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A pure quantum extension of the time-dependent semiclassical Boltzmann-Uehling-Uhlenbeck equation

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The Boltzmann equation is the traditional framework in which one extends the time-dependent mean field classical description of a many-body system to include the effect of particle-particle collisions in an approximate manner. A semiclassical extension of this approach to quantum many-body systems was suggested by Uehling and Uhlenbeck in 1933 for both Fermi and Bose statistics, and many further developments of this approach are known as the Boltzmann-Uehling-Uhlenbeck (BUU) equations. Here I introduce a pure quantum version of the BUU type of equations, which is mathematically equivalent to a generalized Time-Dependent Density Functional Theory extended to superfluid systems. As expected, during non-equilibrium processes the quantum Boltzmann one-body entropy increases during evolution.

The dynamics of a classical N-particle system can be described fully using the Liouville equation for the time-dependent probability distribution function $f_N(\boldsymbol{q}_1\ldots\boldsymbol{q}_N,\boldsymbol{p}_1\ldots\boldsymbol{p}_N,t)$, where $\boldsymbol{q}_k,\boldsymbol{p}_k$ are the canonical coordinates and momenta of the particles and $k=1\ldots N$. Integrating over $N-s$ coordinates and momenta one can introduce the s-particle time-dependent probability distributions $f_s(\boldsymbol{q}_1\ldots\boldsymbol{q}_s,\boldsymbol{p}_1\ldots\boldsymbol{p}_s,t)$ and derive the Bogoliubov-Born-Green-Kirkood-Yvon hierarchy of equations [1]. The lowest order approximation to the exact BBGKY hierarchy is the Vlasov equation for the one-particle time-dependent probability distribution function $f(\boldsymbol{q},\boldsymbol{p},t)$

$$\frac{\partial f}{\partial t}+\frac{\boldsymbol{p}}{m}\cdot\frac{\partial f}{\partial \boldsymbol{q}}+\boldsymbol{F}\cdot\frac{\partial f}{\partial \boldsymbol{p}}=0, \tag{1}$$

where m is the particle mass (assuming that all particles have the same mass) and $\boldsymbol{F}$ is the average force experienced by a particle from all the other particles

$$\boldsymbol{F}(\boldsymbol{q}_k)=-\sum_{l\neq k}^{N}\int d\boldsymbol{q}_l d\boldsymbol{p}_l f(\boldsymbol{q}_l,\boldsymbol{p}_l,t)\frac{V(|\boldsymbol{q}_k-\boldsymbol{q}_l|)}{\partial \boldsymbol{q}_k}, \tag{2}$$

assuming only two-particle interactions. One can show that in the semiclassical approximation the time-dependent Hartree-Fock equations reduce to the Vlasov equation Eq. (1). Boltzmann had the key insight to add an additional collision integral to this equation, assuming "molecular chaos" prior to the two particle collision, and thus arriving at a kinetic equation. Uehling and Uhlenbeck [2] generalized the Boltzmann equation by modifying the collision integral to take into account the quantum statistics, known as the Boltzmann-Uehling-Uhlenbeck (BUU) equation, and see also Bertsch and Das Gupta [3] for applications to nuclear physics,

$$\frac{\partial f}{\partial t}+\frac{\boldsymbol{p}}{m}\cdot\frac{\partial f}{\partial \boldsymbol{q}}+\boldsymbol{F}\cdot\frac{\partial f}{\partial \boldsymbol{p}}=I_{\rm coll}(\boldsymbol{p},t), \tag{3}$$

$$I_{\rm coll}(\boldsymbol{r},\boldsymbol{p},t)=-\frac{1}{(2\pi\hbar)^3}\int d\Omega\int d\boldsymbol{p}_2\int d\boldsymbol{p}_4\, v\frac{d\sigma(q,\Omega)}{d\Omega} \tag{4}$$

$$\times\{f(\boldsymbol{r},\boldsymbol{p},t)f(\boldsymbol{r},\boldsymbol{p}_2,t)[1+\theta f(\boldsymbol{r},\boldsymbol{p}_3,t)][1+\theta f(\boldsymbol{r},\boldsymbol{p}_4,t)]$$
$$-f(\boldsymbol{r},\boldsymbol{p}_3,t)f(\boldsymbol{r},\boldsymbol{p}_4,t)[1+\theta f(\boldsymbol{r},\boldsymbol{p},t)][1+\theta f(\boldsymbol{r},\boldsymbol{p}_2,t)]\},$$
$$\times\,\delta(\boldsymbol{p}+\boldsymbol{p}_2-\boldsymbol{p}_3-\boldsymbol{p}_4),$$
$$mv=q=|\boldsymbol{p}-\boldsymbol{p}_2|. \tag{5}$$

Here $\theta=\pm1$ for bosons/fermions respectively and $\theta\equiv0$ in the original Boltzmann equation. $\frac{d\sigma(q,\Omega)}{d\Omega}$ is the differential cross section of particles with initial/final $\boldsymbol{p},\boldsymbol{p}_2$ and final/initial momenta $\boldsymbol{p}_{3,4}$ into a solid angle $d\Omega$. The integrals of the first and the second terms in the curly brackets in Eq. (4) are often referred as the loss and gain terms in this kinetic equation.

The numerical solution of the BUU equation is significantly simpler than the solution of the time-dependent Hartree-Fock (TDHF) equations. For example, for a nuclear system in a simulation box $L^3=50^3$ fm^3 and with a momentum cutoff of $p_{\rm cut}=600$ MeV/c there are $4L^3(2p_{\rm cut})^3/(2\pi\hbar)^3\approx4.56\times10^5$ quantum phase-space cells, while a TDHF solution a system of $N=500$ nucleons in the same volume 50^3 fm^3 and with a spatial lattice constant of $l=1$ fm, which corresponds to the same momentum cutoff $p_{\rm cut}=\pi\hbar/l\approx600$ MeV/c, has a total of $2NL^3(2p_{\rm cut})^3/(2\pi\hbar)^3\approx1.1\times10^8$ quantum phase-space cells. However, since collisions are absent in TDHF framework, the role of equilibration processes are severely underestimated, even though TDHF describes more accurately the single-particle quantum dynamics and operates in a bigger space.

Similarly to the original Boltzmann equation, the BUU equation is valid only for a quantum dilute weakly interacting system in the semiclassical approximation. Therefore the particle-particle interaction has to be weak and short-ranged, and the average interparticle separation should be smaller than the interaction range. However,

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most of the quantum many-body systems of interest are dense, as the interaction range is of the order of the average interparticle separation or even larger, and the interaction strength is typically strong and in such situations the evaluation of the collision integral relies on various approximations and assumptions, and their accuracy and/or validity is almost impossible to evaluate. In a dense system the use of the free space cross section $\frac{d\sigma(q,\Omega)}{d\omega}$ is highly questionable, n-body collisions with $n > 2$ shall be taken into account, the assumption that a collision occurs at a well defined point in space $\boldsymbol{r}$ and absence of memory effects are inconsistent with the quantum uncertainty principle.

There were many attempts over the years to develop time-dependent descriptions of many-nucleon systems beyond the mean field, in order to describe missing two-body correlations, and in particular to allow for the equilibration of the single-particle degrees of freedom, while at the same time aiming towards a correct description of the quantum single-particle dynamics. The earliest attempts can be traced back to the generator coordinate method (GCM) and its time-dependent extension suggested by Wheeler and collaborators [4, 5], see a recent review [6]. One can try to introduce explicitly the two-body densities as well, see the recent review [7]. Other authors have suggested adding stochastic terms to the TDHF equations and I refer the interested reader to Ref. [8], where a number of such approaches are discussed. It suffice to say that these attempts have limited success in practice for many-fermion systems, apart from applications to rather idealized and simple cases.

I will present arguments that a generalization of the extension of the Time-Dependent Density Functional Theory (TDDFT) to superfluid systems is a generalized mean field framework, which can accommodate two body collisions. I will use the acronym gTDDFT for this further generalization, which will be still local, in the spirit of the Kohn-Sham approach [9] to the Density Functional Theory (DFT), often referred in literature as the local density approximation (LDA) or its further extensions [10]. The DFT is in principle mathematically equivalent with the many-body Schrödiger equation at the level of one-body density [9–13]. The difficulties with both these quantum many-body approaches are well known. The Schödinger equation requires the nucleon-nucleon interactions, which are not known exactly, and for systems of large many nucleons the numerical solution of this equation is practically impossible, unless various approximations are introduced. Within DFT one needs to know the energy density functional (EDF), which cannot be independently measured, its relation with the nucleon-nucleon interaction cannot be accurately established, and for time-dependent phenomena memory effects maybe important [12, 13]. The current difficulties of *ab initio* calculations and their relation with DFT approaches were recently discussed by Salvioni *et al.* [14].

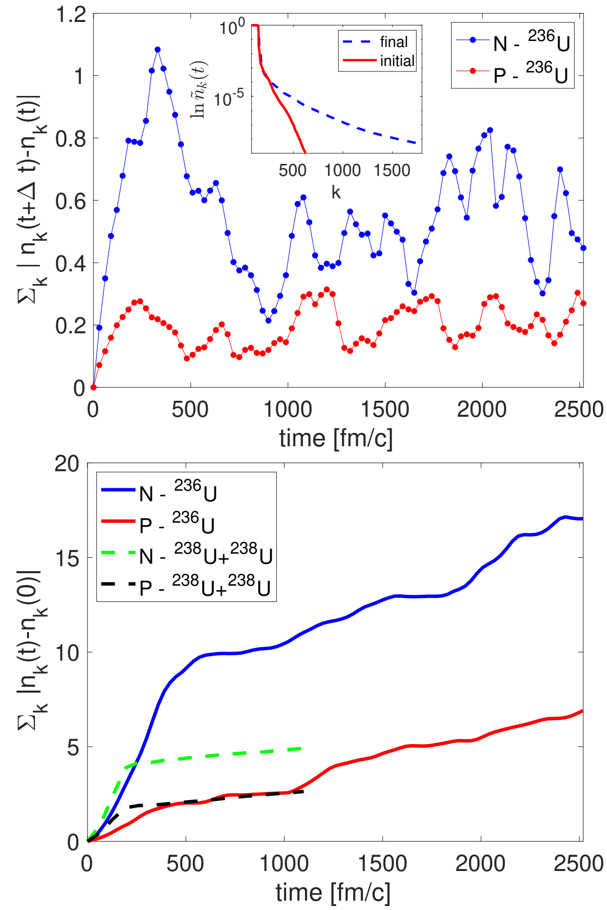

FIG. 1. Typical time evolutions of the nucleon occupation probabilities in a TDSLDA simulation of induced fission of ^{236}U started near the top of the outer fission barrier until complete fission fragment separation and for XY-collision (see Ref. [15] for convention) at zero impact parameter of ^{238}U+^{238}U with 1,500 MeV initial center of mass frame energy. The simulations were performed with the nuclear EDF SeaLL1 [16] with the LISE code [17]. Scission occurs at $t \approx 2,300$ fm/c. In collision the two final fragments are fully separated at $t > 1,000$ fm/c. In the upper panel displays the evolution of short-time evolution of the cumulative nucleon occupation probability $\sum_k |n_k(t+\Delta t) - n_k(t)|$ with $\Delta t \approx 30$ fm/c for fission and $\Delta t \approx 64$ fm/c for collisions. In the inset the canonical neutron occupation probabilities $\tilde{n}_k(t)$, ordered by size, are displayed at the start and finish of the simulation and one can clearly see the formation of the long momentum tails. The total change in the nucleon occupation probability $\sum_k |n_k(t) - n_k(0)|$ as a function of time is shown in the lower panel. Note that for any Δt in the absence of pairing $\sum_k |n_k(t+\Delta t) - n_k(t)| \equiv 0$. Here $n_k(t) = \sum_{\sigma=\uparrow,\downarrow} \int d\boldsymbol{r} |v_k(\boldsymbol{r},\tau,\sigma,t)|^2$, for either $\tau = n, p$, see Eq. (9).

The generalized TDDFT (gTDDFT), which is a further extension TDDFT to superfluid systems [18–22], which apart from allowing to describe static and time-dependent superfluid systems, has the side-effect of describing a particular class of two-body collisions. We often refer to the TDDFT extended to superfluid systems in the spirit of Kohn-Sham local density approximation DFT [9] as the time-dependent superfluid local density approximation (TDSLDA), which will become thus gTD-SLDA accordingly. As Bertsch initially suggested [23–26], while a nucleus adiabatically elongates during fission the single-particle energy levels display typically avoided crossings. The naive picture is that at such an avoided level crossing a Landau-Zener transition may occur. If a nucleon does not undergo a transition it will stay on the up-sloping level and a vacancy below the highest occupied level (the Fermi level) will be created by the down-sloping level. That means the nucleus will acquire an intrinsic excitation energy with a volume character, since the local Fermi surface will cease to be spherically symmetric. The dynamics of nuclei at relatively low energies is that of an incompressible quantum fluid, and its evolution is dominated by the surface tension and the shape of the electric charge distribution mostly [27, 28], with significant corrections due to shell-effects [29, 30]. After many such avoided level crossings the nucleus will acquire a volume excitation energy in case of Landau-Zener transitions, an evolution unexpected for an incompressible fluid. That is the main reason why within a TDHF description of fission nuclei fail to reach scission [31–34] and the presence of the pairing correlations in TDSLDA proved to be the crucial lubricant [35–37], as expected for a long-time [23–26]. Pairing correlations provide the mechanism for the nucleus to follow the dynamics of an incompressible fluid, where the volume energy component does not dramatically change. The single-particle levels are typically characterized by Kramers degeneracies and when a nucleus approaches a level crossing two nucleons jump together as a "Cooper pair" and the nucleus remains "cold." Such a transition is also Bose enhanced in the presence of a pairing condensate [35–37]. Because of the presence of pairing correlations in both neutron and proton systems within TDSLDA nuclei can easily undergo fission, unlike in a TDHF framework, when the initial configuration is close to the outer fission barrier. The evolution mechanism championed by Bertsch [23] however implies the presence of neutron and proton pairing condensates. On the other hand, the overwhelming experimental evidence is that the fission dynamics is not an adiabatic process, which is at odds with the prevailing microscopic approaches, based on the assumption of adiabaticity of the large amplitude collective motion [6, 38–40]. The fission fragments emerge with a significant total excitation energy, which is up to 20% of the total $Q = M_{\rm ini}c^2 - M^{\rm H}c^2 - M^{\rm L}c^2$ of the reaction, where $M_{\rm ini}, M^{\rm H}$ and $M^{\rm L}$ are the masses of the initial fissioning nucleus in case of spontaneous fission and of the ground states of the prompt fission fragments, and c is the speed of light. If on the way from saddle-to-scission the emerging fission fragments become hot the presence of neutron and/or proton pairing condensates becomes highly questionable along with the mechanism suggested in Ref. [23].

The TDSLDA is formulated in terms of Bogoliubov quasi-particle wave functions (qpwfs). The evolution of nucleon qpwfs is governed by the equations:

$$i\hbar\frac{\partial}{\partial t}\begin{pmatrix}u_{k\uparrow}\\u_{k\downarrow}\\v_{k\uparrow}\\v_{k\downarrow}\end{pmatrix}=\begin{pmatrix}h_{\uparrow\uparrow} & h_{\uparrow\downarrow} & 0 & \Delta\\ h_{\downarrow\uparrow} & h_{\downarrow\downarrow} & -\Delta & 0\\ 0 & -\Delta^* & -h^*_{\uparrow\uparrow} & -h^*_{\uparrow\downarrow}\\ \Delta^* & 0 & -h^*_{\downarrow\uparrow} & -h^*_{\downarrow\downarrow}\end{pmatrix}\begin{pmatrix}u_{k\uparrow}\\u_{k\downarrow}\\v_{k\uparrow}\\v_{k\downarrow}\end{pmatrix}, \quad (6)$$

where I have suppressed the spatial $\boldsymbol{r}$ and time coordinate t, and k labels the qpwfs (including the isospin) $[u_{k\sigma}(\boldsymbol{r},t), v_{k\sigma}(\boldsymbol{r},t)]$, with $\sigma = \uparrow,\downarrow$ the z-projection of the nucleon spin. The single-particle (sp) Hamiltonian $h_{\sigma\sigma'}(\boldsymbol{r},t)$, and the pairing field $\Delta(\boldsymbol{r},t)$ are functionals of various neutron and proton densities, which are computed from the qpwfs [17, 41].

Typical evolution of the nucleon occupation probabilities in a TDSLDA are shown in Fig. 1, which is absent in any TDHF, where $\dot{n}_k(t) \equiv 0$. In case of fission the emerging fission fragments have an excitation energy of ≈ 20 MeV each. In the case of collision ^{238}U+^{238}U the final fragments have excitation energies of about 400 and 600 MeV respectively and the distance of closest approach is reached at ≈ 250 fm/c, leading to a heavy fragment with $Z \approx 123$ and $N \approx 198$. At these excitations energies the neutron and proton pairing "gaps" have also significant spatial variations, the long-range order [42] is absent, and the pairing "gaps" have also significantly decreased in magnitude and the "true" pairing condensates are therefore absent. On the other hand, the effect of these pairing "gaps" on the nucleon wave functions $v_{k\uparrow}v_{l\downarrow} \leftrightarrow u_{m\uparrow}u_{n\downarrow}$ is basically the quantum equivalent of the action of the collision term in Eq. (4) $f_1f_2 \leftrightarrow (1-f_3)(1-f_4)$. It is notable that the rate of the single-particle occupation probability redistribution shown in Fig. 1

$$\sum_k |\dot{n}_k(t)| \approx \text{const.} \quad \text{for} \quad t > t_0, \quad (7)$$

is fairly constant after some initial time, $t_0 \approx 350$ fm/c in case of fission and $t_0 \approx 200$ fm/c in case of heavy-ion collisions, even after the reaction fragments are spatially separated. This is expected, as the thermal equilibration is a slower process. While $\sum_k \dot{n}_k(t) \equiv 0$ is always satisfied, in the absence of pairing correlations an even stronger constraint is in effect, $\dot{n}_k(t) \equiv 0$ for all k's. During these initial transitory times $t < t_0$, nuclei start with well-defined nn- and pp–pairing condensates, when the rates of pair transitions are higher due to the Bose enhancement mechanism. Since in case of heavy-ion collisions the excitation energies are higher, the magnitudes of the remnant pairing fields are smaller than in the case of fission. In the case of ^{236}U fission one can demonstrate that the quantum Boltzmann one-body entropy,

$$S(t) = -\sum_k [\tilde{n}_k(t)\ln\tilde{n}_k(t) + (1-\tilde{n}_k(t))\ln(1-\tilde{n}_k(t))], \quad (8)$$

changes from $S(t_{\rm ini}) = 12.4$ to $S(t_{\rm fin}) = 23.0$, and thus entropy increases as expected in a non-equilibrium evolution, where $\tilde{n}_k(t)$ are canonical occupation probabilities. Performing neutron and proton particle projection of the fissioning nucleus at the initial and final times, as described in Ref. [43], leads to $S(t_{\rm ini}) = 10.1$ to $S(t_{\rm fin}) = 18.8$ for ^{236}U and $S(t_{\rm ini}) = 10.2$ to $S(t_{\rm fin}) = 19.9$ for ^{238}Pu.

A simple qualitative argument, assuming that paring condensates are present, was presented in Refs. [23–26]. During the fissioning of an axially symmetric fissioning nucleus in a TDHF framework the projection of the single-particle angular momentum are conserved. In the initial nucleus the maximum nucleon orbital angular momentum is $l_z \approx k_F r_0 A^{1/3}$, which is noticeably larger than the maximum orbital angular momentum in a fission fragment $l_z \approx k_F r_0 (A/2)^{1/3}$. Here $k_F \approx 1.35$ fm^{-1} is the Fermi wave vector and $r_0 = 1.2$ fm. Within TDHF the single-particle occupation probabilities are conserved and in the absence of an effective mechanism for redistribution of the single-particle occupation probabilities the waist of the fission fragments are artificially kept large as in the initial nucleus, instead of shrinking by $\approx 2^{-1/3} \approx 0.79$. In an axially symmetric nucleus two nucleons with conjugate momenta can easily jump simultaneously if a transition $(m, -m) \to (m', -m')$ is allowed. Such a transition is controlled by a two-body matrix element $\langle m, -m|V|m', -m'\rangle$, which describes a $nn-$ or pp-collision with the pair quantum numbers $L = S = 0, T_z = \pm 1$. Therefore, as in the case of Boltzmann equation, the pairing correlations allow for $nn-$ and pp-collisions, but only with $L = S = 0, T_z = \pm 1$. However, unlike the Boltzmann equation, the TDSLDA also allows for the Bose enhancement of such transitions.

The absence of np-pair jumps is a major difference with the role played by the collision integral in the BUU equation. In heavy nuclei the number of np-pairs is larger than the sum of the numbers of nn- and pp-pairs and it is hard to accept that their role could be neglected in fission for example, particularly in the absence of genuine nn- and pp-pairing condensates. I will show here how one can generalize the TDSLDA to include np-collisions with pair quantum numbers $L = 0, S = 0, 1$. It is important to appreciate the fact that even if the long-range order of the pairing field/condensate is lost, these two-nucleon transitions survive at large excitation energies of the fissioning nucleus and in the fission fragments, which emerge with an excitation energy ≈ 20 MeV, corresponding to intrinsic temperatures ≈ 1 MeV or higher, as illustrated in Fig. 1. At these excitation energies both neutron and proton "pairing" fields have no phase coherence anymore, which means that the nucleons in the "Cooper pairs" have finite center-of-mass momenta, which varies from point-to-point inside the nucleus, and the pairing fields have large spatial variation of their magnitudes [35–37]. In spite of that, the rate of the redistribution of the nucleon occupation probabilities does not diminish for $t > t_0$, see Fig. 1. The addition of np-pairing short-range correlations is going to play a significant role in definition of the mass and charges fission yields, similarly in heavy-ion collisions. In nuclear and cold atoms physics pairing is attributed to an attractive short-range interaction, which as a result leads to very long momentum tails of the nucleon occupation probabilities $n(k) \propto 1/k^4$, and which at the same time are always present due to the presence of short-range correlations [44, 45] and have been recently unequivocally been put in evidence in experiments [46], particularly in the case np-pairs, which, as I advocate here, are likely the most important ones in dynamics.

I introduce generalized Bogoliubov quasiparticle $u-$ and $v-$components and corresponding generalized fermionic quasiparticle creation and annihilation operators

$$u_k(x) = u_k(\boldsymbol{r}, \tau, \sigma, t), \quad v_k(x) = v_k(\boldsymbol{r}, \tau, \sigma, t), \tag{9}$$

$$\alpha_k^\dagger = \sumint dx [\mathrm{u}_k(x)\psi^\dagger(x) + \mathrm{v}_k(x)\psi(x)], \tag{10}$$

$$\alpha_k = \sumint dx [v_k^*(x)\psi^\dagger(x) + u_k^*(x)\psi(x)], \tag{11}$$

$$\{\alpha_k^\dagger, \alpha_l\} = \delta_{kl}, \quad \{\alpha_k, \alpha_l\} = 0, \tag{12}$$

$$\{\psi^\dagger(x), \psi(y)\} = \delta(x - y), \quad \{\psi(x), \psi(y)\} = 0, \tag{13}$$

where $\tau = n, p$ and $\sigma =\uparrow, \downarrow$ and $\sumint$ stands for integration of spatial coordinates and summation over spin and isospin degrees of freedom. These new quasiparticle operators do not necessarily have a well defined isospin quantum number, they mix the neutrons and protons in the same manner as the spin degrees of freedom where mixed in previous approaches. With these definitions of quasiparticle states and with the restriction that the relevant anomalous densities be local in space one has to introduce the following four different types in case when only $L = 0$ is allowed:

$$\kappa_\tau(\boldsymbol{r}) = \sum_k v_k^*(\boldsymbol{r}, \tau, \downarrow) u_k(\boldsymbol{r}, \tau, \uparrow),\ \tau = n, p \tag{14}$$

$$\kappa_0(\boldsymbol{r}) = \sum_k v_k^*(\boldsymbol{r}, n, \downarrow) u_k(\boldsymbol{r}, p, \uparrow), \tag{15}$$

$$\kappa_1(\boldsymbol{r}) = \sum_k v_k^*(\boldsymbol{r}, n, \uparrow) u_k(\boldsymbol{r}, p, \uparrow), \tag{16}$$

where $\alpha_k|\Phi\rangle = 0$. Here $\kappa_{n,p}(\boldsymbol{r})$ are the usual neutron and proton anomalous densities, while $\kappa_0(\boldsymbol{r})$ describes pn-pairs with $S_z = 0$ and $\kappa_1(\boldsymbol{r})$ describes pn-pairs with $S_z = \pm 1$. $\kappa_0(\boldsymbol{r})$ has exactly the same form as the anomalous density for the unitary Fermi gas, in which case p and n would refer to atoms in different hyperfine states, which sometimes could be different atom species. In $\kappa_1(\boldsymbol{r})$ the roles of spin and isospin are switched when compared with $\kappa_\tau(\boldsymbol{r})$. The normal densities have a similar spin-isospin structure

$$n_\tau(\boldsymbol{r}) = \sum_{k,\sigma} v_k^*(\boldsymbol{r}, \tau, \sigma) v_k(\boldsymbol{r}, \tau, \sigma), \tag{17}$$

$$n_{np}(\boldsymbol{r}) = \sum_{k,\sigma} v_k^*(\boldsymbol{r}, n, \sigma) v_k(\boldsymbol{r}, p, \sigma), \tag{18}$$

$$\boldsymbol{\sigma}_\tau(\boldsymbol{r}) = \sum_{k,\sigma,\sigma'} v_k^*(\boldsymbol{r}, \tau, \sigma) \boldsymbol{\sigma}_{\sigma,\sigma'} v_k(\boldsymbol{r}, \tau, \sigma'), \tag{19}$$

$$\boldsymbol{\sigma}_{np}(\boldsymbol{r}) = \sum_{k,\sigma,\sigma'} v_k^*(\boldsymbol{r}, n, \sigma) \boldsymbol{\sigma}_{\sigma,\sigma'} v_k(\boldsymbol{r}, p, \sigma'), \tag{20}$$

and where $\boldsymbol{\sigma}$ are Pauli matrices. Other type of densities (density gradients, currents, etc.) are also needed [47, 48]. The gTDSLDA equations read in this case

$$i\hbar\frac{\partial}{\partial t}\begin{pmatrix}\mathrm{u}_k(x,t)\\ \mathrm{v}_k(x,t)\end{pmatrix} = \begin{pmatrix}H & \Delta\\ \Delta^\dagger & -H^*\end{pmatrix}\begin{pmatrix}\mathrm{u}_k(x,t)\\ \mathrm{v}_k(x,t)\end{pmatrix}, \tag{21}$$

where $\mathrm{u}_k(x,t)$ and $\mathrm{v}_k(x,t)$ are 4-column vectors (9) and H and Δ are 4×4 matrix operators with the structure

$$H = \begin{pmatrix} h_{n\uparrow,n\uparrow}(\boldsymbol{r}) & h_{n\uparrow,n\downarrow}(\boldsymbol{r}) & h_{n\uparrow p\uparrow}(\boldsymbol{r}) & h_{n\uparrow,p\downarrow}(\boldsymbol{r})\\ h_{n\downarrow,n\uparrow}(\boldsymbol{r}) & h_{n\downarrow,n\downarrow}(\boldsymbol{r}) & h_{n\downarrow p\uparrow}(\boldsymbol{r}) & h_{n\downarrow,p\downarrow}(\boldsymbol{r})\\ h_{p\uparrow,n\uparrow}(\boldsymbol{r}) & h_{p\uparrow,n\downarrow}(\boldsymbol{r}) & h_{p\uparrow p\uparrow}(\boldsymbol{r}) & h_{p\uparrow,p\downarrow}(\boldsymbol{r})\\ h_{p\downarrow,n\uparrow}(\boldsymbol{r}) & h_{p\downarrow,n\downarrow}(\boldsymbol{r}) & h_{p\downarrow p\uparrow}(\boldsymbol{r}) & h_{p\downarrow,p\downarrow}(\boldsymbol{r}) \end{pmatrix} \tag{22}$$

and

$$\Delta = \begin{pmatrix} 0 & \Delta_n(\boldsymbol{r}) & \Delta_1(\boldsymbol{r}) & \Delta_0(\boldsymbol{r})\\ -\Delta_n(\boldsymbol{r}) & 0 & -\Delta_0(\boldsymbol{r}) & \Delta_1(\boldsymbol{r})\\ -\Delta_1(\boldsymbol{r}) & -\Delta_0(\boldsymbol{r}) & 0 & \Delta_p(\boldsymbol{r})\\ \Delta_0(\boldsymbol{r}) & -\Delta_1(\boldsymbol{r}) & -\Delta_p(\boldsymbol{r}) & 0 \end{pmatrix}. \tag{23}$$

I did not include the chemical potentials in Eq. (21), as their presence is not necessary in the time-dependent formulation. The equations (21) are derived via an EDF, which should satisfy all the usual required symmetries. In particular the number and anomalous mixed neutron-proton densities can enter in such an EDF only as combinations $|\kappa_0(\boldsymbol{r})|^2$, $|\kappa_1(\boldsymbol{r})|^2$, $|n_{np}(\boldsymbol{r})|^2$, and $|\boldsymbol{\sigma}_{np}(\boldsymbol{r})|^2$, in order to satisfy isospin invariance. One can then show that both average neutron and proton numbers are conserved separately. Moreover, the average number of neutrons and protons with either spin-up or spin-down is conserved as well, unless an external time-dependent time-odd one-body field is present. If one assumes isospin symmetry then the three anomalous densities $|\kappa_{n,p}(\mathbf{r})|^2$ and $|\kappa_0(\boldsymbol{r})|^2$ should appear in the EDF with the same coupling constant. The absence of a di-neutron bound state and existence of a deuteron suggests however that np-pairs with $S=1, T=0$ could be controlled by a stronger effective s-wave coupling constant than the pairing coupling constant for $S=0, T=1$ pairs [49, 50]. This np-interaction can be derived either by eliminating the tensor interaction using second order perturbation theory or an approach similar to in medium similarity renormalization group [51]. A conclusive experimental evidence of the presence of a genuine neutron-proton pairing condensate in nuclear ground states is absent, with perhaps the exception of $N=Z$ nuclei and remains a matter of debate [48–50, 52–54]. The extension of the present analysis to $L\geq 1$ pairs is straightforward, see Ref. [48].

Fission or heavy-ion collisions of superfluid nuclei are typically started from states with vanishing mixed normal and anomalous densities, which will remain so during the entire time-dependent evolution in the absence of np-mixing. The neutron-proton pairing correlations can lead to a significant redistribution of single-particle occupation probabilities, similar to the role played by the collision integral in BUU simulations (4). As a simple example one can consider the case of a nucleus where both nn- and pp-pairing correlations are absent and include only np-pairing/short-range correlations/collisions using the magic nucleus ^{100}Sn. In the TDHF+TDBCS approximation the time evolution equations have a canonical form by design [55], the occupation probabilities evolve according to

$$i\hbar\frac{dn_k}{dt} = \Delta_k\kappa_k^* - \Delta_k^*\kappa_k, \quad i\hbar\frac{d\kappa_k}{dt} = \Delta_k(1-2n_k) \tag{24}$$

where now one couples a neutron state k with spin-up with a proton state k with spin-up in case of $S=1$ for example, thus inter-changing the roles of the spin and isospin. These equations have exactly the same structure as in the case of either nn- or pp-pairing correlations, but with a different content of the pairing field, which now will describe the jumps of np-pairs. Similar to BUU equation, a condensate is not needed to facilitate mass and charge transport. If the system is susceptible to develop wide mass and charge distributions one can initially simply seed relatively small pairing fields $\Delta_{0,1}(\boldsymbol{r})$ as in Ref. [56], and with an initial excitation energy corresponding to a larger level density. The Boltzmann one-body entropy will grow with time from $S(t_{\text{ini}})=0$, driving the system towards the most probable outcomes, as expected in a non-equilibrium process. Another option is to treat the pairing fields as phenomenological inputs as in nuclear BUU simulations. Since the occupation redistribution mechanism described here is similar to that present in the BUU equation, there is likely no need to generate np-components of the mean field part of Eq.(22), which were never considered in the BUU equation as far as I know. The "true" mean field h_{np} components are never dominant and since they will lead only to uncorrelated one-particle jumps their role is negligible.

In conclusion, noticing that the TDSLDA describes transitions of $nn-$ and $pp-$pairs even in the absence of genuine pairing condensates I have presented an extension of the TDSLDA framework, here dubbed gTDSLDA to account for $nn-$, $pp-$, and $np-$collisions, in a manner similar to the semiclassical BUU equation. The collision intergal in BUU equation accounts of the loss and gain processes

$$f(x_1)f(x_2) \leftrightarrow [1-f(x_3)][1-f(x_4)]. \tag{25}$$

Exactly the same type of transitions are performed by the $nn-$, $pp-$, and $np-$pseudo-pairing fields where transitions of the type [23–26]

$$v_k(\mathbf{r},\sigma_1,\tau_1)v_l(\boldsymbol{r},\sigma_2,\tau_2) \leftrightarrow u_m(\boldsymbol{r},\sigma_3,\tau_3)u_n(\boldsymbol{r},\sigma_4,\tau_4) \tag{26}$$

are enabled. In BUU and gTDSLDA frameworks transitions occur at the same position in space. The $nn-$ and $pp-$pairs jumps have been shown to occur consistently in the past TDSLDA calculations [35–37, 57] and Fig. 1, both in the presence of genuine pairing condensates as well in their absence and they lead to an increase of the Boltzmann one-body entropy, see Eq. (8). gT-DDFT/gTDSLDA thus incorporates naturally both the long-range mean field effects and the short-range correlations between nucleons.

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[1] K. Huang, *Statistical Mechanics* (Wiley, New York, 1987).

[2] E. A. Uehling and G. E. Uhlenbeck, "Transport Phenomena in Einstein-Bose and Fermi-Dirac Gases. I," Phys. Rev. **43**, 552 (1933).

[3] G. F. Bertsch and S. Das Gupta, "A guide to microscopic models for intermediate energy heavy ion collisions," Phys. Rep. **160**, 189 (1988).

[4] D. L. Hill and J. A. Wheeler, "Nuclear Constitution and the Interpretation of Fission Phenomena," Phys. Rev. **89**, 1102 (1953).

[5] J. J. Griffin and J. A. Wheeler, "Collective Motions in Nuclei by the Method of Generator Coordinates," Phys. Rev. **108**, 311 (1957).

[6] M. Verriere and D. Regnier, "The Time-Dependent Generator Coordinate Method in Nuclear Physics," Frontiers in Physics **8**, 233 (2020).

[7] M. Toyama, "Applications of Time-Dependent Density Matrix Approach," Frontiers in Physics **8**, 67 (2020).

[8] A. Bulgac, S. Jin, and I. Stetcu, "Unitary evolution with fluctuations and dissipation," Phys. Rev. C **100**, 014615 (2019).

[9] W. Kohn and L. J. Sham, "Self-consistent equations including exchange and correlation effects," Phys. Rev. **140**, A1133 (1965).

[10] R. M. Dreizler and E. K. U. Gross, *Density Functional Theory: An Approach to the Quantum Many–Body Problem* (Springer-Verlag, Berlin, 1990).

[11] P. Hohenberg and W. Kohn, "Inhomogeneous Electron Gas," Phys. Rev. **136**, B864 (1964).

[12] M. A. L. Marques, C. A. Ullrich, F. Nogueira, A. Rubio, K. Burke, and E. K. U. Gross, eds., *Time-Dependent Density Functional Theory*, Lecture Notes in Physics, Vol. 706 (Springer-Verlag, Berlin, 2006).

[13] M. A. L. Marques, N. T. Maitra, F. M. S. Nogueira, E. K. U. Gross, and A. Rubio, eds., *Fundamentals of Time-Dependent Density Functional Theory*, Lecture Notes in Physics, Vol. 837 (Springer, Heidelberg, 2012).

[14] G. Salvioni, J. Dobaczewski, C. Barbieri, G. Carlsson, A. Idini, and A. Pastore, "Model nuclear energy density functionals derived from ab initio calculations," J. Phys. G: Nucl. Part. Phys. **47**, 085107 (2020).

[15] C. Golabek and C. Simenel, "Collision Dynamics of Two ^{238}U Atomic Nuclei," Phys. Rev. Lett. **103**, 042701 (2009).

[16] A. Bulgac, M. M. Forbes, S. Jin, R. N. Perez, and N. Schunck, "Minimal nuclear energy density functional," Phys. Rev. C **97**, 044313 (2018).

[17] S. Jin, K. J. Roche, I. Stetcu, A. Abdurrahman, and A. Bulgac, "The LISE package: Solvers for static and time-dependent superfluid local density approximation equations three dimensions," Compiut. Phus. Commun. **269**, 108130 (2021).

[18] A. Bulgac, "Local-density-functional theory for superfluid fermionic systems: The unitary gas," Phys. Rev. A **76**, 040502 (2007).

[19] A. Bulgac, "The Long Journey from Ab Initio Calculations to Density Functional Theory for Nuclear Large Amplitude Collective Motion," J. Phys. G: Nucl. Part. Phys. **37**, 064006 (2010).

[20] A. Bulgac, M. M. Forbes, and P. Magierski, "The unitary Fermi gas: From Monte Carlo to density functionals," in *The BCS–BEC Crossover and the Unitary Fermi Gas*, Lecture Notes in Physics, Vol. 836, edited by W. Zwerger (Springer, Berlin Heidelberg, 2012) Chap. 9, pp. 127 – 191.

[21] A. Bulgac, "Time-Dependent Density Functional Theory and the Real-Time Dynamics of Fermi Superfluids," Ann. Rev. Nucl. and Part. Sci. **63**, 97 (2013).

[22] A. Bulgac, "Time-Dependent Density Functional Theory for Fermionic Superfluids: from Cold Atomic gases, to Nuclei and Neutron Star Crust," Physica Status Solidi B **256**, 1800592 (2019).

[23] G. Bertsch, "The nuclear density of states in the space of nuclear shapes," Phys. Lett. B **95**, 157 (1980).

[24] F. Barranco, G.F. Bertsch, R.A. Broglia, and E. Vigezzi, "Large-amplitude motion in superfluid Fermi droplets," Nucl. Data Sheets Phys. A **512**, 253 (1990).

[25] G. F. Bertsch and A. Bulgac, "Comment on "Spontaneous Fission: A Kinetic Approach"," Phys. Rev. Lett. **79**, 3539 (1997).

[26] G. F. Bertsch, "The shapes of nuclei," Int. J. Mod. Phys. **26**, 1740001 (2017).

[27] L. Meitner and O. R. Frisch, "Disintegration of Uranium by Neutrons: a New Type of Nuclear Reaction," Nature **143**, 239 (1939).

[28] N. Bohr and J. A. Wheeler, "The Mechanism of Nuclear Fission," Phys. Rev. **56**, 426 (1939).

[29] V. M. Strutinsky, "Shell effects in nuclear masses and deformation energies," Nucl. Phys. A **95**, 420 (1967).

[30] M. Brack, J. Damgaard, A. S. Jensen, H. C. Pauli, V. M. Strutinsky, and C. Y. Wong, "Funny Hills: The Shell-Correction Approach to Nuclear Shell Effects and Its Applications to the Fission Process," Rev. Mod. Phys. **44**, 320 (1972).

[31] P. Goddard, P. Stevenson, and A. Rios, "Fission dynamics within time-dependent Hartree-Fock: Deformation-induced fission," Phys. Rev. C **92**, 054610 (2015).

[32] P. Goddard, P. Stevenson, and A. Rios, "Fission dynamics within time-dependent Hartree-Fock. II. Boost-induced fission," Phys. Rev. C **93**, 014620 (2016).

[33] Y. Tanimura, D. Lacroix, and G. Scamps, "Collective aspects deduced from time-dependent microscopic mean-field with pairing: Application to the fission process," Phys. Rev. C **92**, 034601 (2015).

[34] G. Scamps, C. Simenel, and D. Lacroix, "Superfluid dynamics of ^{258}Fm fission," Phys. Rev. C **92**, 011602 (2015).

[35] A. Bulgac, P. Magierski, K. J. Roche, and I. Stetcu, "Induced Fission of ^{240}Pu within a Real-Time Microscopic Framework," Phys. Rev. Lett. **116**, 122504 (2016).

[36] A. Bulgac, S. Jin, K. J. Roche, N. Schunck, and I. Stetcu, "Fission dynamics of ^{240}Pu from saddle to scission and beyond," Phys. Rev. C **100**, 034615 (2019).

[37] A. Bulgac, S. Jin, and I. Stetcu, "Nuclear Fission Dynamics: Past, Present, Needs, and Future," Frontiers in Physics **8**, 63 (2020).

[38] P. Ring and P. Schuck, *The Nuclear Many-Body Problem*, 1st ed., Theoretical and Mathematical Physics Series No. 17 (Springer-Verlag, Berlin Heidelberg New York, 2004).

[39] N. Schunck and L. M. Robledo, "Microscopic theory of nuclear fission: a review," Rep. Prog. Phys. **79**, 116301 (2016).

[40] J. K. Krappe and K. Pomorski, *Theory of Nuclear Fission* (Springer Heidelberg, 2012).

[41] S. Jin, A. Bulgac, K. Roche, and G. Wlazłowski, "Coordinate-space solver for superfluid many-fermion systems with the shifted conjugate-orthogonal conjugate-gradient method," Phys. Rev. C **95**, 044302 (2017).

[42] C. N. Yang, "Concept of Off-Diagonal Long-Range Order and the Quantum Phases of Liquid He and of Superconductors," Rev. Mod. Phys. **34**, 694 (1962).

[43] A. Bulgac, "Restoring Broken Symmetries for Nuclei and Reaction Fragments," Phys. Rev. C **104**, 054601 (2021).

[44] R. Sartor and C. Mahaux, "Self-energy, momentum distribution, and effective masses of a dilute fermi gas," Phys. Rev. C **21**, 1546 (1980).

[45] S. Tan, "Large momentum part of a strongly correlated Fermi gas," Ann. Phys. **323**, 2971 (2008).

[46] O. Hen, M. Sargsian, L. B. Weinstein, E. Piasetzky, H. Hakobyan, D. W. Higinbotham, M. Braverman, W. K. Brooks, S. Gilad, K. P. Adhikari, J. Arrington, G. Asryan, H. Avakian, J. Ball, N. A. Baltzell, M. Battaglieri, A. Beck, S. May-Tal Beck, I. Bedlinskiy, W. Bertozzi, A. Biselli, V. D. Burkert, T. Cao, D. S. Carman, A. Celentano, S. Chandavar, L. Colaneri, P. L. Cole, V. Crede, A. D'Angelo, R. De Vita, A. Deur, C. Djalali, D. Doughty, M. Dugger, R. Dupre, H. Egiyan, A. El Alaoui, L. El Fassi, L. Elouadrhiri, G. Fedotov, S. Fegan, T. Forest, B. Garillon, M. Garcon, N. Gevorgyan, Y. Ghandilyan, G. P. Gilfoyle, F. X. Girod, J. T. Goetz, R. W. Gothe, K. A. Griffioen, M. Guidal, L. Guo, K. Hafidi, C. Hanretty, M. Hattawy, K. Hicks, M. Holtrop, C. E. Hyde, Y. Ilieva, D. G. Ireland, B. I. Ishkanov, E. L. Isupov, H. Jiang, H. S. Jo, K. Joo, D. Keller, M. Khandaker, A. Kim, W. Kim, F. J. Klein, S. Koirala, I. Korover, S. E. Kuhn, V. Kubarovsky, P. Lenisa, W. I. Levine, K. Livingston, M. Lowry, H. Y. Lu, I. J. D. MacGregor, N. Markov, M. Mayer, B. McKinnon, T. Mineeva, V. Mokeev, A. Movsisyan, C. Munoz Camacho, B. Mustapha, P. Nadel-Turonski, S. Niccolai, G. Niculescu, I. Niculescu, M. Osipenko, L. L. Pappalardo, R. Paremuzyan, K. Park, E. Pasyuk, W. Phelps, S. Pisano, O. Pogorelko, J. W. Price, S. Procureur, Y. Prok, D. Protopopescu, A. J. R. Puckett, D. Rimal, M. Ripani, B. G. Ritchie, A. Rizzo, G. Rosner, P. Roy, P. Rossi, F. Sabatié, D. Schott, R. A. Schumacher, Y. G. Sharabian, G. D. Smith, R. Shneor, D. Sokhan, S. S. Stepanyan, S. Stepanyan, P. Stoler, S. Strauch, V. Sytnik, M. Taiuti, S. Tkachenko, M. Ungaro, A. V. Vlassov, E. Voutier, N. K. Walford, X. Wei, M. H. Wood, S. A. Wood, N. Zachariou, L. Zana, Z. W. Zhao, X. Zheng, I. Zonta, and Jefferson Lab CLAS Collaboration, "Momentum sharing in imbalanced fermi systems," Science **346**, 614 (2014).

[47] M. Bender, P.-H. Heenen, and P.-G. Reinhard, "Self-consistent mean-field models for nuclear structure," Rev. Mod. Phys. **75**, 121 (2003).

[48] E. Perlińska, S. G. Rohoziński, J. Dobaczewski, and W. Nazarewicz, "Local density approximation for proton-neutron pairing correlations: Formalism," Phys. Rev. C **69**, 014316 (2004).

[49] G. F. Bertsch and Y. Luo, "Spin-triplet pairing in large nuclei," Phys. Rev. C **81**, 064320 (2010).

[50] A. Gezerlis, G. F. Bertsch, and Y. L. Luo, "Mixed-Spin Pairing Condensates in Heavy Nuclei," Phys. Rev. Lett. **106**, 252502 (2011).

[51] S. R. Stroberg, H. Hergert, S. K. Bogner, and J. D. Holt, "Non-Empirical Interactions for the Nuclear Shell Model: An Update," Ann. Rev. of Nucl. Part. Sci. **69**, 307 (2019).

[52] S. Frauendorf and A.O. Macchiavelli, "Overview of neutron–proton pairing," Prog. Part. Nucl. Phys. **78**, 24 (2014).

[53] A.M. Romero, J. Dobaczewski, and A. Pastore, "Symmetry restoration in the mean-field description of proton-neutron pairing," Phys. Lett. B **795**, 177 (2019).

[54] B. Cederwall, X. Liu, Ö. Aktas, A. Ertoprak, W. Zhang, C. Qi, E. Clément, G. de France, D. Ralet, A. Gadea, A. Goasduff, G. Jaworski, I. Kuti, B. M. Nyakó, J. Nyberg, M. Palacz, R. Wadsworth, J. J. Valiente-Dobón, H. Al-Azri, A. Ataç Nyberg, T. Bäck, G. de Angelis, M. Doncel, J. Dudouet, A. Gottardo, M. Jurado, J. Ljungvall, D. Mengoni, D. R. Napoli, C. M. Petrache, D. Sohler, J. Timár, D. Barrientos, P. Bednarczyk, G. Benzoni, B. Birkenbach, A. J. Boston, H. C. Boston, I. Burrows, L. Charles, M. Ciemala, F. C. L. Crespi, D. M. Cullen, P. Désesquelles, C. Domingo-Pardo, J. Eberth, N. Erduran, S. Ertürk, V. González, J. Goupil, H. Hess, T. Huyuk, A. Jungclaus, W. Korten, A. Lemasson, S. Leoni, A. Maj, R. Menegazzo, B. Million, R. M. Perez-Vidal, Zs. Podolyak, A. Pullia, F. Recchia, P. Reiter, F. Saillant, M. D. Salsac, E. Sanchis, J. Simpson, O. Stezowski, Ch. Theisen, and M. Zielińska, "Isospin properties of nuclear pair correlations from the level structure of the self-conjugate nucleus ^{88}Ru," Phys. Rev. Lett. **124**, 062501 (2020).

[55] G. Scamps, D. Lacroix, G. F. Bertsch, and K. Washiyama, "Pairing dynamics in particle transport," Phys. Rev. C **85**, 034328 (2012).

[56] A. Bulgac, I. Abdurrahman, and G. Wlazłowski, "Sensitivity to the intial conditions of the Time-Dependent Density Functional Theory (2021)," arXiv:2108.10858.

[57] I. Stetcu, A. Bulgac, P. Magierski, and K. J. Roche, "Isovector giant dipole resonance from the 3D time-dependent density functional theory for superfluid nuclei," Phys. Rev. C **84**, 051309 (R) (2011).