Density-Constrained TDDFT with Application to Fission

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Time-dependent DFT theory with density constraint
Fusion of light systems for astrophysics
Fission dynamics

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DC-TDHF for fusion
Fusion of neutron-rich systems
Capture for superheavy formations

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Quantitative Large Amplitude Shape Dynamics: Fusion and Fission INT-2013
Mean Field or Energy Density Functional (EDF)

\[ \langle \Psi | H | \Psi \rangle = E \]

\[ E = \langle \Phi | H_{\text{eff}} | \Phi \rangle = \int d^3r \left\{ H(\rho, \tau, j, s, T, J_{\mu \nu}; r) + H_{\text{Coulomb}}(\rho_p) \right\} \]

Single-(one-) particle density \textit{etc.} in terms of s.p. states

\[ \rho_q(\mathbf{r}) = \sum_{i=1}^{A} \sum_{\sigma} \phi_i^*(\mathbf{r}, \sigma, q) \phi_i(\mathbf{r}, \sigma, q) \]

EDF in NP more complicated

\[ v = v_{\text{NN-eff}} \rightarrow DFT(\text{Hartee – Fock}) \]

\[ v \neq v_{\text{NN-eff}} \rightarrow DFT(\text{Kohn – Sham}) \]

- **Structure**
  - Neutron Star Crust
  - Small and large amplitude collective phenomena

**Quantitative Large Amplitude Shape Dynamics:**

\[ \delta S = \int_{t_1}^{t_2} dt \langle \Phi(t) | H_{\text{eff}} - i \hbar \frac{\partial}{\partial t} | \Phi(t) \rangle = 0 \]

- Time-dependent generalization TDDFT (variational or Runge-Gross)

- Considerable effort testing TDDFT in recent years

\[ i \frac{\partial}{\partial t} \varphi_\alpha = \hbar (\rho, \tau, j, s, T, J_{\mu \nu}; r) \varphi_\alpha \]

**self-consistent**
\[ H_S(r) = \frac{\hbar^2}{2m} \tau + \frac{1}{2} \left[ t_0 \left( 1 + \frac{1}{2} x_0 \right) \rho^2 - \frac{1}{2} \left( \frac{1}{2} + x_0 \right) \left[ \rho^2_p + \rho^2_n \right] \right] + \frac{1}{4} \left[ t_1 \left( 1 + \frac{1}{2} x_1 \right) + t_2 \left( 1 + \frac{1}{2} x_2 \right) \right] \left( \rho \tau - j^2 \right) \]

\[-\frac{1}{4} \left( t_1 \left( \frac{1}{2} + x_1 \right) - t_2 \left( \frac{1}{2} + x_2 \right) \right) \left( \rho^2_p \tau_p + \rho^2_n \tau_n - j^2_p - j^2_n \right) - \frac{1}{16} \left[ 3t_1 \left( 1 + \frac{1}{2} x_1 \right) - t_2 \left( 1 + \frac{1}{2} x_2 \right) \right] \rho \nabla^2 \rho \]

\[ + \frac{1}{16} \left[ 3t_1 \left( \frac{1}{2} + x_1 \right) + t_2 \left( \frac{1}{2} + x_2 \right) \right] \left( \rho_p \nabla^2 \rho_p + \rho_n \nabla^2 \rho_n \right) \]

\[ + \frac{1}{12} t_3 \left[ \rho^{\alpha+2} \left( 1 + \frac{1}{2} x_3 \right) - \rho^\alpha \left( \rho^2_p + \rho^2_n \right) \left( x_3 + \frac{1}{2} \right) \right] \]

\[ + \frac{1}{4} t_0 x_0 s^2 - \frac{1}{4} t_0 (s^2_n + s^2_p) + \frac{1}{24} \rho^\alpha t_3 x_3 s^2 - \frac{1}{24} t_3 \rho^\alpha (s^2_n + s^2_p) \]

\[ + \frac{1}{32} \left( t_2 + 3t_1 \right) \sum_q s_q \cdot \nabla^2 s_q - \frac{1}{32} (t_2 x_2 - 3 t_1 x_1) s \cdot \nabla^2 s \]

\[ + \frac{1}{8} (t_1 x_1 + t_2 x_2) \left( s \cdot T - J^2 \right) + \frac{1}{8} (t_2 - t_1) \sum_q \left( s_q \cdot T_q - J^2_q \right) \]

\[ - \frac{t_4}{2} \sum_{qq'} \left( 1 + \delta_{qq'} \right) \left[ s_q \cdot \nabla \times j_{q'} + \rho_q \nabla_{\mu'} \cdot J_{\mu'} \right] \]

(s,j,T) time-odd, vanish for static HF calculations of even-even nuclei non-zero for dynamic calculations, odd mass nuclei, cranking etc.
Worldwide Nuclear TDDFT Efforts (partial list)

- Surrey – P. Stevenson
- Frankfurt/GSI – J. A. Maruhn
- Erlangen – P.-G. Reinhard
- Kyoto – Itagaki
- Tokyo – Iwata/Otsuka
- RIKEN/Tsukuba – Nakatsukasa
- Yabana....
- Frankfurt/GSI – J. A. Maruhn
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- RIKEN/Tsukuba – Nakatsukasa
- Yabana....
- Vanderbilt – Umar, Oberacker
- INT – Bulgac (TDDFT+TDHFB)
- GANIL -- Denis Lacroix
- ANU – C. Simenel

Existing programs

In progress
New Vanderbilt TDDFT Code

- 3-D Cartesian lattice – no geometrical simplification
- Complete EDF including all terms (time-even, time-odd, tensor)
- Coded in Fortran-95 and OpenMP
- Basis-Spline discretization for high accuracy

*Frankfurt/Vanderbilt/Surrey code will be submitted to Computer Physics Communications this year
Minimize energy with density constraint during unhindered TDDFT

\[ E_{DC}(t) = \min_{\rho} \left\{ E(\rho_n, \rho_p) + \int d^3r \nu_n(r)[\rho_n(r) - \rho_{n,tddft}(r, t)] + \int d^3r \nu_p(r)[\rho_p(r) - \rho_{p,tddft}(r, t)] \right\} \]

**Ion-Ion Potential**

\[ V(R(t)) = E_{DC}(t) - E_{A_1} - E_{A_2} \]

Subtract binding energies

- DC-TDHF finds underlying microscopic potential \( V(R) \)
- Parameter-free, only depends on chosen EDF
- Dynamical, energy-dependent
- Calculate \( E^*(t) \) and \( M(R) \)

Traditional method double-folding with frozen densities + CC

**Microscopic calculations of heavy-ion fusion reactions**

Nuclear fusion refers to the process in which two atomic nuclei combine to form a single larger nucleus. The two fundamental forces that dominate the probability of nuclear fusion are the electrostatic Coulomb force and the strong nuclear force. The Coulomb force is only long-range and attractive. On the other hand, the strong nuclear force acts between two nucleons, which are protons and neutrons. It is a short-range (about \( 1 \times 10^{-15} \) m) and a linear force. The fusion process is limited by the Coulomb repulsion between the two nuclei as they come closer. As they approach, the nuclei cool down, fuse, and stick together, forming a single nucleus.

Using the general formula, the energy density of the system includes the chemical potential energy of the nuclear matter. The fusion process begins when the nuclei come close enough to overcome the Coulomb barrier. The energy released in the fusion of light nuclei is much greater than the energy released in the fusion of heavy nuclei. This is because the mass excess of the light nuclei is much smaller than the mass excess of the heavy nuclei. The fusion of light nuclei releases a significant amount of energy, while the fusion of heavy nuclei releases only a small amount of energy.

**Approaches to calculating ion-ion potentials**

1. Microscopic models are the most general, but also the most computationally intensive. These models include the following:
   - **Fusion and Fission**
   - **Microscopic calculations**
   - **Dynamical, energy-dependent**
   - **Calculate \( E^*(t) \) and \( M(R) \)**

2. Traditional methods double-folding with frozen densities + CC.
Numerical Implementation

- Standard 1-body constraint $Q$ becomes

$$\lambda \hat{Q} \longrightarrow \int d^3r \, \lambda(r) \hat{\rho}(r) = \lambda(r)$$

1-body

- Iterative scheme for lambda is

$$\lambda^{n+1}(r) = \lambda^n(r) + c_0 \frac{\delta \rho^{n+1/2}}{2x_0 \rho^n(r) + d_0}$$

$$\delta \rho^{n+1/2}(r) \equiv \rho^{n+1/2}(r) - \rho_0(r)$$

- Full iteration becomes ($c_0$, $d_0$ parameters)

$$\chi^{n+1}_\chi = \mathcal{O}[\chi^{n+1/2}_\chi - x_0(\lambda^{n+1}(r) - \lambda^n(r) + \delta \lambda^n(r))\chi^{n+1/2}_\chi]$$

$$\delta \lambda^n(r) = c_0 \frac{\rho^n(r) - \rho_0(r)}{2x_0 \rho^n(r) + d_0}$$

Recent Applications of the Method (last three years)

**Neutron-rich systems – Superheavy formations**

- Microscopic study of the $^{132,124}$Sn+$^{96}$Zr reactions
- Dynamic microscopic study of pre-equilibrium giant resonance excitation and fusion in $^{132}$Sn+$^{48}$Ca and $^{124}$Sn+$^{40}$Ca
- Microscopic study of Ca+Ca fusion
- Microscopic analysis of sub-barrier fusion enhancement in $^{132}$Sn+$^{40}$Ca versus $^{132}$Sn+$^{48}$Ca
- *Entrance channel dynamics of hot and cold fusion reactions leading to superheavy elements*

**Light systems - astrophysics**

- *Microscopic Study of the Triple-$\alpha$ Reaction*
- *Linear-Chain Structure of Three-Alpha Clusters in $^{12}$C, $^{16}$C, and $^{20}$C*
- *Localization in light nuclei*
- *Microscopic composition of ion-ion interaction potentials*
- *Microscopic sub-barrier fusion calculations for the neutron star crust*

**Fission and miscellaneous**

- *Microscopic description of nuclear fission dynamics*
- *Single-particle dissipation in a time-dependent Hartree-Fock approach studied from a phase-space perspective*
- *Time-dependent coupled-cluster method for atomic nuclei*
Reactions Relevant for Neutron Star Crust

S-Factors for Reactions Relevant for Neutron Star Crust
Nuclear Fission

- Almost all of the theoretical work focuses on static fission barrier properties.

- Static-Adiabatic self-consistent calculation of barriers in terms of collective degrees of freedom.

- Improved by configuration mixing, projections, etc.

(HF+BCS, GCM+GOA+Cranking)

Pei, Nazarewicz, Sheikh, Kerman, Phys. Rev. Lett. 102, 192501 (2009)  
(Finite-temperature DFT or HFB)

Burvenich, Bender, Maruhn, Reinhard, PRC 69, 014307 (2004)  
(RMF + Skyrme HF systematics)

Bender, Heenen, Bonche, PRC 70, 054304 (2004)  
(HF+BCS, angular momentum projection)

(HFB with Gogny + GCM)
Types of Fission Dynamics

- Spontaneous or Prompt Fission

- Induced Fission (neutrons etc.)

- Quasi-Fission
  - Product of heavy-ion reactions
  - Important for superheavy formations

- Fusion-Fission

QF and FF may be amenable to study directly with TDDFT (see C. Simenel, EPJA 48, 152 (2012) )

We are currently studying QF in $^{48}\text{Ca} + ^{248}\text{Cm}$

Start from ground or excited state
Effective potential barrier description may be an oversimplification

In a many-body system different states see different barriers

Dynamical system may not follow the static PES path

Certain symmetry breakings are not included in adiabatic-static approach

How do we restore broken symmetries?
Understanding the *dynamics* of prompt and induced fission is a challenge

By dynamics we mean real-time microscopic dynamics – not in collective subspace

Is TDHF-TDDFT suitable to study some aspects of fission dynamics?

Periodic TDHF equations are too hard to solve for spontaneous fission

Multi-configuration or stochastic dynamics may be necessary
Historical attempts to use TDHF for fission dynamics

Our attempts to use TDHF for fission dynamics

Fusion-fission, quasi-fission studies using TDHF and DC-TDHF

One success – not yet explained!
Fission – TDHF - History

Most well known attempt to study fission via TDHF:
Dynamics of Induced Fission, Negele, Koonin, Möller, Nix, Sierk, PRC 17, 1098 (1978)

- Nucleus is initialized via quadrupole constraint with energy 1 MeV below and beyond the saddle point.

- Crude numerical methods; axial symmetry, reflection symmetry, no spin-orbit, BKN force.

- To break symmetries and couple angular momenta time-dependent BCS was introduced in conjunction TDHF calculations. Only reason?

- Results depend strongly on gap parameter.

Furthermore, there exists a conceptually clear program in which, in principle, the initial adiabatic TDHF wave function provides an ensemble of initial conditions from which all fission observables may be unambiguously calculated microscopically without any free parameters.
Fission – TDHF - History

Other fission studies using TDHF:

- Fission studied with TDHF for slabs by giving a collective boost to the initial HF state
  1. Fission was not seen when using small velocity field for boost but higher fields resulted in fission.
  2. Instead, the initial states were constructed by exciting single particle states into higher unoccupied states. Easier to induce fission from these configurations.

- Nucleus is initialized inside and just outside of the barrier and given a quadrupole boost
- Two different method used to create the initial HF states:
  1. A single center regular HF state – no fission achieved for different initializations.
  2. A spherical two-center initial HF state leads to fission almost always.

Jung, Cassing, Mosel, Cusson. NP A477, 256(1988)
- Studied multi-fragmentation and fission using TDHF (very adhoc).
  1. Initialize by multiplying density with some r-dependent function $c(r)$.
  2. Boost by $\exp(i\mathbf{k}\cdot\mathbf{r})$ but with $\mathbf{k}$ having different sign for different parts of the nucleus. Limiting $k > 0.5 \text{ fm}^{-1}$
In studying fusion with TDHF we sometimes see states that are coalesced and show kind of a resonance behavior.

For light nuclei these have been associated with shape-isomers and nuclear molecular resonances.

In heavy systems these intermediary states may:

bits a kind of resonance behavior. Therefore, it is tempting to speculate\textsuperscript{52} that these characteristics might be associated with single-particle wave functions which are approximate eigenstates\textsuperscript{53} of the instantaneous Hartree-Fock (HF) Hamiltonian $h(t_f)$ as $t_f \to \infty$. The many-body wave function constructed from these single-particle wave functions could then be considered a “transition state” to processes that are not taken into account in TDHF theory.\textsuperscript{52}

\textsuperscript{52}Private comm. A.K. Kerman

Initiating Fission with Boosts

Simple \( \exp(\imath \mathbf{k} \cdot \mathbf{r}) \) boost with \( \mathbf{k} \) having opposite sign for each half

- \( k_x \sim 0.4 \text{ fm}^{-1} \) limit for breakup
- about 700 MeV excitation!
- symmetric fission
- asymmetric boost makes the c.m. move, box problems

Boost using collective operators \( \exp(\imath \alpha q_{20} + \imath \beta q_{30}) \) etc.

- Similar to above case, need very large boosts
- Is there a magic boost operator?

Bertsch et al. PRC 17, 1646 (1978) find \( E/A > 2 \text{ MeV} \)

\( ^{238}\text{U g.s.} \)
Initializing with excited states

- Construct excited states by promoting s.p. levels
  - generate g.s. wavefunctions and store
  - read them into the Gram-Schmidt routine when running HF again
  - orthogonalize selected state(s) to all of the g.s. wavefunctions

\[
\langle \Phi | \Phi ' \rangle = \text{det} (\langle \varphi_i | \varphi'_j \rangle) = 0
\]

- For light nuclei very interesting breakups occur during the static iteration!
  \[12C^* \text{ promote } 1p \text{ to } 2s\]
- Harder for heavier systems – the effect not as pronounced
- Is there a way to select particular states to excite in induced fission?
Roll Down Approach

- Initialize the system close to the saddle point and let it evolve via TDHF.
- With no boost or reasonable collective boosts, the system does not fission.
- In all of these, the system has difficulty reorganizing to have a two-center configuration.
- Tried with and without pairing (frozen in TDHF).
Different approach – fission after a collision

- Recently, we have investigated fission after a low-energy collision
- We have studied collision of $^{100}\text{Zr} + ^{140}\text{Xe} \rightarrow ^{240}\text{Pu}^*$
- Long-time oscillatory behavior, followed up to 2600 fm/c ($E_{cm}=250$ MeV)
Unconstrained Fusion/Fission Isomer

- Start from TDHF fusion state and minimize energy by DC
- Starting from DC-TDHF result minimize energy with no constraint

\[ \beta_2 = 2.27 \quad Q_{20} = 230 \text{ b} \quad Q_{30} = -28 \text{ b}^{3/2} \]
Initiation of Fission

Boost this state by a unitary collective boost operator $e^{ipq_{20}(r)}$ where for $p = 0.0025$ we get 7.5 MeV excitation.

SHOW MOVIE!
Fission Path

Follow fission path with DC-TDHF

A$_1$, Z$_1$ = 106, 42
A$_2$, Z$_2$ = 134, 52
Further Experimentation and a Special Case

- Obtained the 238U initial state from Skyax with BCS-LN (SLy4)
  
  a) Do a density-constraint to reproduce the same density in 3D with no L-N
  
  b) Do a density-constraint to reproduce the same density in 3D with L-N
  
  c) Do a q2 constraint to reproduce the same density in 3D with L-N

  Energies (no c.m. correction):
  
  \[ E_{(a)} = -1761.2 \text{ MeV} \]
  \[ E_{(b)} = -1762.9 \text{ MeV} \]
  \[ E_{(c)} = -1763.4 \text{ MeV} \]

  (E_{g.s.} = -1772 \text{ MeV})

- Initialize TDHF with **no boost** using the above configurations

- Only case (a) can go to fission, others like before
Details of the Dynamics with No Boost

![Diagram showing the dynamics with no boost.]

- Boost $t=6400 \text{ fm/c}$ state with $|k|=0.02-0.03 \text{ fm}^{-1}$
- $E^*=5-10 \text{ MeV}$

Note the $q_{31}$ symmetry breaking (also $q_{11}$)!

$A_1, Z_1 = 134.5, 51.8$
$A_2, Z_2 = 103.5, 40.2$
How Do We Make Progress?

We can say that we have a reasonable handle on fusion

We may have a handle on quasi-fission

Understanding of fission dynamics is an outstanding challenge in NP

Can we describe many-body quantal fission with TDDFT? Does the KS theory apply here? If yes, what are the ingredients?

Issues with initialization must be better understood for prompt and induced fission. When a neutron transfers its energy to the nucleus what mode or state does this energy go into? What is this state in TDDFT?

Too many variables in the problem, collective boost operator, boost strength, different initial and final states, ...

Brute numerical force may not be sufficient – need more insight