

# New variational principle for dynamical low-complexity approximations for the time-dependent Schrödinger equation

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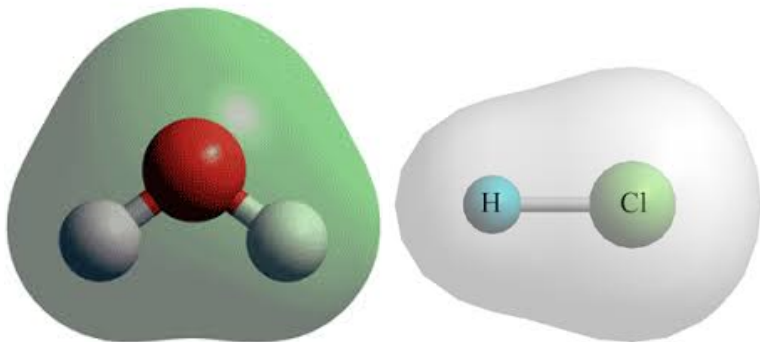
# Outline of the talk

- 1 Aim and motivation
- 2 Variational formulation of the time-dependent Schrödinger equation
- 3 Application to the many-body electronic Schrödinger problem
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# Motivation: electronic structure calculation for molecules



Computation of the **evolution in time of the state of the set of electrons** in a molecule:  
electrical, magnetical, optical properties...

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- $M$  nuclei, that are assumed to be (fixed) classical point charges, whose positions and electric charges are denoted by  $R_1, \dots, R_M \in \mathbb{R}^3$  and  $Z_1, \dots, Z_M \in \mathbb{N}^*$  respectively;



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- $N$  electrons, considered as quantum particles: at time  $t \in \mathbb{R}$ , the state of the electrons is represented by a complex-valued function  $\psi(t) : \mathbb{R}^{3N} \rightarrow \mathbb{C}$ . The function  $\psi(t)$  is called the **wavefunction** of the system of electrons at time  $t \in \mathbb{R}$ .

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## Physical interpretation of the wavefunction:

For  $x_1, \dots, x_N \in \mathbb{R}^3$ , the quantity  $|\psi(t, x_1, \dots, x_N)|^2$  represents the probability density at time  $t$  of the positions  $x_1, \dots, x_N$  of the  $N$  electrons.

For  $B \subset \mathbb{R}^{3N}$ ,

$$\int_B |\psi(t, \cdot)|^2: \text{probability that the electrons are located in the set } B \text{ at time } t.$$

# Time-dependent Schrödinger equation

$$\begin{cases} i\partial_t\psi(t) - H\psi(t) = 0, & t \in (0, T) \\ \psi(0) = \psi_0 \end{cases} \quad (1)$$

where the operator

$$H = H_0 + A$$

is a self-adjoint operator on  $\mathcal{H} = L^2(\mathbb{R}^{3N})$  with domain  $D(H) = H^2(\mathbb{R}^{3N})$  called the **Hamiltonian** of the system of electrons and is given by

$$H_0 = -\Delta_{x_1, \dots, x_N} \quad (\text{kinetic energy})$$

and

$$A = V(x_1, \dots, x_N) = \sum_{k=1}^M \sum_{i=1}^N \frac{-Z_k}{|x_i - R_k|} + \sum_{1 \leq i < j \leq N} \frac{1}{|x_i - x_j|} \quad (\text{coulombic energy})$$

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**$N$  large  $\Rightarrow$  Curse of dimensionality!**

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**Dynamical low-complexity approximation:** The aim is to compute an approximation  $\tilde{\psi}$  of  $\psi$  such that  $\tilde{\psi}(t) \in \Sigma$  for all  $t$ .

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Assume that  $\Sigma$  is a **regular submanifold** of  $\mathcal{H}$ .

**Dirac-Frenkel variational principle:** Find  $\tilde{\psi}$  such that for almost all  $t$ ,

$$\partial_t \tilde{\psi}(t) \in \operatorname{argmin}_{v \in T_{\tilde{\psi}(t)} \Sigma} | -iH\tilde{\psi}(t) - v |^2$$

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$$\langle (i\partial_t - H)\tilde{\psi}(t), \delta\tilde{\psi} \rangle = 0, \quad \forall \delta\tilde{\psi} \in T_{\tilde{\psi}(t)} \Sigma. \quad (2)$$

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**Problem:** In general, the low-complexity sets  $\Sigma$  which are used in practice are not regular everywhere. As a consequence, except in some particular situations, one can only obtain **the local existence in time** of a solution  $\tilde{\psi}$  to (2).



## Alternative: quadratic variational formulation of the TD Schrödinger equation

Our aim here is to express equivalently the solution  $\psi$  of (8) as the solution of a variational problem of the form

$$\forall \varphi \in \mathcal{X}_H, \quad a(\psi, \varphi) = b(\varphi)$$

with

- $\mathcal{X}_H$  a Hilbert space of functions depending both on the time and space variable;
- $a : \mathcal{X}_H \times \mathcal{X}_H$  a continuous hermitian coercive sesquilinear form
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**There are several ways to do so!**

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- well-defined on the whole time interval  $(0, T)$  whatever the value of the final time  $T$
- certified a posteriori error estimator

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## Notation and definition of weak solutions

- Let  $\mathcal{H}$  be a Hilbert space equipped with a scalar product  $\langle \cdot, \cdot \rangle$  and associated norm  $|\cdot|$
- Let  $H$  be a self-adjoint operator on  $\mathcal{H}$  with domain  $D(H)$
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### Definition (Notion of weak solutions)

A function  $u^* \in L^2(I; \mathcal{H})$  is said to be a weak solution to (3) if and only if

$$(C1) \quad \forall v \in C_c^0(I, D(H)) \cap C_c^1(I, \mathcal{H}),$$

$$(u^* | (i\partial_t - H)v)_{L^2(I; \mathcal{H})} = (f | v)_{L^2(I; \mathcal{H})}$$

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**Remark:** Actually, (C1) implies that  $u^* \in C^0(\bar{I}; \mathcal{H})$ , which enables to give a meaning to (C2)

## A first variational formulation (not useful)

Define

$$\mathcal{X}_H = \{u^* \in L^2(I; \mathcal{H}) : \exists(u_0, f) \in \mathcal{H} \times L^2(I; \mathcal{H}) \text{ such that } u^* \text{ solves (3)}\}$$

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This space is a Hilbert space when equipped with the inner product

$$\forall u, v \in \mathcal{X}_H, (u, v)_{\mathcal{X}_H} = \langle u(0), v(0) \rangle + T \|(i\partial_t - H)u\|_{L^2(I; \mathcal{H})} \|(i\partial_t - H)v\|_{L^2(I; \mathcal{H})} \quad (4)$$

The associated norm is then denoted by

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**Problem:** what is the space  $\mathcal{X}_H$ ?

## Theorem

*The application*

$$\begin{cases} L^2(I; \mathcal{H}) & \rightarrow & L^2(I; \mathcal{H}) \\ u & \mapsto & e^{itH} u \end{cases} \quad (6)$$

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**Problem again:** the evolution group  $e^{-itH}$  is not easy to compute/characterize in general

Main idea:  $H = H_0 + A$

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The proofs of the following results rely on **Kato's smoothing theory** [Reed, Simon, 1978]

## Assumptions (A):

- (A1) The operator  $H_0$  is a self-adjoint operator on  $\mathcal{H}$  with domain  $D(H_0)$
- (A2) The operator  $A$  is a closed symmetric operator on  $\mathcal{H}$  such that  $D(H_0) \subset D(A)$
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$$\forall u \in \mathcal{X}_{H_0}, \quad \frac{\alpha}{1+T} \|u\|_{\mathcal{X}_{H_0}} \leq \|u\|_{\mathcal{X}_H} \leq C(1+T) \|u\|_{\mathcal{X}_{H_0}}$$

## Consequence: second variational formulation

$$u^* = \operatorname{argmin}_{u \in \mathcal{X}_{H_0}} |u(0) - u_0|^2 + \|(i\partial_t - H_0 - A)u - f\|_{L^2(I; \mathcal{H})}^2$$



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and  $e^{itH_0} H_0 e^{-itH_0} v = e^{itH_0} e^{-itH_0} H_0 v$  because  $H_0$  commutes with  $e^{-itH_0}$ .

### Theorem

Let  $H_0$  and  $A$  be operators on  $\mathcal{H}$  satisfying (A). Let  $u_0 \in \mathcal{H}$  and  $f \in L^2(I; \mathcal{H})$ .



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**Remark:** We obtain a similar result in the case when  $u^*$  is the solution of a time-dependent Schrödinger equation of the form

$$\begin{cases} i\partial_t u^*(t) - (H_0 + A + B(t))u^*(t) = f(t), & t \in I, \\ u^*(0) = u_0 \end{cases}$$

where  $B : I \ni t \mapsto B(t)$  is a strongly continuous family of **bounded** self-adjoint operators on  $\mathcal{H}$ .

# Outline of the talk

- 1 Aim and motivation
- 2 Variational formulation of the time-dependent Schrödinger equation
- 3 Application to the many-body electronic Schrödinger problem**
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# Many-body electronic Schrödinger problem

$$\begin{cases} i\partial_t\psi(t) - H\psi(t) = 0, & t \in (0, T) \\ \psi(0) = \psi_0 \end{cases} \quad (8)$$

where the operator  $H = H_0 + A$  is a self-adjoint operator on  $\mathcal{H} = L^2(\mathbb{R}^{3N})$  with domain  $D(H) = H^2(\mathbb{R}^{3N})$  is given by

$$H_0 = -\Delta_{x_1, \dots, x_N} \quad (\text{kinetic energy})$$

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$$A = V(x_1, \dots, x_N) = \sum_{k=1}^M \sum_{i=1}^N \frac{-Z_k}{|x_i - R_k|} + \sum_{1 \leq i < j \leq N} \frac{1}{|x_i - x_j|} \quad (\text{coulombic energy})$$

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stems from **Kato-Yajima inequality**: [Kato, Yajima, 1989], [Burq, 2004]

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Then, there exist constants  $C, \alpha > 0$  such that for any  $v \in H^1(I, L^2(\mathbb{R}^{3N}))$ ,

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Rather look for  $\tilde{\psi} = e^{it\Delta} \tilde{v}$  where  $\tilde{v}$  is defined as a solution to

$$\tilde{v} \in \operatorname{argmin}_{\tilde{w} \in H^1(I; \Sigma)} F(\tilde{w}) \quad (12)$$

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Let  $\Sigma$  be a *weakly closed subset* of  $\mathcal{H}$ . Then,  $H^1(I; \Sigma)$  is a weakly closed subset of  $H^1(I; \mathcal{H})$ . Hence, there always exists at least one solution to (12).

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- **global in time existence** of dynamical low-complexity approximations provided that  $\Sigma$  is a weakly closed subset of  $\mathcal{H} = L^2(\mathbb{R}^{3N})$ .

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Let  $\Sigma$  be a *weakly closed subset* of  $\mathcal{H}$ . Then,  $H^1(I; \Sigma)$  is a weakly closed subset of  $H^1(I; \mathcal{H})$ . Hence, there always exists at least one solution to (12).

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### Very nice property for low-complexity approximations:

- **low-rank tensor formats**:  $e^{it\Delta}$  is a pure tensor product of operators:

$$e^{it\Delta_{x_1, \dots, x_N}} = e^{it\Delta_{x_1}} \otimes \dots \otimes e^{it\Delta_{x_N}}$$

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**How does it compare with the dynamical low-complexity approximation given by the Dirac-Frenkel variational principle?**

**Finite-dimensional version of the Schrödinger equation:**  $n \in \mathbb{N}^*$

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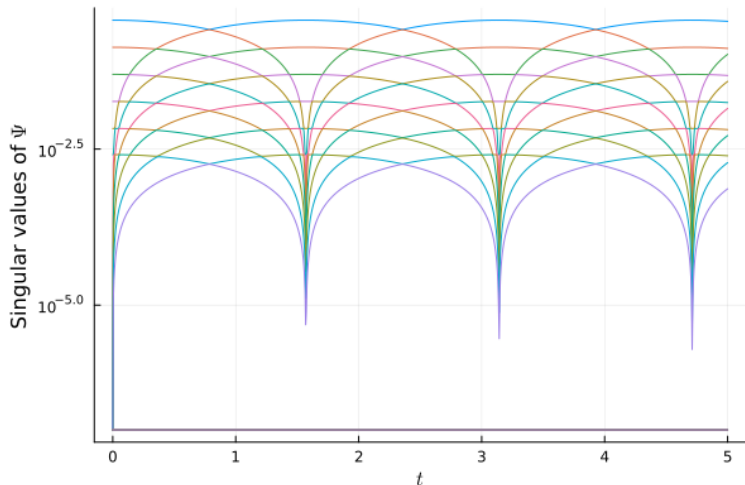
We look for a **rank- $r$**  approximation  $\tilde{\Psi}$  of  $\Psi$ , i.e. for  $r \in \mathbb{N}^*$ ,

$$\Sigma = \left\{ \tilde{\Psi} = \underbrace{\Phi_x}_{\in \mathbb{C}^{m,r}} \underbrace{\Phi_y^T}_{\in \mathbb{C}^{r,n}} \right\}$$



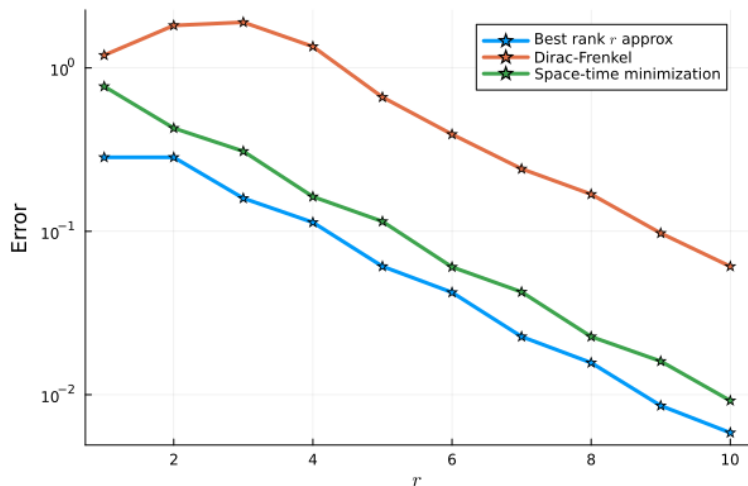
# Singular value decomposition

For all  $t \in [0, T]$ , a best approximation of  $\Psi(t)$  by an element of  $\Sigma$  is given as a **rank- $r$  truncated Singular Value Decomposition** of  $\Psi(t)$ .



# Comparison of dynamical low-rank approximations

Errors in  $L^\infty((0, T); \mathbb{C}^{n \times n})$  between the various dynamical low-rank approximations and the exact solution as a function of the rank  $r$



$$\Sigma = \left\{ e^{-(x-q)^T A (x-q) + i p^T x} : p, q \in \mathbb{R}^{3N}, A = P + iQ \text{ with } P, Q \in \mathbb{R}_{\text{sym}}^{3N \times 3N}, P \text{ definite positive} \right\}.$$

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**See Clément Guillot's poster on Thursday for more details!**

# Outline of the talk

- 1 Aim and motivation
- 2 Variational formulation of the time-dependent Schrödinger equation
- 3 Application to the many-body electronic Schrödinger problem
- 4 Dynamical low complexity approximations
- 5 Summary**

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  - **Open question:** how to impose norm conservation in this global space-time formulation? Not completely obvious...

**Thank you for your attention!**