Time-dependent current density functional theory: Rigorous Lattice Formulation

I. V. Tokatly

Universidad del Pais Vasco UPV/EHU, San Sebastian, Spain
and
IKERBASQUE Basque Foundation for Science

ikerbasque
Basque Foundation for Science

Universidad del País Vasco
Euskal Herriko Unibertsitatea

ETSF

Benasque, January 2012

DFT: A Theory of Collective Variables

Most experiments probe the dynamics of some collective variables

density and current	controlled ⟷ probed	Electro-magnetic potentials
$n(\mathbf{r},t), \quad \mathbf{j}(\mathbf{r},t)$		$\varphi(\mathbf{r},t), \quad \mathbf{A}(\mathbf{r},t)$
typical collective variables		typical “experimental tools”

TD(C)DFT is an ideal theoretical setup to address this situation directly

The standard many-body theory: $\{\varphi, \mathbf{A}\} \mapsto |\Psi\rangle \mapsto \{n, \mathbf{j}\}$

Time-dependent (current) density functional theory: $\{\varphi, \mathbf{A}\} \mapsto \{n, \mathbf{j}\}$

In TD(C)DFT the “intermediate” many-body problem is avoided because the collective variables completely determine the state of the system

TDDFT: $n \mapsto |\Psi\rangle = |\Psi[n]\rangle$

TDCDFT: $\mathbf{j} \mapsto |\Psi\rangle = |\Psi[\mathbf{j}]\rangle$

Closed theory of collective variables

(i) TDCDFT: Collective response to a general electro-magnetic field

$$\partial_t n + \partial_\mu j_\mu = 0,$$
$$m\partial_t j_\mu = [\mathbf{j} \times \mathbf{B}]_\mu + nE_\mu - \partial_\nu \Pi_{\mu\nu}[\mathbf{j}]$$

$\Pi_{\mu\nu}[\mathbf{j}] \leftarrow \langle \Psi[\mathbf{j}] | \hat{\Pi}_{\mu\nu} | \Psi[\mathbf{j}] \rangle$

(ii) TDDFT: Density dynamics driven by a scalar potential

$$m\partial_t^2 n = \partial_\mu(n\partial_\mu \varphi) + \partial_\mu \partial_\nu \Pi_{\mu\nu}[n]$$

$\Pi_{\mu\nu}[n] \leftarrow \langle \Psi[n] | \hat{\Pi}_{\mu\nu} | \Psi[n] \rangle$

Such closed theories do exist if we can guarantee the existence of the observable-to-WF map $\mathcal{N} \mapsto |\Psi\rangle$, which can be viewed as a consequence of the observable-to-potential map $\mathcal{N} \leftrightarrow \mathcal{V}$

Standard quantum mechanics solves the "direct problem": $\mathcal{V} \mapsto \mathcal{N}$

TD(C)DFT assumes solvability of the "inverse problem": $\mathcal{N} \mapsto \mathcal{V}$

Question: How to pose the inverse problem mathematically?

Formulation of the problem in TDCDFT

Consider most general many-body Hamiltonian (in a temporal gauge)

$$H[\mathbf{A}] = \sum_{j=1}^{N} \frac{(-i\nabla_j - \mathbf{A}(\mathbf{r}_j, t))^2}{2m} + \frac{1}{2}\sum_{j\neq k} V(|\mathbf{r}_j - \mathbf{r}_k|)$$

Direct problem (given $\mathbf{A}$, $|\Psi_0\rangle$)

$$i\partial_t|\Psi(t)\rangle = H[\mathbf{A}]|\Psi(t)\rangle, \quad |\Psi(0)\rangle = |\Psi_0\rangle$$

$$\hat{n}(\mathbf{r}) = \sum_{j=1}^{N}\delta(\mathbf{r}-\mathbf{r}_j), \quad \hat{\mathbf{j}}^p(\mathbf{r}) = \frac{-i}{2m}\sum_{j=1}^{N}\{\nabla_j, \delta(\mathbf{r}-\mathbf{r}_j)\}$$

$$\mathbf{j}(\mathbf{r},t) = \langle\Psi(t)|\hat{\mathbf{j}}^p(\mathbf{r})|\Psi(t)\rangle - \frac{n(\mathbf{r},t)}{m}\mathbf{A}(\mathbf{r},t), \quad n(\mathbf{r},t) = \langle\Psi(t)|\hat{n}(\mathbf{r})|\Psi(t)\rangle$$

Inverse problem (given $\mathbf{j}$, $|\Psi_0\rangle$)

$$i\partial_t|\Psi(t)\rangle = H[\mathbf{A}]|\Psi(t)\rangle, \quad |\Psi(0)\rangle = |\Psi_0\rangle$$

$$\mathbf{A}(\mathbf{r},t) = \frac{m}{n(\mathbf{r},t)}\left[\langle\Psi(t)|\hat{\mathbf{j}}^p(\mathbf{r})|\Psi(t)\rangle - \mathbf{j}(\mathbf{r},t)\right]$$

This is a nonlinear (self-consistent) quantum many-body problem!

The solution, if exists, gives us $\Psi[\mathbf{j}, \Psi_0]$, $\mathbf{A}[\mathbf{j}, \Psi_0]$

Simple example of the inverse problem: N=1

$$i\partial_t \Psi(\mathbf{r},t) = \frac{1}{2m}(-i\nabla - \mathbf{A}(\mathbf{r},t))^2\Psi(\mathbf{r},t), \quad \Psi(\mathbf{r},0) = \Psi_0(\mathbf{r}) \equiv \sqrt{n_0(\mathbf{r})}e^{i\varphi_0(\mathbf{r})},$$

$$\mathbf{A}(\mathbf{r},t) = \frac{1}{|\Psi|^2}\left[\frac{-i}{2}(\Psi^*\nabla\Psi - \Psi\nabla\Psi^*) - m\mathbf{j}(\mathbf{r},t)\right]$$

This nonlinear problem is exactly solvable!

$$\begin{aligned}
\Psi[\Psi_0,\mathbf{j}](\mathbf{r},t) &= \sqrt{n(\mathbf{r},t)}e^{i\varphi(\mathbf{r},t)}, \\
\mathbf{A}[\Psi_0,\mathbf{j}](\mathbf{r},t) &= \nabla\varphi(\mathbf{r},t) - m\frac{\mathbf{j}(\mathbf{r},t)}{n(\mathbf{r},t)},
\end{aligned}$$

where the functions $n(\mathbf{x},t)$ and $\varphi(\mathbf{x},t)$ are defined as follows

$$n(\mathbf{r},t) = n_0(\mathbf{r}) - \int_0^t dt' \nabla\mathbf{j}(\mathbf{r},t'),$$

$$\varphi(\mathbf{r},t) = \varphi_0(\mathbf{r}) + \int_0^t dt' \left[\frac{\nabla^2\sqrt{n(\mathbf{r},t')}}{2m\sqrt{n(\mathbf{r},t')}} - \frac{m\mathbf{j}^2(\mathbf{r},t')}{2n^2(\mathbf{r},t')}\right]$$

Two ways to approach the general N-body inverse problem of TDCDFT

$$i\partial_t|\Psi(t)\rangle = H[\mathbf{A}]|\Psi(t)\rangle, \quad |\Psi(0)\rangle = |\Psi_0\rangle \tag{1}$$

$$\mathbf{A}(\mathbf{r},t) = \frac{m}{n(\mathbf{r},t)}\left[\langle\Psi(t)|\hat{\mathbf{j}}^p(\mathbf{r})|\Psi(t)\rangle - \mathbf{j}(\mathbf{r},t)\right] \tag{2}$$

(I) "NLSE" approach - Plug $\mathbf{A}[\Psi]$ from Eq. (2) into Eq. (1). The result is a nonlinear Schrödinger equation (NLSE)

$$i\partial_t|\Psi(t)\rangle = \tilde{H}_{\mathbf{j}}[\Psi]|\Psi(t)\rangle, \quad |\Psi(0)\rangle = |\Psi_0\rangle$$

TDCDFT is valid if there exists a unique solution to this NLSE.

(II) "Potential fixed point" approach – Solve Eq. (1) to get $\Psi[\mathbf{A}](t)$, and plug it into Eq. (2). The result is a "fixed point"-type problem:

$$\mathbf{A} = \frac{m}{n}\left[\langle\Psi[\mathbf{A}]|\hat{\mathbf{j}}^p|\Psi[\mathbf{A}]\rangle - \mathbf{j}\right] \equiv \mathcal{F}_{\mathbf{j}}[\mathbf{A}]$$

The existence of TDCDFT is equivalent to the existence of a unique fixed point of the mapping $\mathcal{F}_{\mathbf{j}}[\mathbf{A}] \, : \, \mathcal{B}_{\mathbf{A}} \mapsto \mathcal{B}_{\mathbf{A}}$

Time-dependent current density functional theory on a lattice (NLSE approach)

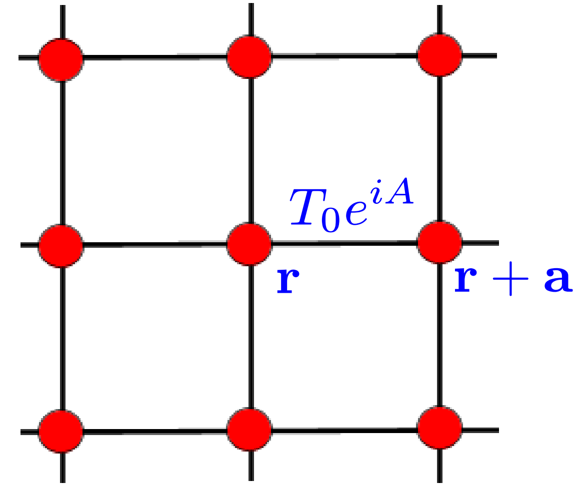
Many-body theory on a lattice (temporal gauge):

N particles on M-site lattice $(N_{\mathcal{H}} = M^N)$

Many-body wave function: $\psi(\mathbf{r}_1, \mathbf{r}_2 \ldots \mathbf{r}_N; t)$

Driving vector potential enters via hopping phases:

$$T(\mathbf{r}, \mathbf{r}+\mathbf{a}) \to T_0 e^{iA(\mathbf{r},\mathbf{r}+\mathbf{a};t)}$$



$$A(\mathbf{r}, \mathbf{r}+\mathbf{a}; t) = \int_{\mathbf{r}}^{\mathbf{r}+\mathbf{a}} \mathbf{A}(\mathbf{x}; t) d\mathbf{x}$$ - link vector potential (two-point object)

<u>Time-dependent Schrödinger equation on a lattice</u>

$$i\partial_t \psi(\mathbf{r}_1 \ldots \mathbf{r}_N; t) = -\sum_{j=1}^{N} \sum_{\mathbf{a}} T_0 e^{iA(\mathbf{r}_j, \mathbf{r}_j+\mathbf{a}; t)} \psi(\ldots \mathbf{r}_j + \mathbf{a} \ldots; t) + \sum_{i>j} V_{\mathbf{r}_i - \mathbf{r}_j} \psi(\mathbf{r}_1 \ldots \mathbf{r}_N; t)$$

$$\psi(\mathbf{r}_1 \ldots \mathbf{r}_N; t_0) = \psi_0(\mathbf{r}_1 \ldots \mathbf{r}_N)$$

Cauchy problem for a system of $N_{\mathcal{H}}$ ordinary differential equations!

Density of particles and the current density on a lattice (identical particles)

On-site density: $n(\mathbf{r};t) = N \sum_{\mathbf{r}_2 \ldots \mathbf{r}_N} |\psi(\mathbf{r},\mathbf{r}_2 \ldots \mathbf{r}_N;t)|^2$

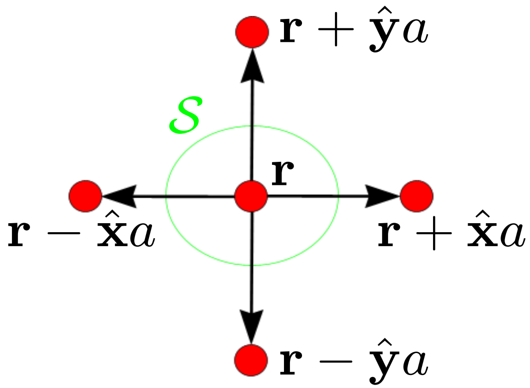
Local current is expressed in terms of a "link density" (density matrix on a link):

$$\rho(\mathbf{r},\mathbf{r}+\mathbf{a}) = N \sum_{\mathbf{r}_2 \ldots \mathbf{r}_N} \psi^*(\mathbf{r},\mathbf{r}_2 \ldots \mathbf{r}_N)\psi(\mathbf{r}+\mathbf{a},\mathbf{r}_2 \ldots \mathbf{r}_N)$$

Link current: $J(\mathbf{r},\mathbf{r}+\mathbf{a};t) = 2\mathrm{Im}\left\{T_0 e^{iA(\mathbf{r},\mathbf{r}+\mathbf{a};t)}\rho(\mathbf{r},\mathbf{r}+\mathbf{a};t)\right\}$

Continuity equation on a lattice

$$\partial_t n(\mathbf{r};t) = -\sum_{\mathbf{a}} J(\mathbf{r},\mathbf{r}+\mathbf{a};t)$$



$K(\mathbf{r},\mathbf{r}+\mathbf{a};t) = 2\mathrm{Re}\left\{T_0 e^{iA(\mathbf{r},\mathbf{r}+\mathbf{a};t)}\rho(\mathbf{r},\mathbf{r}+\mathbf{a};t)\right\}$ - local kinetic energy on a link

Many-body NLSE on a lattice

I. Start from the definition of the current:

$$J(\mathbf{r},\mathbf{r}+\mathbf{a};t) = 2\mathrm{Im}\left\{T_0 e^{iA(\mathbf{r},\mathbf{r}+\mathbf{a};t)}\rho(\mathbf{r},\mathbf{r}+\mathbf{a};t)\right\}$$

$$|K(\mathbf{r},\mathbf{r}+\mathbf{a};t)|^2 + |J(\mathbf{r},\mathbf{r}+\mathbf{a};t)|^2 = 4T_0^2|\rho(\mathbf{r},\mathbf{r}+\mathbf{a};t)|^2$$

II. Express the vector potential as $A[J,\psi]$ $\Downarrow$

$$T_0 e^{iA(\mathbf{r},\mathbf{r}+\mathbf{a};t)} = \frac{\sqrt{4T_0^2|\rho(\mathbf{r},\mathbf{r}+\mathbf{a};t)|^2 - J^2(\mathbf{r},\mathbf{r}+\mathbf{a};t)} + iJ(\mathbf{r},\mathbf{r}+\mathbf{a};t)}{2\rho(\mathbf{r},\mathbf{r}+\mathbf{a};t)} = T_J[\psi](\mathbf{r},\mathbf{r}+\mathbf{a})$$

III. Insert it into the Schrödinger equation: $\Downarrow$

$$i\partial_t\psi(\mathbf{r}_1\ldots\mathbf{r}_N) = -\sum_{j,\mathbf{a}} T_J[\psi](\mathbf{r}_j,\mathbf{r}_j+\mathbf{a})\psi(\ldots\mathbf{r}_j+\mathbf{a}\ldots) + \sum_{i>j} V_{\mathbf{r}_i-\mathbf{r}_j}\psi(\mathbf{r}_1\ldots\mathbf{r}_N)$$

$$\psi(\mathbf{r}_1\ldots\mathbf{r}_N;t_0) = \psi_0(\mathbf{r}_1\ldots\mathbf{r}_N)$$

Hence the problem reduces to a system of $N_{\mathcal{H}}$ nonlinear ODE:

$$\dot{\boldsymbol{\psi}} = \mathbf{F}(\boldsymbol{\psi},t), \qquad \boldsymbol{\psi}(t_0) = \boldsymbol{\psi}_0,$$

which, by Picard's theorem, has a unique solution if $\mathbf{F}(\boldsymbol{\psi},t)$ is Lipschitz in $\boldsymbol{\psi}$-variables

The nonlinearity in NLSE is determined by the hopping parameters:

$$T_J[\psi](\mathbf{r},\mathbf{r}+\mathbf{a};t) = \frac{\sqrt{4T_0^2|\rho(\mathbf{r},\mathbf{r}+\mathbf{a};t)|^2 - J^2(\mathbf{r},\mathbf{r}+\mathbf{a};t)} + iJ(\mathbf{r},\mathbf{r}+\mathbf{a};t)}{2\rho(\mathbf{r},\mathbf{r}+\mathbf{a};t)}$$

On a lattice the physical (A-representable) link currents are bounded from above

$$|J(\mathbf{r},\mathbf{r}+\mathbf{a})| < 2T_0|\rho(\mathbf{r},\mathbf{r}+\mathbf{a})| \qquad (1)$$

The physical reason is that the maximal "hopping rate" is bounded by T_0

[Eq. (1) implies a bound on $\partial_t n$ (Baer, Ullrich, Verdozzi)]

$|J(\mathbf{r},\mathbf{r}+\mathbf{a})| = 2T_0|\rho(\mathbf{r},\mathbf{r}+\mathbf{a})| \implies |K(\mathbf{r},\mathbf{r}+\mathbf{a})| = 0$ - vanishing kinetic energy on the link

Using Cauchy-Schwarz inequality we find an upper bound on A-representable currents:

$$|J(\mathbf{r},\mathbf{r}+\mathbf{a})| < 2T_0\sqrt{n(\mathbf{r})n(\mathbf{r}+\mathbf{a})} \leq 2T_0, \quad \text{(for fermions)}$$

In general inequality (1) determines a subset of "A-representability" in the Hilbert space $\mathcal{H}$

NLSE: $i\partial_t \psi(t) = \hat{T}_J[\psi]\psi(t) + \hat{V}\psi(t), \quad \psi(t_0) = \psi_0$

Theorem (The existence of lattice TDCDFT)

Let $J(\mathbf{r}, \mathbf{r}+\mathbf{a}; t)$ be continuous functions of t, such that in the extended phase space $\mathcal{H} \times R$ there exists a subset Ω defined by

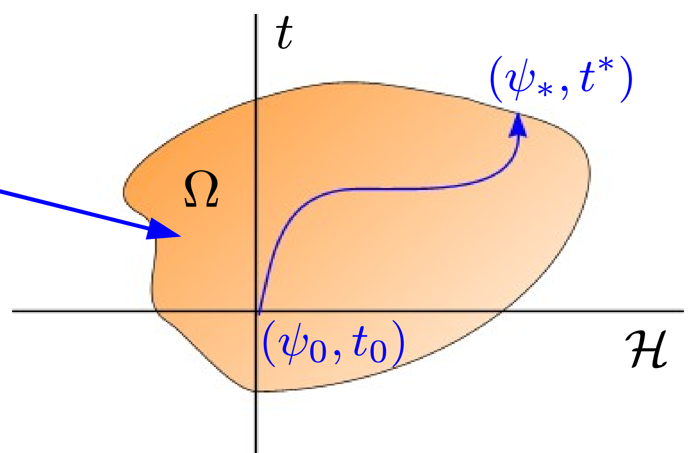
$$2T_0|\rho(\mathbf{r}, \mathbf{r}+\mathbf{a})| > |J(\mathbf{r}, \mathbf{r}+\mathbf{a}; t)|.$$

If the initial point $(\psi_0, t_0) \in \Omega$, then

(i) There is a neighborhood of (ψ_0, t_0) where the $\psi(t)$ is a unique functional of $J(t)$ and ψ_0, and the map $J \leftrightarrow A$ is unique and invertible;

(ii) The statement (i) can not be extended beyond some maximal existence time t^*, if and only if at time t^* the boundary of Ω is reached.

A-representability subset (non-zero kinetic energy)



The solution is not global only if it hits the boundary, i. e., at least for one link:

$t \to t^*, \quad |K(\mathbf{r}, \mathbf{r}+\mathbf{a}; t)| \to 0$

Explicit example: One particle on a lattice (N=1)

$$i\partial_t \psi(\mathbf{r};t) = -\sum_{\mathbf{a}} T_J[\psi](\mathbf{r},\mathbf{r}+\mathbf{a};t)\psi(\mathbf{r}+\mathbf{a};t), \qquad \psi(\mathbf{r};t_0) = |\psi_0(\mathbf{r})|e^{i\chi_0(\mathbf{r})}$$

$$T_J[\psi](\mathbf{r},\mathbf{r}+\mathbf{a};t) = \frac{\sqrt{4T_0^2|\psi^*(\mathbf{r};t)\psi(\mathbf{r}+\mathbf{a};t)|^2 - J^2(\mathbf{r},\mathbf{r}+\mathbf{a};t)} + iJ(\mathbf{r},\mathbf{r}+\mathbf{a};t)}{2\psi^*(\mathbf{r};t)\psi(\mathbf{r}+\mathbf{a};t)}$$

The exact solution: $\psi(\mathbf{r};t) = |\psi(\mathbf{r},t)|e^{i\chi(\mathbf{r},t)}$

$$|\psi(\mathbf{r},t)| = \sqrt{|\psi_0(\mathbf{r})|^2 - \int_{t_0}^{t}\sum_{\mathbf{a}} J(\mathbf{r},\mathbf{r}+\mathbf{a};t')dt'}$$

$$K(\mathbf{r},\mathbf{r}+\mathbf{a};t) = \sqrt{4T_0^2|\psi(\mathbf{r};t)|^2|\psi(\mathbf{r}+\mathbf{a};t)|^2 - J^2(\mathbf{r},\mathbf{r}+\mathbf{a};t)}$$

$$\chi(\mathbf{r},t) = \chi_0(\mathbf{r}) + \int_{t_0}^{t}\frac{\sum_{\mathbf{a}} K(\mathbf{r},\mathbf{r}+\mathbf{a};t')}{2|\psi(\mathbf{r},t')|^2}dt'$$

The maximal existence time, if $t^* < \infty$, is determined by $K(\mathbf{r},\mathbf{r}+\mathbf{a};t^*) = 0$

The behavior of one-particle system is generic!
There is no conceptual difference between N=1 and N>1

General comments on the existence theorem for the lattice TDCDFT

1. For a “physical” initial state any continuous in t current J(t) is locally A-representable. Since the statement of the theorem does no depend on interactions both interacting and noninteracting A-representability is guaranteed locally.

2. If the current J(t) is t-analytic, then it is locally (both interacting and noninteracting) A-representable. The corresponding potential A(t) is also t-analytic. This completes the van Leeuwen-type argumentation by proving the convergence of the power series for the potential.

3. In general the conditions for the global existence may be different for interacting and noninteracting systems. Currently we can not exclude a situation when a physical (for interacting system) current will drive the KS system to the border if its A-representability domain.

4. The t-continuity restriction on the currents/potentials can be easily relaxed to a piecewise continuity, which is sufficient to cover most physically relevant cases.

Generalizations, Open Questions, Problems, etc...

1. Extension to the lattice TDDFT (*see poster by Mehdi Farzanehpour*)

2. Relation of NLSE to the fixed point approach of Ruggenthaler and van Leeuwen (*clearly the unique solution of NLSE implies the existence of a unique fixed point of the map* $\mathcal{F}_J[A]$. *Does it mean that this mapping is contractive?*)

3. It looks like in the "hydrodynamic" implementation of TDCDFT the boundary of the A-representability subset is never reached. Can we really prove this?

4. Can we say anything about topology of the A-representability subset. Are there some general relations between those subsets for interacting and KS systems.

5. Big Open Question is the continuum limit. (*An encouraging observation is that the exact solution for one particle on a lattice perfectly converges to its continuum counterpart. Can we expect a similar behavior for the rest of the theory.*)