A time-dependent mean field framework beyond the typical mean field approach

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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-Uehling-Uhlenbeck (BUU) equations. Here I suggest 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.

The dynamics of a classical $N$-particle system can be described fully using the Liouville equation for the time-dependent probability distribution function $f_N(q_1 \ldots q_N, p_1 \ldots p_N, t)$, where $q_k, p_k$ are the canonical coordinates and momenta of the particles $k = 1 \ldots N$. Integrating over $N - s$ coordinates and momenta one can introduce the $s$-particle time-dependent probability distributions $f_s(q_1 \ldots q_s, p_1 \ldots p_s, t)$ and derive the Bogoliubov-Born-Green-Kirkwood-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 $f(q, p, t)$

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

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

\[
F(q_k) = -\sum_{l \neq k}^N \int dq_l dp_l f(q_l, p_l, t) \frac{V(|q_l - q_k|)}{\partial q_k}, \tag{2}
\]

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,

\[
\frac{\partial f}{\partial t} + \frac{p}{m} \cdot \frac{\partial f}{\partial q} + F \cdot \frac{\partial f}{\partial p} = I_{\text{coll}}(p, t), \tag{3}
\]

\[
I_{\text{coll}}(r, p, t) = \frac{1}{(2\pi \hbar)^3} \int d\Omega \int dp_2 \int dp_3 \frac{d^3q \Omega}{d\Omega} \times \left\{ f(r, p, t)f(r, p_2, t)[1 + \theta f(r, p_3, t)][1 + \theta f(r, p_4, t)] - f(r, p_3, t)f(r, p_4, t)[1 + \theta f(r, p, t)][1 + \theta f(r, p_2, t)]\right\},
\]

\[
\times \delta(p + p_2 - p_3 - p_4),
\]

\[
mv = q = |p - p_2|. \tag{5}
\]

Here $\theta = \pm 1$ for bosons/fermions respectively and $\theta \equiv 0$ in the original Boltzmann equation. $\frac{d^3q \Omega}{d\Omega}$ is the differential cross section of particles with initial final $p, p_2$ and final/initial momenta $p_{3,4}$ 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 $50^3$ fm$^3$ and with a momentum cutoff of 600 MeV/c there are $\approx 4 \times 10^6$ quantum phase-space cells, while a TDHF solution a system of 500 nucleons in the same volume $50^3$ fm$^3$ and with a spatial lattice constant of $l = 1$ fm, which corresponds to a the same momentum cutoff $p_{\text{cut}} = \pi \hbar/l \approx 600$ MeV/c, has a total of $27 \times 10^6$ quantum phase-space cells. Moreover, as 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, 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 situa-

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tions the evaluation of the collision integral relies on various approximations and assumptions, and their accuracy and/or validity is impossible to evaluate.

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 allow for thermalization of the intrinsic degrees of freedom, while at the same time having a correct description of the quantum single-particle dynamics. The earliest attempts can be traced back to the generator coordinate method (GCM) 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]. Others 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 none of these earlier attempts proved successful in the case of 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, similarly the Kohn-Sham approach to Density Functional Theory (DFT) [9]. The DFT is in principle mathematically equivalent with the many-body Schrödiger equation at the level of one-body density [10–13] and the formalism presented here is the quantum equivalent of the Boltzmann equation. The difficulties with both these quantum many-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. The present difficulties of ab initio calculations and their relation with DFT approaches were recently discussed by Salvioni et al. [14].

I describe here 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 additional 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 gTDSLDA accordingly. As Bertsch initially suggested [23–26], while a nucleus in particular it (slowly)

FIG. 1. Typical time evolutions of the neutron and proton occupation probabilities obtained during a TDSLDA simulation of induced fission of $^{238}$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] using the LISE code [17]. Scission occurs at $t \approx 2,300$ fm/c and 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 $\sum |n_k(t+\Delta t)-n_k(t)|$ over a relatively short time interval $T \approx 30$ fm/c for fission and $\Delta t \approx 64$ fm/c for collisions. The total change in the nucleon occupation probability $\sum |n_k(t)-n_k(0)|$ as a function of time is shown in the lower panel. Note that for $T \approx 10\Delta t$ in the absence of pairing $\sum |n_k(t+\Delta t)-n_k(t)| \equiv 0$. Here $n_k(t) = \sum_{\sigma=\uparrow,\downarrow} \int d\mathbf{r} |\psi_\mathbf{r}(\tau,\sigma, t)|^2$, for $\tau = n,p$, see Eq. (7).
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 crossing the nucleus will acquire a volume excitation energy, as opposed to an expected surface character and the role of the shape of the charge distribution. That is the main reason why within TDHF description of fission nuclei fail to reach scission [31–33] and the presence of the pairing correlations in TDDFT proved to be the crucial lubricant [34–36], as expected for a long-time [23–26]. Pairing correlations provide the mechanism for the nucleus to follow such a dynamics, 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 [34–36]. Because of the presence of pairing correlations in both neutron and proton systems within the extended TDDFT to superfluid systems nuclei can easily undergo fission, unlike in a TDHF framework, when the initial configuration is close to the outer fission barrier.

Typical evolution of the nucleon occupation probabilities in a TDHF extended to superfluid systems are shown in Fig. 1, which is absent in any TDHF or TDDFT simulation in the absence of pairing correlations. In case of fission the emerging fission fragments have an excitation energy of $\approx 20$ MeV each. In the case of collision $^{238}\text{U} + ^{238}\text{U}$ the final fragments have excitation energies of about 400 and 600 MeV respectively and the distance of closest approach is reached at $\approx 250$ fm/c, leading to a heavy fragment with $Z \approx 123$ and $N \approx 198$. At these excitation energies the neutron and proton pairing “gaps” have significant spatial variations, the long range order is absent, and the magnitude of the pairing “gaps” have also decreased and the “true” pairing condensates are absent. In this respect the action of these pairing “gaps” on the nucleon wave functions is basically the quantum equivalent of the action of the collision term in Eq. (4). It is notable that the rate single-particle occupation probability redistribution shown in the lower panel of Fig. 1

$$\sum_k \left| \dot{u}_k(t) \right| \approx \text{const.} \quad \text{for} \quad t > t_0, \quad (6)$$

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. (Remember that $\sum_k \dot{u}_k(t) = 0$ in this case and that in the absence of pairing correlations $\dot{u}_k(t) \equiv 0$ for all $k$’s.) This is expected, as the thermal equilibration is a slower process. During these initial transitory times 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.

A simple qualitative argument 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 nuclear 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 $\approx 2^{-1/3}$. In an axially symmetric nucleus two nucleons with conjugate momenta can easily jump simultaneously if a transition $(m, -m) \rightarrow (m', -m')$ 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 $L = 0, S = 0, T_z = \pm 1$. 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 extended TDDFT to superfluid systems I describe here also allows for the Bose enhancement of such transitions. I will show here how one can generalize the TDDFT extension to superfluid systems to include $np$-collisions with pair quantum numbers $L = 0, S = 0, 1$. The inclusion of higher partial waves $L \geq 1$ can be implemented by introducing new densities as in Skyrme-like mean field approaches [37]. It is important to appreciate the fact that even if the long-range order of the pairing field is lost, which is equivalent to the existence of a pairing condensate with Copper pairs at rest, these two-nucleon transition survive at large excitation energies of the fissioning nucleus and in the fission fragments, which emerge with and excitation energy $\approx 20$ MeV, corresponding to intrinsic temperatures $\approx 1$ MeV or higher. 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, and the pairing fields have large spatial variation of their magnitudes [34–36]. In spite of that, the rate of the redistribution of the nucleon occupation probabilities does not diminish, see Fig. 1. The addition of $np$-pairing correlations is going to play a significant role in definition of the mass and charges fission yields, similarly to what one observes in the case of the BUU equation in heavy-ion collisions.

I introduce generalized Bogoliubov quasiparticle $u$- and $v$-components and corresponding generalized fermionic quasiparticle creation and annihilation oper-
Thenormal densities have a similar isospin-spin structure, states, which sometimes could be different atom species.)

\[ S \] and proton anomalous densities, while \( \kappa \) and where \( p_n \) and \( \alpha \)

\[ \kappa_0(r) = \sum_k v_k^* (r, n, \downarrow) u_k (r, p, \uparrow), \]

\[ \kappa_1(r) = \sum_k v_k^* (r, n, \uparrow) u_k (r, p, \uparrow), \]

where \( \alpha_k \Phi = 0 \). Here \( \kappa_{n,p}(r) \) are the usual neutron and proton anomalous densities, while \( \kappa_0(r) \) describes \( pn \)-pairs with \( S_z = 0 \) and \( \kappa_1(r) \) describes \( pn \)-pairs with \( S_z = \pm 1 \). (NB \( \kappa_0(r) \) 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.) The normal densities have a similar isospin-spin structure

\[ n_\tau (r) = \sum_{k, \sigma} v_k^* (r, \tau, \sigma) u_k (r, \tau, \sigma), \]

\[ n_{np}(r) = \sum_{k, \sigma} v_k^* (r, n, \sigma) u_k (r, p, \sigma), \]

\[ \sigma_\tau (r) = \sum_{k, \sigma, \sigma'} v_k^* (r, \tau, \sigma) \sigma_{\sigma, \sigma'} v_k (r, \tau, \sigma'), \]

\[ \sigma_{np} (r) = \sum_{k, \sigma, \sigma'} v_k^* (r, n, \sigma) \sigma_{\sigma, \sigma'} v_k (r, p, \sigma'), \]

and where \( \sigma \) are Pauli matrices. Other type of densities (density gradients, currents, etc.) are also needed and they can be constructed in a straightforward manner [37]. In this gTDDFT formulation the presence of the mixed neutron-proton densities is new, they are not present in traditional time-dependent Hartree-Fock-Bogoliubov or in the TDDFT extended to superfluid systems.

The gTDDFT equations read in this case

\[ i \hbar \frac{\partial}{\partial t} \begin{pmatrix} u_k(x,t) \\ v_k(x,t) \end{pmatrix} = \begin{pmatrix} H & \Delta \\ \Delta^\dagger & -H^* \end{pmatrix} \begin{pmatrix} u_k(x,t) \\ v_k(x,t) \end{pmatrix}, \]

where \( u_k(x,t) \) and \( v_k(x,t) \) are 4-column vectors (7) and \( H \) and \( \Delta \) are \( 4 \times 4 \) matrix operators with the structure

\[ H = \begin{pmatrix} h_{n\uparrow,n\downarrow}(r) & h_{n\downarrow,n\uparrow}(r) & h_{n\uparrow,p\uparrow}(r) & h_{n\downarrow,p\downarrow}(r) \\ h_{p\uparrow,n\downarrow}(r) & h_{p\downarrow,n\uparrow}(r) & h_{p\uparrow,p\uparrow}(r) & h_{p\downarrow,p\downarrow}(r) \\ h_{p\downarrow,n\downarrow}(r) & h_{p\uparrow,n\uparrow}(r) & h_{p\uparrow,p\uparrow}(r) & h_{p\downarrow,p\downarrow}(r) \\ h_{p\uparrow,n\downarrow}(r) & h_{p\downarrow,n\uparrow}(r) & h_{p\uparrow,p\uparrow}(r) & h_{p\downarrow,p\downarrow}(r) \end{pmatrix}, \]

and

\[ \Delta = \begin{pmatrix} 0 & \Delta_n(r) & \Delta_1(r) & \Delta_0(r) \\ -\Delta_n(r) & 0 & -\Delta_0(r) & \Delta_1(r) \\ -\Delta_1(r) & -\Delta_0(r) & 0 & \Delta_p(r) \\ \Delta_0(r) & -\Delta_1(r) & -\Delta_p(r) & 0 \end{pmatrix}. \]

I did not include the chemical potentials in Eq. (19), as their presence is not necessary in the time-dependent formulation.

The equations (19) 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 \( |\kappa_0(r)|^2, |\kappa_1(r)|^2, |n_{np}(r)|^2, \) and \( |\sigma_{np}(r)|^2, \) in order to satisfy isospin invariance. One can then show then that both average proton and neutron numbers are conserved separately. Moreover, the average number of neutron 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 three three anomalous densities \( |\kappa_{n,p}(r)|^2 \) and \( |\kappa_0(r)|^2 \) should appear in the EDF with the same coupling constant. The absence of a di-neutron bound state and existence of a deuteron would suggest that \( pn \)-pairs with \( S = 1, T = 0 \) could be controlled by a stronger coupling constant then the pairing coupling constant for \( S = 0, T = 1 \) pairs. So far a conclusive experimental evidence of the presence of neutron-proton pairing condensate in nuclear ground states is absent, in spite of decades of theoretical and experimental effort to either prove its existence or detect it, with perhaps the exception of \( N = Z \) nuclei and still remains a matter of considerable debate, see Refs. [38–42] and references therein.

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 \( pn \)-mixing. The neutron-proton pairing correlations, even in the absence of a condensate, can lead to a significant redistribution of single-particle occupation probabilities, similar to the role played by the collision integral in BUU simulations (4). In fission and low energy heavy-ion collisions the transition of \( nn \)--, \( pp \)-- and \( np \)--nucleon pairs with \( L = 0 \) alone (21) can lead to wide mass and charge fragment yields distributions. The structure of the pairing field (21) is the same in the case of \( L = 2 \) nucleon pairs, only the corresponding anomalous densities will depend on derivatives of the quasi-particle wave functions. As in the case of BUU, 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 $\Delta_{0,1}(r)$ and with time they would grow. One option is to follow one of the prescriptions outlined in Ref. [43]. In this respect gTDDFT simulations would acquire a stochastic character. Another option is to treat the pairing fields as energy dependent phenomenological inputs as in the BUU simulations. Since the occupation redistribution mechanism described here is similar to one present in the BUU equation, there is likely no need to generate $np$-components of the mean field part of Eq. (20), which were never considered in the BUU equation as far as I know. The mean field $np$-components are never dominant and since they will lead only to uncorrelated one-particle jumps, not of pair-jumps, their role is expected to be negligible. Such components appear naturally if one would follow a Hartree-Fock procedure [38], but not within DFT framework, in which one typically postulates the form and density dependence of the EDF.

In conclusion, noticing that the TDDFT extension to superfluid systems (TDSLDA) describes transitions of $nn$ and $pp$ pairs even in the absence of true pairing condensates I have presented an extension of the TDDFT framework, here dubbed gTDDFT/gTDSLDA to account for $nn$, $pp$, and $np$ collisions, in a manner similar to the semiclassical BUU equation. I have limited the formal analysis to only $L = 0$ nucleon-nucleon pairs as the extension to $L \geq 1$ pairs is straightforward.

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