHF, the so-called density constraint TDHF (DC-TDHF) approach [37], it is also possible to compute the excitation energy in the fragments.

In this manuscript we focus our discussion on the extraction of the time evolution of the moment of inertia and of the excitation energy (temperature). These are indeed the relevant quantities for fragment mass-angle distributions and angular distribution analyses. These quantities are computed for collisions between calcium isotopes and actinides for which the TDHF approach has been shown to provide a deep insight into the quasifission reaction mechanisms [35, 45, 46].

II. FORMALISM

A. TDHF and DC-TDHF approaches

The theoretical formalism for the microscopic description of complex many-body quantum systems and the understanding of the nuclear interactions that result in self-bound, composite nuclei possessing the observed properties are the underlying challenges for studying low energy nuclear physics. The Hartree-Fock approximation and its time-dependent generalization, the time-dependent Hartree-Fock theory, have provided a possible means to study the diverse phenomena observed in low energy nuclear physics [43, 44]. In general modern TDHF calculations provide a useful foundation for a fully microscopic many-body description of large amplitude collective motion including collective surface vibrations and giant resonances [50–63] nuclear reactions in the vicinity of the Coulomb barrier, such as fusion [38, 41, 64–71], deep-inelastic reactions and transfer [21, 72–77], and dynamics of (quasi)fission fragments [35, 45–49].

Despite its successes, the TDHF approach has important limitations. In particular, it assumes that the many-body state remains a single Slater determinant at all time. It describes the time-evolution of an independent particle system in a single mean-field corresponding to the dominant reaction channel. As a result, it induces a classical behavior of many-body observables. A well known example is that TDHF does not include tunneling of the many-body wave-function and, thus, it is unable to describe sub-barrier fusion. It also underestimates width of fragment mass and charge distributions in strongly dissipative collisions [72, 74, 78]. Thus, to obtain multiple reaction channels or widths of one-body observables one must in principle go beyond TDHF [74, 79–82].

In fact, different reaction outcomes can also be obtained from mean-field calculations if the initial state (defined by the center of mass energy $E_{c.m.}$ and by the orbital angular momentum $L$ of the collision) is best described by a superposition of independent (quasi)particle states and that each of these states can be assumed to evolve in its own mean-field. This is the case, for instance, if a collision partner is deformed in its intrinsic frame. In this case, each orientation of the deformed nucleus may encounter a different mean-field evolution [83, 84]. Such orientation dependence of reaction mechanisms has been experimentally studied in quasifission with actinide targets [85–91] and confirmed in TDHF studies [27, 35, 44]. In particular, these studies have shown that collisions of a spherical projectile with the tip of prolately deformed actinides lead to fast quasifission (with contact time smaller than 10 zs) while collisions with the side of the actinide may induce longer contact times, larger mass transfer, and possible fusion. It is therefore common practice to investigate a subset of specific orientations depending on the reaction mechanism one is interested to investigate. For instance, it is sufficient to study “side collisions” in order to investigate the competition between quasifission and fusion in collisions with actinide nuclei [45].

In recent years it has become numerically feasible to perform TDHF calculations on a 3D Cartesian grid without any symmetry restrictions and with much more accurate numerical methods [44, 54, 75, 92–94]. In addition, the quality of effective interactions has been substantially improved [95–98]. In order to overcome the lack of quantum tunneling preventing direct studies of sub-barrier fusion, the DC-TDHF method was developed to compute heavy-ion potentials [37] and excitation energies [99] di-
rectly from TDHF time-evolution. For instance, this method was applied to calculate capture cross-sections for hot and cold fusion reactions leading to superheavy element $Z = 112$ [67].

### B. Fragment angular distributions

Experimental analysis of the fragment angular distributions $W(\theta)$ is commonly expressed in terms of a two-component expression for fusion-fission and quasifission parts [28, 100–102],

$$W(\theta) = \sum_{J=0}^{J_{CN}} F^{(FF)}(\theta) + \sum_{J=J_{CN}}^{J_{max}} F^{(QF)}(\theta),$$

where

$$F^{(\alpha)}(\theta) = \frac{(2J + 1)^2 \exp[-(J + 1/2)^2 \sin^2 \theta/4K^2_0(\alpha)] J_0[i(J + 1/2)^2 \sin^2 \theta/4K^2_0(\alpha)]}{\text{erf}[(J + 1/2)/(2K^2_0(\alpha))^{1/2}]}$$

and $\alpha \equiv FF$ (fusion-fission) or QF (quasi-fission). Here, $J_{CN}$ defines the boundary between fusion-fission and quasifission, assuming a sharp cutoff between the angular momentum distributions of each mechanism. The detailed definition of various mathematical functions can be found in Refs. [23, 29].

The quantum number $K$ is known to play an important role in fission [103]. The latter is defined as the projection of the total angular momentum along the deformation axis. In the Transition State Model (TSM) [103], the characteristics of the fission fragments are determined by the $K$ distribution at scission. The argument $K_0$ entering Eq. 3 is the width of this distribution which is assumed to be Gaussian. It obeys

$$K_0^2 = T\Xi_{eff}/h^2,$$

where the effective moment of inertia, $\Xi_{eff}$, is computed from the moments of inertia for rotations around the axis parallel and perpendicular to the principal deformation axis

$$\frac{1}{\Xi_{eff}} = \frac{1}{\Xi_\parallel} - \frac{1}{\Xi_\perp},$$

and $T$ is the nuclear temperature at the saddle point. The physical parameters of the fusion-fission part are relatively well known from the liquid-drop model [104, 105]. In contrast, the quasifission process never reaches statistical equilibrium. In principle, it has to be treated dynamically, while equation 3 is based on a statistical approximation. In addition, the usual choice for the nuclear moment of inertia for the quasifission component, $\Xi_0/\Xi_{eff} = 1.5$ [23, 28, 36], is somewhat arbitrary. Here, $\Xi_0$ is the moment of inertia of an equivalent spherical nucleus.

In order to illustrate the sensitivity of the fragment angular distribution with the chosen value of the moment of inertia, we have plotted the ratio $W(\theta)/W(90^\circ)$ in Figure 1 with $\Xi_0/\Xi_{eff} = 1.1$ and 1.5. Here, we used the value of $J_{CN} = 46$ obtained directly from TDHF calculations of quasifission for this system. Similarly, temperature was taken to be $T = 1.2$ MeV. The deviation is mostly visible at most forward and backward angles, where it can reach up to 20%. It is interesting to note that if we assume that the red curve of Fig. 1 is the actual measured quantity and try to fit it by varying the $J_{CN}$ value of the black curve we obtain a value of $J_{CN} = 37$. This would then be the error for using the wrong $J_{CN}$ value.

In the following section we outline the extraction of these ingredients directly from TDHF time-evolution of collisions resulting in quasifission.

### III. RESULTS

The feasibility of using TDHF for quasifission has only been recognized recently [35, 44, 45]. By virtue of long
contact-times for quasifission and the energy, orientation and impact parameter dependencies, these calculations require extremely long CPU times and numerical accuracy [92–94, 106]. During the collision process, the nuclear densities, as described by TDHF time-evolution, undergo complicated shape changes including vibration and rotation. Such evolutions finally lead to two separated final fragments identified as quasifission due to the long contact-time for the reaction as well as the mass/charge of the fragments [35, 45].

**FIG. 2.** (Color online) Quasi-fission in the reaction $^{48}$Ca+$^{249}$Bk at $E_{c.m.} = 218$ MeV with impact parameter $b = 2.0$ fm. Shown is a contour plot of the time evolution of the mass density. The actual numerical box is larger than the one shown in the frames.

In Fig. 2 we show a few time snapshots of the evolving mass density for the QF reaction of the $^{48}$Ca+$^{249}$Bk system at $E_{c.m.} = 218$ MeV with impact parameter $b = 2.0$ fm. The times are given in zeptoseconds ($1\,\text{zs} = 10^{-21}\,\text{s}$). The very large elongation of the separating fragments is noteworthy. The initial orientation of the deformed $^{249}$Bk was chosen such that the spherical $^{48}$Ca nucleus collides with the side of the actinide nucleus. In the following we will refer to this as the “side” orientation.

**A. Incoming and outgoing potentials**

As quasifission is part of the capture process, it is therefore possible to compute the nucleus-nucleus potential down to the inside barrier region with the DC-TDHF technique. The latter is represented in Fig. 3 for the incoming channel of a $^{40}$Ca+$^{238}$U central collision. After contact, the two fragments encounter a significant mass transfer toward symmetry. The outgoing potential is therefore different than the incoming one. In fact, the outgoing potential does not exhibit any barrier, leading to a re-separation of the fragments.

**FIG. 3.** (Color online) DC-TDHF nucleus-nucleus potential for the $^{40}$Ca+$^{238}$U central collision at $E_{c.m.} = 211$ MeV and with the side orientation. Both the potentials for the incoming and outgoing channels are shown.

**B. Moment of inertia**

The first collective observable of interest for fission and quasi-fission (both dynamical and statistical) studies is the moment of inertia of the system. The proper way to calculate the moment-of-inertia for such time-dependent densities (particularly for non-zero impact parameters) is to directly diagonalize the moment-of-inertia tensor represented by a $3 \times 3$ matrix with elements

$$\mathcal{I}_{ij}(t)/m = \int d^3r \, \rho(r,t)\left(r^2\delta_{ij} - x_i x_j\right),$$

where $\rho$ is the local number-density calculated from TDHF evolution, $m$ is the nucleon mass, and $x_i = 1, 2, 3$ denote the Cartesian coordinates. The TDHF calculations are done in three-dimensional Cartesian geometry [93]. Numerical diagonalization the matrix $\mathcal{I}$ gives three eigenvalues. One eigenvalue corresponds to the moment-of-inertia $\mathcal{I}_\parallel$ for the nuclear system rotating about the principal axis. The other two eigenvalues define the moments of inertia for rotations about axes perpendicular to the principal axis. Naturally, for triaxial density distributions, the two perpendicular components are not exactly equal but for practical calculations they are close enough and always larger than the parallel component. We thus use a single average value for these moments of inertia denoted by $\mathcal{I}_\perp$.

Using the time-dependent moment-of-inertia obtained from the TDHF collision one can calculate the so-called effective moment-of-inertia defined in Eq. (5). It is standard to compute the effective moment of inertia relative to a spherical system using the mass independent quantity $\mathcal{I}_0/\mathcal{I}_{eff}$, where $\mathcal{I}_0$ is the moment-of-inertia
of a spherical nucleus with the same number of nucleons [23, 29]. The expression for the moment-of-inertia for a rigid sphere is given by $\Theta_0/m = 2AR_0^2/\hbar^2$, where $R_0$ can be chosen as $R_0 = 1.225A^{1/3}\text{ fm}$ [29].

1. Role of impact parameter

The moment-of-inertia ratio calculated for the $^{48}\text{Ca} + ^{249}\text{Bk}$ non-central collisions at $E_{\text{c.m.}} = 218 \text{ MeV}$ is shown in Fig. 4. These trajectories all lead to quasi-fission with a large mass transfer and orbiting before the separation of the fragments. This is illustrated in Fig. 2 for the impact parameter $b = 2 \text{ fm}$ case. For this system using the value of $\hbar^2/m = 41.471 \text{ MeV-fm}^2$ and total mass number $A = 297$, we get $\Theta_0 = 191.359 \hbar^2\text{-MeV}^{-1}$. At the point of final touching configuration the moment-of-inertia ratios are in the range $1.4 - 1.8$, suggesting a relatively strong impact parameter dependence. Impact parameters smaller than $b = 0.5 \text{ fm}$ lead to contact time between the fragments exceeding 35 zs. We can therefore consider that the nuclei have fused and would ultimately form a compound nucleus.

2. Role of center of mass energy

Next we consider central collisions of the $^{40}\text{Ca} + ^{238}\text{U}$ systems. Similar to the $^{249}\text{Bk}$ case, $^{238}\text{U}$ exhibits a strong prolate deformaion and its alignment with respect to the collision axis changes the quasifission characteristics [35]. The value of $\Theta_0$ is $171.394 \hbar^2\text{-MeV}^{-1}$ for $A = 278$.

Figure 5 shows the time-evolution of the moment-of-inertia ratio at two different center of mass energies for central collisions with the side orientation. The two calculations have an energy difference of only 3 MeV in order to compare reactions which lead to similar mass asymmetry of the fragments. It is interesting to observe that up to $\sim 8 \text{ zs}$, the two evolutions of the moment of inertia are almost identical. This is likely to be due to the fact that the translational kinetic energy is rapidly dissipated in both cases, leading to a system with similar compactness but different excitation energies. For longer times, we observe a faster re-separation of the fragments at the highest center of mass energy. However, the final value of the ratio $\Theta_0/\Theta_{eff} \simeq 1.2$ is approximately the same for both energies. Note that this value is again different from the assumed ratio of 1.5, but in this case it is significantly smaller than in the $^{48}\text{Ca} + ^{249}\text{Bk}$ system, indicating that the $^{40}\text{Ca} + ^{238}\text{U}$ system leads to more compact configurations.

3. Role of the orientation

Figure 6 shows the dependence of the moment-of-inertia ratio on the orientation of the $^{238}\text{U}$ nucleus for the $^{48}\text{Ca} + ^{238}\text{U}$ system ($\Theta_0 = 179.693 \hbar^2\text{-MeV}^{-1}$ for $A = 286$). The shorter contact time for the tip orientation (elongation axis of $^{238}\text{U}$ parallel to the collision axis) is evident from the figure. We also observe that at the initial contact and for most of the evolution, the ratios are significantly different, originating from the very different initial density configurations. However, as the two reactions approach the scission point the ratios become relatively close to one another: The neck breaks at $\Theta_0/\Theta_{eff} \sim 1.1 - 1.2$. These values are, again, signifi-
significantly smaller than the assumed value of 1.5.

C. Collective dynamics and temperature

In this Section we study the evolution of the TDHF density relative to the dynamical potential energy surface (PES) using the DC-TDHF method. In the DC-TDHF method a parallel static calculation is performed at given time intervals which constrains the instantaneous TDHF density and finds the corresponding minimum energy state. This allows us to trace the TDHF dynamical trajectory on the PES, albeit restricted to the shapes calculated in TDHF evolution. At the same time these calculations provide the dynamical excitation energy, \( E^*(t) \) [99]. The DC-TDHF calculations for heavy-systems are extremely computationally extensive and the calculation of the trajectories performed here took about two months of computing on a 16 processor modern workstation utilizing all processors.

The temperature used in Eq. (4) is taken to be the temperature at the saddle point of a fissioning system [23, 36]. Since this is adopted from modeling of fission it may not be appropriate for quasifission, which does not really have a saddle point in the sense of ordinary fission and it is a fully dynamical process.

We have computed the dynamical temperature of the system during the quasifission path using the calculated excitation energy, \( E^*(t) \), as \( T(t) = \sqrt{E^*(t)/(A/8.5)} \) in MeV. Figure 7(a) shows the TDHF results for the \( ^{40}\text{Ca} + ^{238}\text{U} \) central collision at \( E_{\text{c.m.}} = 211 \text{ MeV} \) and side orientation. We see that the temperature rapidly rises during the initial overlap phase of the collision and reaches a value of about 1.0 MeV. After this the temperature rises more slowly being in the range of 1.25–1.5 MeV along the scission path.

While the calculation of the full temperature dynamics is very computationally expensive one can calculate the temperature if the dynamical density is stored at a chosen time. This would then allow us to obtain all the ingredients of the parameter \( K_0 \) of Eq. (4). One problem is that so far we do not have a convention for knowing exactly at what point one is supposed to evaluate the ingredients of \( K_0 \). The closest configuration to a fission saddle point would be the most compact shape formed just after full dissipation of the translational kinetic energy. In this case, this would correspond to the end of the first, fast rise of \( T^*(t) \), with a value of \( \sim 1 \) MeV.

IV. SUMMARY

The fully microscopic TDHF theory has shown itself to be rich in nuclear phenomena and continues to stimulate our understanding of nuclear dynamics. We have used the TDHF theory to study evolution of the nuclear density for reactions resulting in quasifission with a focus on the ingredients that are used to analyze experimental angular distributions to calculate the fusion probability \( P_{CN} \). We show that a number of useful quantities
can be obtained from TDHF dynamics rather than utilizing models that may not be appropriate for quasifission. Among these are the moment-of-inertia parallel and perpendicular to the symmetry axis, temperature through the calculation of the dynamical excitation energy, as well as other quantities like the rotational energy. In addition TDHF can tell us the dependence of these variables on impact parameter, energy, and structure.

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