Constrained Schrödinger Dynamics and the Foundations of TDDFT

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18/03/2026

TDDFT = Time–Dependent Density Functional Theory

Goal : Describe the electronic dynamics submited to a time-dependent external potential, using the one-body density ρ instead of the many-body wavefunction Ψ.

In practice : Use non-interacting fictitious electrons that reproduce ρ_Ψ.

1D Helium with Soft-Coulomb interaction

2 electrons in a singlet state

Interacting system

$$i\partial_t \Psi^S(t,x_1,x_2) = H(t)\Psi^S(t,x_1,x_2)$$

$$H(t) = \sum_{i=1}^{2} h(t,x_i) + \frac{1}{\sqrt{(x_1-x_2)^2+1}}$$

Kohn–Sham system

$$i\partial_t \varphi(t,x) = \big(h + V_{\mathrm{Hxc}}[\rho_\varphi]\big)(t,x)\varphi(t,x)$$

$$h(t,x) = -\tfrac{1}{2}\partial_x^2 - \frac{2}{\sqrt{x^2+1}} + V_{\mathrm{elec}}(t,x)$$

Give ρ^S → Give ρ_φ

$V_{\mathrm{Hxc}}[\rho_\varphi](t,x)$ is chosen so that $\rho^S = \rho_\varphi := 2|\varphi(t,x)|^2$.

Limitations in non–adiabtic regime

- Today, TDDFT is mostly used with **adiabatic approximations** :

$$V_{\mathrm{Hxc}}(\rho_\varphi)(t) = V_{\mathrm{Hxc}}^{\mathrm{LDA,\,GGA,\ldots}}(\rho_\varphi(t))$$

$\Longrightarrow$ Works well in **near-adiabatic regimes** but fail in **non-adiabatic regimes** (e.g. charge-transfer processes).

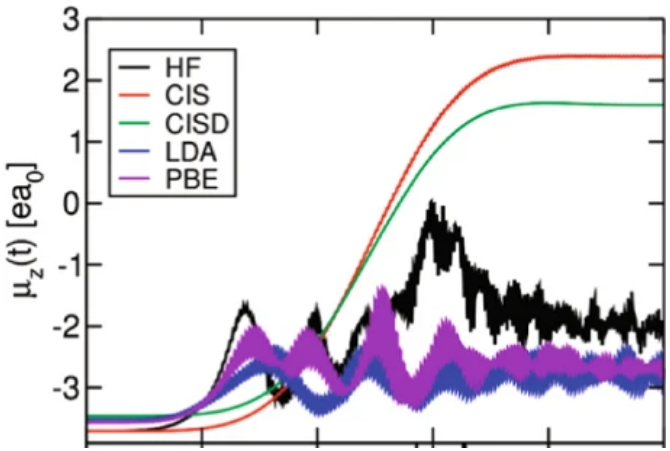

Figure 1 – Charge-transfer in LiCN.

- Main limit : **Absence of memory effects** : $V_{\mathrm{Hxc}}(\rho_\varphi)(t) = F(\rho_\varphi(t'),\, t' \leq t)$

Lionel Lacombe et Neepa T. Maitra. "Non-adiabatic approximations in time-dependent density functional theory : progress and prospects". In : (2023)

Mathematical Foundation of TDDFT

- **Time-dependent variational principle** :

  Assuming the interacting density is known, how can one enforce a Kohn–Sham system to reproduce it ?

  $\Longrightarrow$ Can be achieved by introducing an additional real potential V_{Hxc}.

Mathematical Foundation of TDDFT

- **Time-dependent variational principle** :

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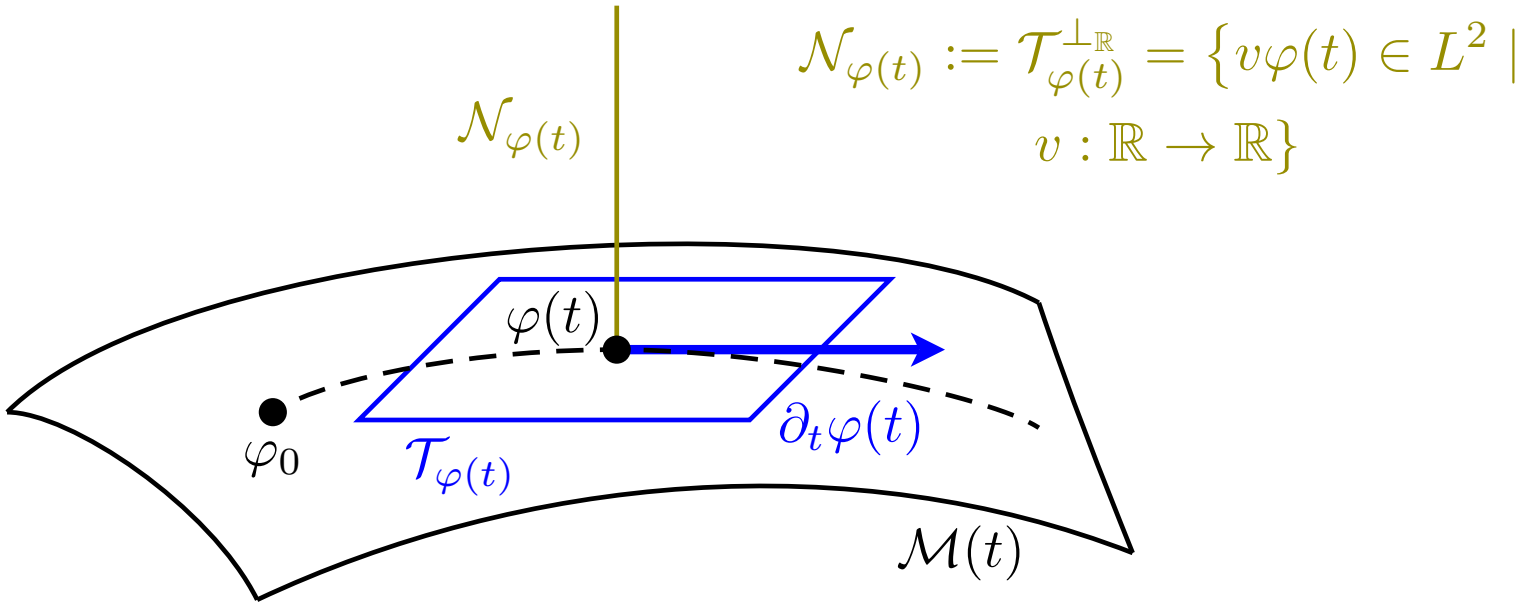
Is the time-dependent variational principle the only natural principle for TDDFT ?

Geometric structure of the problem

We still consider the 1D helium system in a singlet state.
Assume you know the interacting density ρ^S :

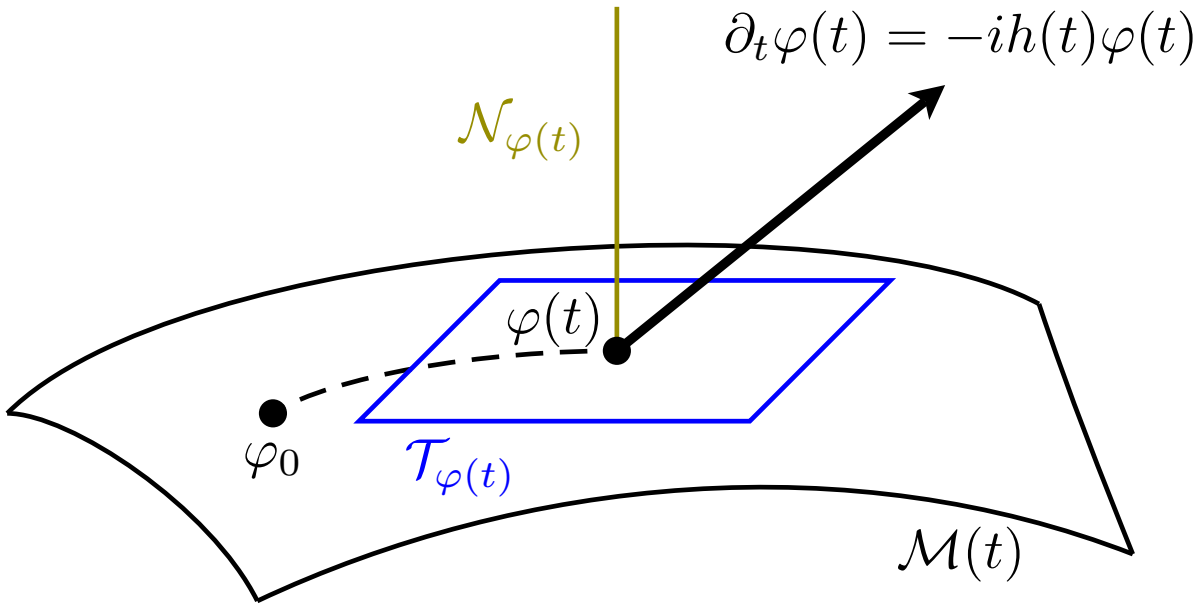


$$\mathcal{M}(t) := \{\varphi(t) \in L^2 \mid \rho_\varphi(t,x) = \rho^S(t,x)\}$$

$\mathcal{M}(t)$ is an infinite-dimensional **real** manifold.

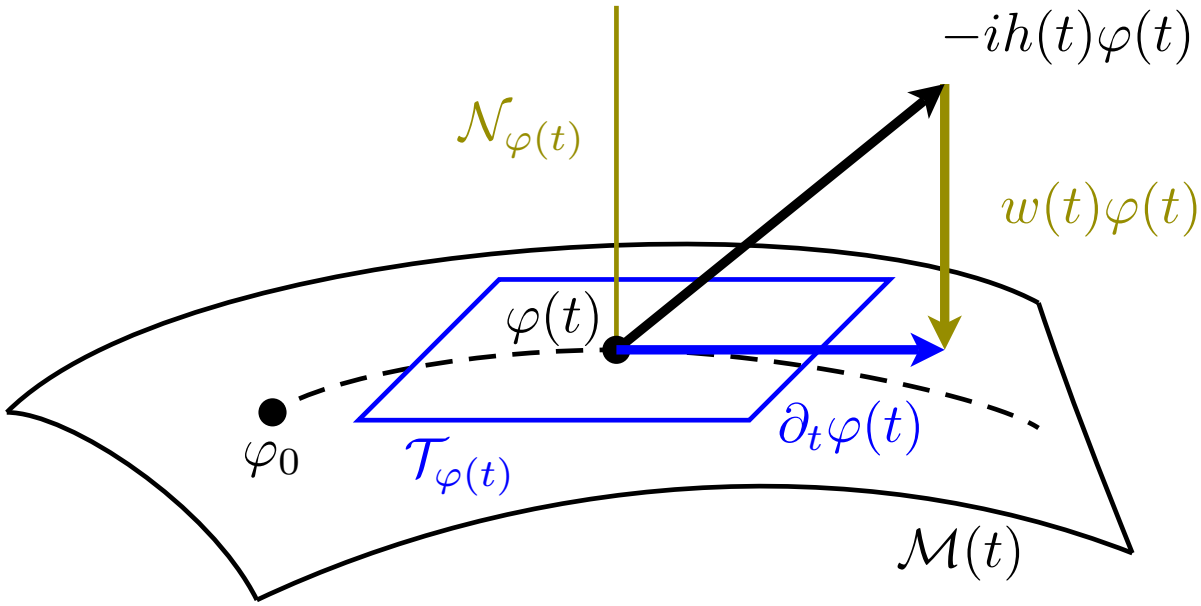
Geometric Principle (GP)

At time t, if no correction term is added, the trajectory leaves the manifold.



Geometric Principle (GP)

Project $-ih(t)\varphi(t)$ onto the real vector space $\mathcal{T}_{\varphi(t)}$!



$$\partial_t\varphi(t) + ih(t)\varphi(t) = \underbrace{w(t)}_{\mathbb{R}\to\mathbb{R}}\varphi(t) \in \mathcal{N}_{\varphi(t)}$$

$\Longrightarrow$ purely imaginary "potential"

Time-dependent Variational Principle (TDVP)

A trajectory $\varphi(t) \in \mathcal{M}(t)$ satisfies the **TDVP** if it is a critical point of the action

$$\mathcal{A}[\eta] = \int_0^T \langle \eta(t) \left[i\partial_t - h(t) \right] \eta(t) \rangle_{L^2} \, \mathrm{d}t$$

defined for paths $\eta(t) \in \mathcal{M}(t)$ such that $\eta(0) = \varphi(0)$ and $\eta(T) = \varphi(T)$.

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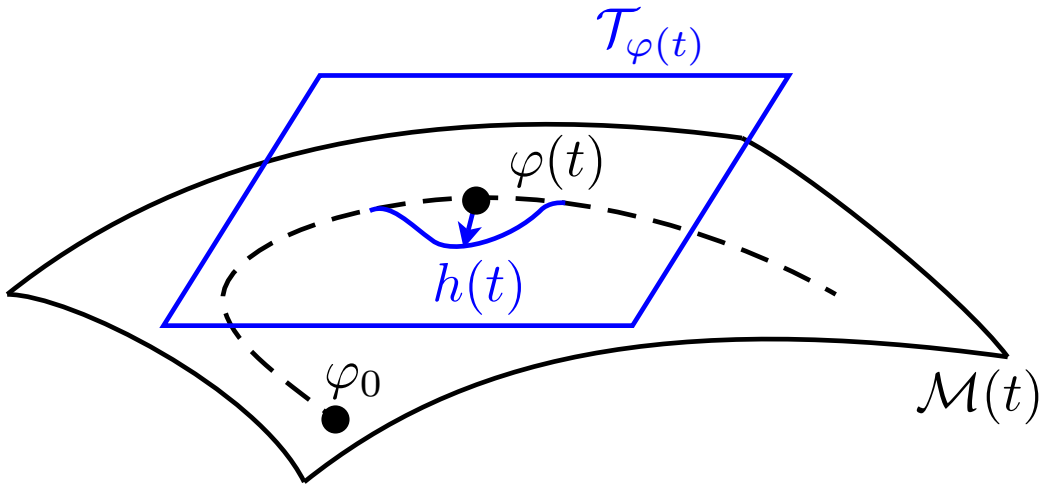

$$\partial_t \varphi(t) + ih(t)\varphi(t) = i \underbrace{v(t)}_{\mathbb{R}\to\mathbb{R}} \varphi(t) \in i\mathcal{N}_{\varphi(t)}$$

Two different principles

- **TDVP** $\Longrightarrow$ Correction in $i\mathcal{N}_{\varphi(t)}$

$$i\partial_t\varphi(t,x) = -\tfrac{1}{2}\partial_x^2\varphi(t,x) + V_{\text{ext}}(t,x)\varphi(t,x) + V(t,x)\varphi(t,x)$$

- **GP** $\Longrightarrow$ Correction in $\mathcal{N}_{\varphi(t)}$

$$i\partial_t\varphi(t,x) = -\tfrac{1}{2}\partial_x^2\varphi(t,x) + V_{\text{ext}}(t,x)\varphi(t,x) + iW(t,x)\varphi(t,x)$$

$\mathcal{M}(t)$ is not a Kähler Manifold !

$$\boxed{i\mathcal{N}_{\varphi(t)} \neq \mathcal{N}_{\varphi(t)}}$$

GP as a correction to TDVP

It is possible to mix TDVP and GP !

For example, if you have a TDVP approximation V, you can add it to your non-interacting system and then apply the GP :

$$\begin{cases} i\partial_t \varphi(t,x) = \left[-\frac{1}{2}\partial_x^2 + V_{\text{ext}}(t,x) + V(t,x) + iW(t,x)\right]\varphi(t,x), \\ \varphi(0,x) = \sqrt{\frac{\rho^S(0,x)}{2}} \end{cases}$$

where V and W are chosen to enforce $\rho_\varphi(t,x) = 2|\varphi(t,x)|^2 = \rho^S(t,x)$.

$\Longrightarrow$ **Some degrees of freedom.**

Hydrodynamic picture

If $\varphi(t,x) = \sqrt{\frac{\rho^S(t,x)}{2}} e^{i\theta(t,x)}$, then the triplet (θ, V, W) solves

$$\begin{cases} W = \dfrac{\partial_t \rho^S + \partial_x(\rho^S \partial_x \theta)}{2\rho^S}, & (1) \\ V = -\partial_t \theta - \dfrac{|\partial_x \theta|^2}{2} + \dfrac{\partial_x^2 \sqrt{\rho^S}}{2\sqrt{\rho^S}} - V_{\text{ext}}. & (2) \end{cases}$$

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Several interesting cases :

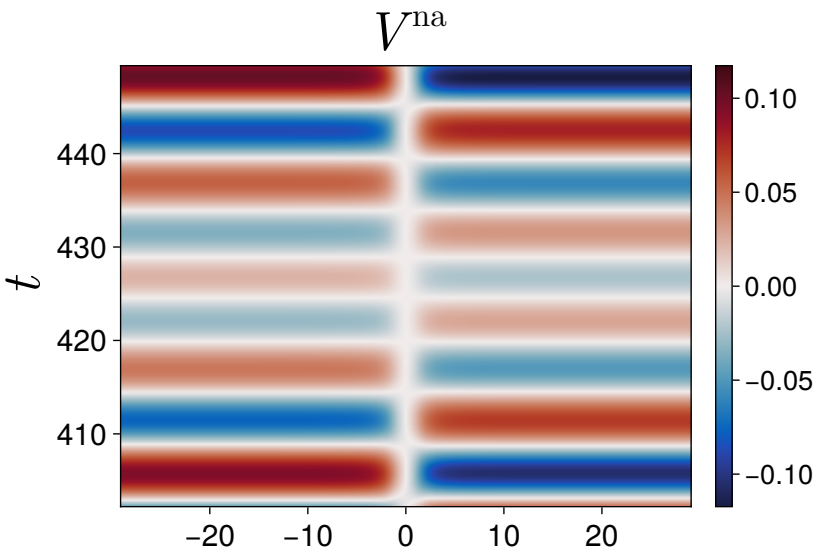
1. **TDVP** : $W = 0 \Rightarrow V^{\text{eKS}} := -\frac{1}{2}\left(\frac{j_\varphi}{\rho^S}\right)^2 - \int^x \partial_t \left(\frac{j_\varphi}{\rho^S}\right) + \frac{\partial_x^2 \sqrt{\rho^S}}{2\sqrt{\rho^S}} - V_{\text{ext}}$.
2. **GP** : $V = 0 \;\Rightarrow$ Solve (2) to find θ and use (1) to deduce W.
3. $\theta = 0$: No equation to solve !

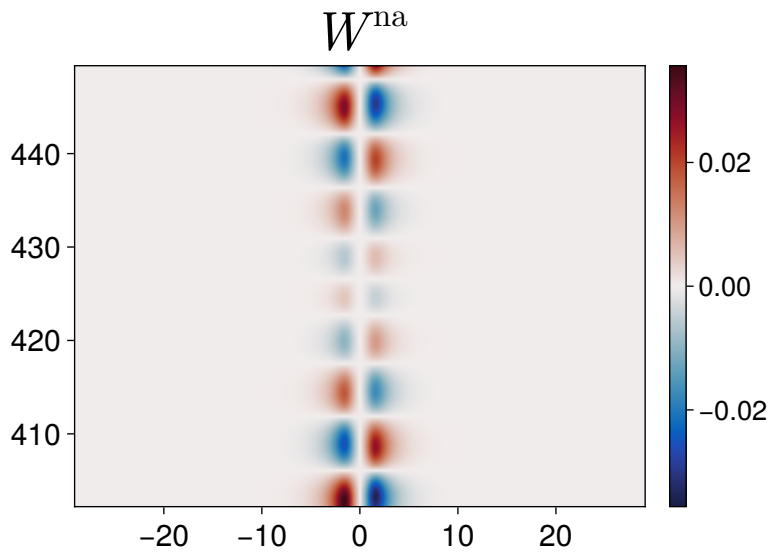
$$V^{\text{eaKS}} := \frac{\partial_x^2 \sqrt{\rho^S}}{2\sqrt{\rho^S}} - V_{\text{ext}}, \quad W^{\text{na}} := \frac{\partial_t \rho^S}{2\rho^S}.$$

We want to compare W^{na} with $V^{\text{na}} := V^{\text{eKS}} - V^{\text{eaKS}}$.

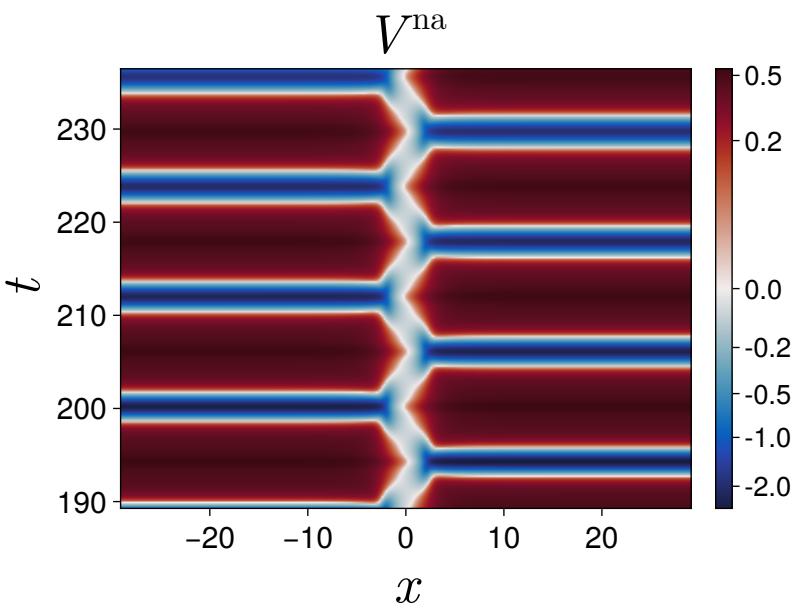
Rabi Oscillation in 1D Helium : $\psi_{\mathrm{gs}} \xrightarrow{T_R} \psi_{\mathrm{exc}} \xrightarrow{T_R} \psi_{\mathrm{gs}} \xrightarrow{T_R} \cdots$

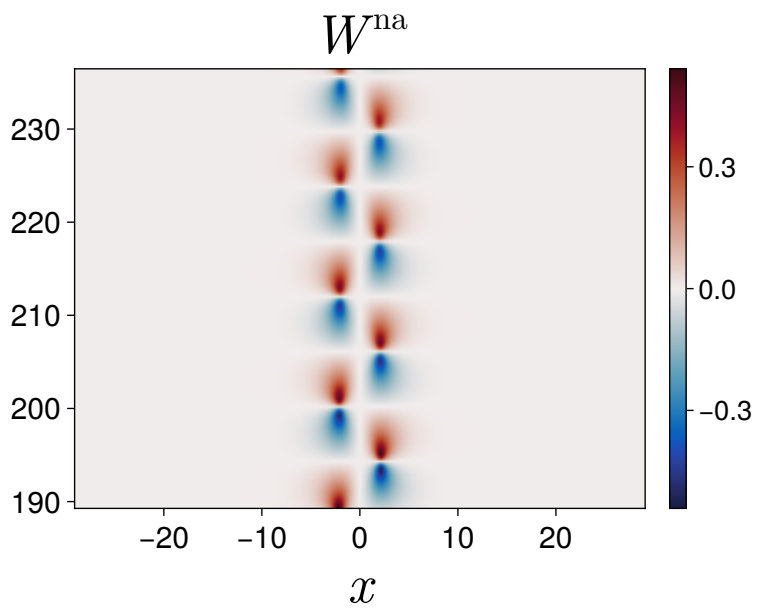
T_R





$T_R/2$





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- Éric Cancès et al. *Geometric theory of constrained Schrödinger dynamics with application to time-dependent density-functional theory on a finite lattice*. 2026. arXiv : 2601.07719. url : https://arxiv.org/abs/2601.07719
  - Abstract constrained Schrödinger equation on a lattice for arbitrary observables.
  - TDVP-representable $\Longrightarrow$ GP-representable
- Éric Cancès et al. *Geometric Time-Dependent Density Functional Theory*. 2026. arXiv : 2601.07724. url : https://arxiv.org/abs/2601.07724
  - Introduce formally the continuous case.
  - Formulation of orbital free TDDFT.

Thank you for your attention !