Low-complexity approximations with least-squares formulation of the time-dependent Schrödinger equation

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July 22, 2025

The atom

- Assume we want to study a system with M protons and N electrons
- Protons are much heavier than electrons $\Rightarrow$ We assume they have fixed position $X_1, ..., X_M \in \mathbb{R}^3$

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- Assume we want to study a system with M protons and N electrons
- Protons are much heavier than electrons $\Rightarrow$ We assume they have fixed position $X_1, ..., X_M \in \mathbb{R}^3$
- The positions $x_1(t), ..., x_N(t) \in \mathbb{R}^3$ of the electrons satisfy the equations of movement:

$$\begin{cases} \ddot{x_j}(t) = -\nabla V(x_1(t), ..., x_N(t)), \\ \dot{x_j}(0) = v_{j,0}, \\ x_j(0) = x_{j,0}, \end{cases}$$

$$V(x_1, ..., x_N) = \underbrace{\sum_{j=1}^{N} \sum_{k=1}^{M} \frac{-1}{|x_j - X_k|}}_{\text{electron-proton}} + \underbrace{\sum_{1 \leq j_1 < j_2 \leq N} \frac{1}{|x_{j_1} - x_{j_2}|}}_{\text{electron-electron}}.$$

Solving the problem

- We have an ODE:

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- Compute ∇V explicitly, or with autodifferentiation
- Solve the equation with RK4, or a symplectic integrator
- Add external electro-magnetic field !

Thank you for your attention !

Professor Schrödinger believes things are too easy

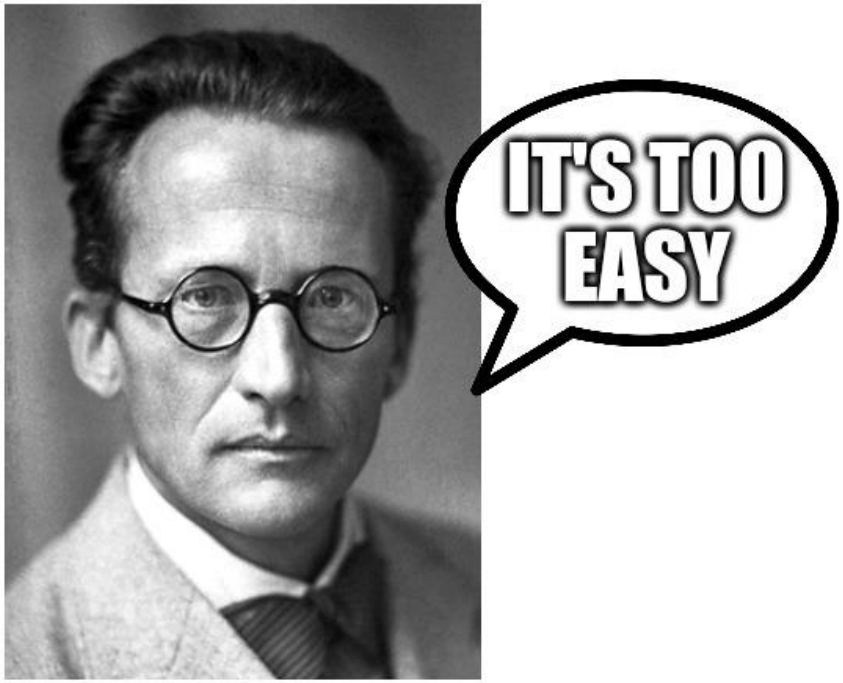

Figure: Erwin Schrödinger in 1933 (Wikipedia)

The Schrödinger equation

- Electrons are not "point" particles, they are *waves*
- Electronic systems are described by a *wave function*
  $\psi : (t, x_1, ..., x_N) \in \mathbb{R} \times \mathbb{R}^3 \times ... \times \mathbb{R}^3 \mapsto \mathbb{C}$

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- $|\psi|^2$ represents the *density of presence* of the electrons in the space of all possible configurations ($\int |\psi(t, x_1, ..., x_N)|^2 \, dx_1 ... dx_N = 1$)
- The movement of the electrons is ruled by the *Schrödinger equation*:

$$\begin{cases} i\partial_t \psi = -\Delta\psi + V\psi, \\ \psi(t=0) = \psi_0 \in L^2\left(\mathbb{R}^{3N}\right), \end{cases}$$

Pros	**Cons**
Linear	Curse of dimensionality
Mathematically well posed	Ubounded domain

Well-posedness

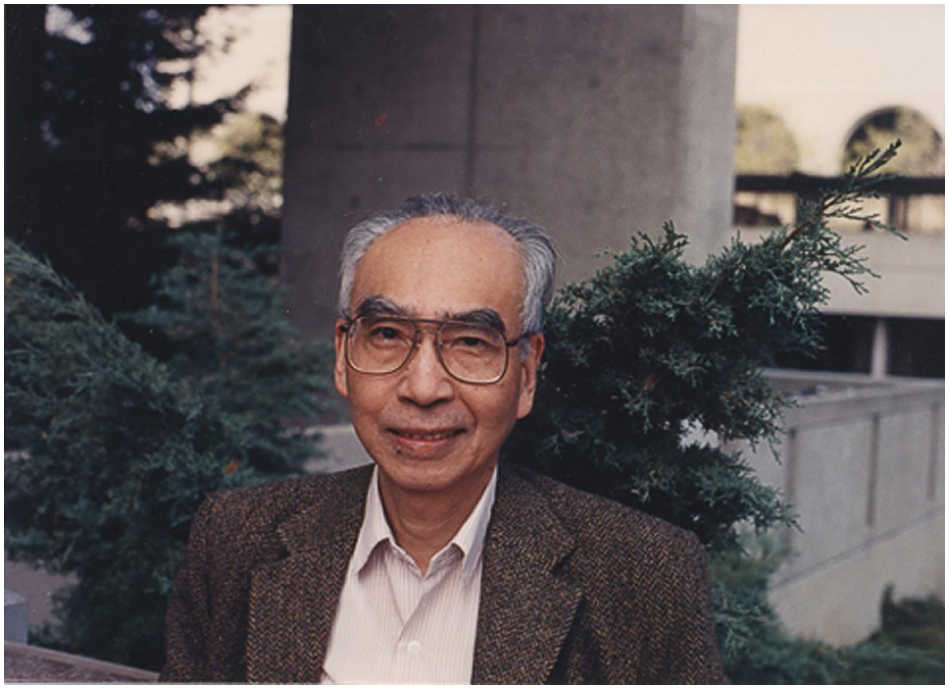
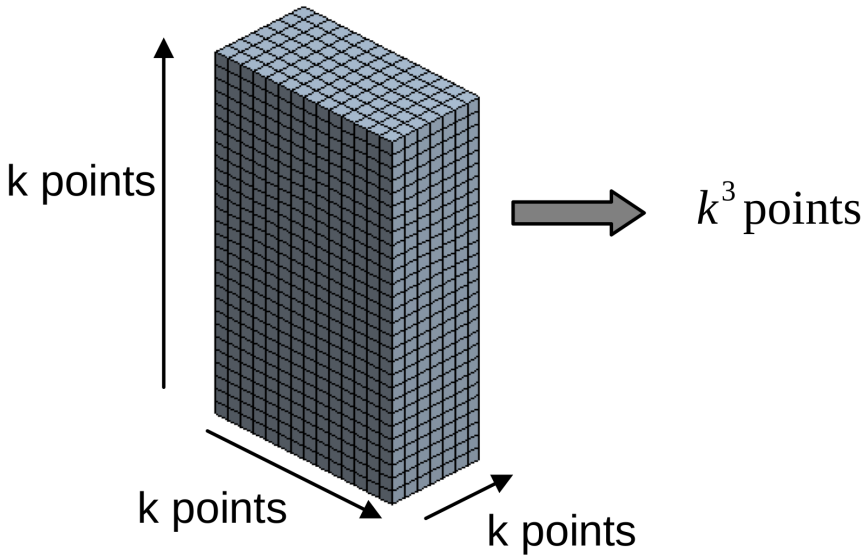


Figure: Tosio Kato (Wikipedia)

- Proved the Kato-Rellich theorem
- Proved that $-\Delta + V$ is self-adjoint $\Rightarrow$ Schrödinger equation is well posed whenever $\psi_0 \in L^2\left(\mathbb{R}^{3N}\right)$.

The curse of dimensionality

"Grid-type" approach with 1 electron:



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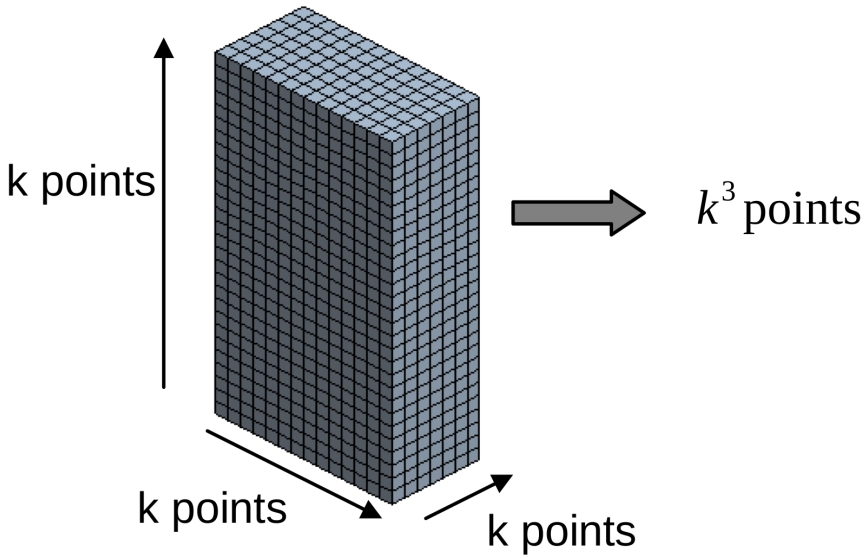

"Grid-type" approach with N electrons:

$\psi(t, x_1, ..., x_N) \Rightarrow k^{3N}$ points

Ex: $k = 10$, $N = 10 \Rightarrow 10^{30}$ *points*

$=$ ***Too expensive !***

Variational formulation for the Schrödinger equation

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- We "twist" the equation: Define $\varphi(t) = \mathrm{e}^{-it\Delta}\,\psi(t)$, then φ is solution to

$$\begin{cases} i\partial_t \varphi = \mathrm{e}^{-it\Delta}\, V\, \mathrm{e}^{it\Delta}\, \varphi, \\ \varphi(0) = \psi_0. \end{cases}$$

(The operator $\mathrm{e}^{-it\Delta}\, V\, \mathrm{e}^{it\Delta}$ follows the *Heisenberg picture*)

Variational formulation for the Schrödinger equation

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Theorem (Dupuy, Ehrlacher, Guillot)

Let

$$F(u) = \|u(0) - \psi_0\|_{L^2}^2 + T \left\| i\partial_t u - \mathrm{e}^{-it\Delta}\, V\, \mathrm{e}^{it\Delta}\, u \right\|_{L^2(I,L^2)}^2 .$$

If V satisfies the condition (C), then (1) has a unique solution φ in $H^1_{loc}(\mathbb{R}, L^2(\mathbb{R}^{3N}))$. For any bounded time interval $I = (0, T)$, φ there exist constants $\alpha, C > 0$ such that

$$\alpha \|u - \varphi\|_{H^1(I,L^2)}^2 \leq F(u) \leq C \|u - \varphi\|_{H^1(I,L^2)}^2 ,$$

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- Condition (C): There exists an $\varepsilon > 0$ such that

$$\sup_{\lambda \in \mathbb{R}} \left\| V(-\Delta + i\varepsilon + \lambda)^{-1} \right\| < 1.$$

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What is it for:

- Now (1) is equivalent to

$$\operatorname*{argmin}_{u \in H^1(I,L^2)} F(u)$$

- Computing $F(u)$ gives an a posteriori error bound

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Remarks:

- The constants α, C depends on T polynomially.
- The potential mentioned earlier (Coulomb and bounded) satisfy condition (C).

Variational formulation for the Schrödinger equation

Hint of proof:

We know that

$$\int_0^\infty \mathrm{e}^{-t(ix+\varepsilon)}\,\mathrm{e}^{-it\xi}\;dt \;=\; \frac{-i}{x+(\xi-i\varepsilon)}.$$

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Then we have Plancherel's indentity:

$$\int_0^\infty \mathrm{e}^{-2\varepsilon t}\left\|V\,\mathrm{e}^{it\Delta}\,\varphi\right\|^2 dt = \int_{\mathbb{R}} \left\|V(-\Delta+(\xi-i\varepsilon))^{-1}\varphi\right\|^2 d\xi$$

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Doing the same for $\int_{-\infty}^0$, we get

$$\int_{\mathbb{R}} \mathrm{e}^{-2\varepsilon|t|}\left\|V\,\mathrm{e}^{it\Delta}\,\varphi\right\|^2 dt = \int_{\mathbb{R}} \left\|V(-\Delta+(\xi-i\varepsilon))^{-1}\varphi\right\|^2 + \left\|V(-\Delta+(\xi+i\varepsilon))^{-1}\varphi\right\|^2$$

Example: Gaussian Wave Packets

- We consider the set of *gaussian wave packets*:

$$\mathcal{G} = \left\{ g : \mathbb{R}^d \to \mathbb{C} \ : \quad \begin{array}{c} \forall x \in \mathbb{R}^d,\ g(x) = \lambda\, \mathrm{e}^{-\frac{1}{2}(x-q)\cdot Q(x-q)}\, \mathrm{e}^{ip\cdot(x-q)}, \\ \lambda \in \mathbb{C},\ p, q \in \mathbb{R}^d,\ Q = A + iB \ \text{ with } A \in \mathcal{S}_d^{+,*} B \in \mathcal{S}_d \end{array} \right\}$$

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- $\mathcal{O}\left(d^2\right)$ parameters (not exponential)
- Computations are often explicit
- $\mathrm{e}^{it\Delta}(\mathcal{G}) = \mathcal{G}$
- $\mathcal{G}$ is a weakly close subset of $L^2\left(\mathbb{R}^d\right)$ (useful to prove the existence of minimizers)

Optimization process: Greedy algorithm

- We want to approximate the solution of

$$\underset{u \in H^1(I,L^2)}{\operatorname{argmin}} \|u(0) - \psi_0\|_{L^2}^2 + T \left\| i\partial_t u - \mathrm{e}^{-it\Delta} V \mathrm{e}^{it\Delta} u \right\|_{L^2(I,L^2)}^2,$$

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- Problem: No reason why the real minimizer should be in (or even close to) $H^1(I, \mathcal{G})$

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- Solution: We use more than one elements, and a *greedy algorithm*:

  Compute $g_1 \leftarrow \operatorname{argmin}_{g\in H^1(I,\mathcal{G})} F(g)$
  Compute $g_2 \leftarrow \operatorname{argmin}_{g\in H^1(I,\mathcal{G})} F(g_1+g)$
  Compute $g_3 \leftarrow \operatorname{argmin}_{G\in H^1(I,\mathcal{G})} F(g_1+g_2+g)$
  ...
  Compute $g_k \leftarrow \operatorname{argmin}_{g\in H^1(I,\mathcal{G})} F(g_1+\ldots+g_{k-1}+g)$
  ...

Optimization process: Greedy algorithm

We consider the greedy algorithm:

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...

Compute $g_k \leftarrow \text{argmin}_{g \in H^1(I,\mathcal{G})} F(g_1 + ... + g_{k-1} + g)$

...

Proposition

If $\mathcal{G}$ is the set of gaussian wave packets described above, the sequence defined by $h_k = g_1 + ... + g_k$ converges in $L^2(\mathbb{R}^{3N})$ to the minimizer of F.

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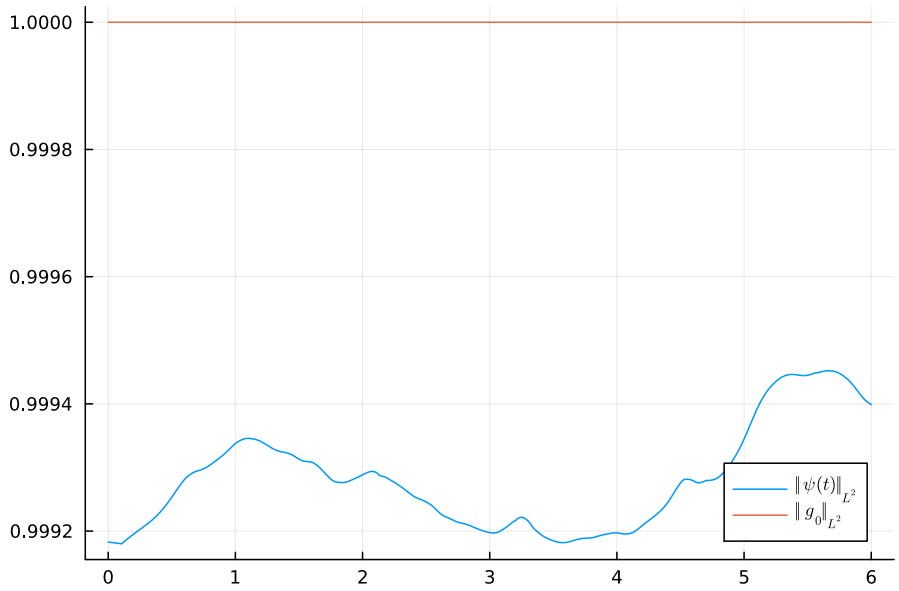
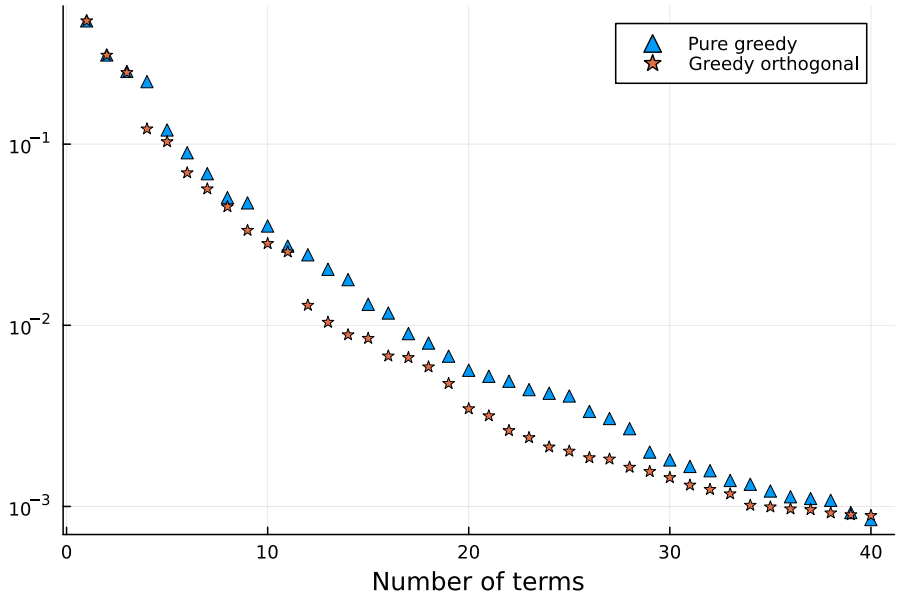


Figure: L^2 norm of the computed wave function with respect to t.



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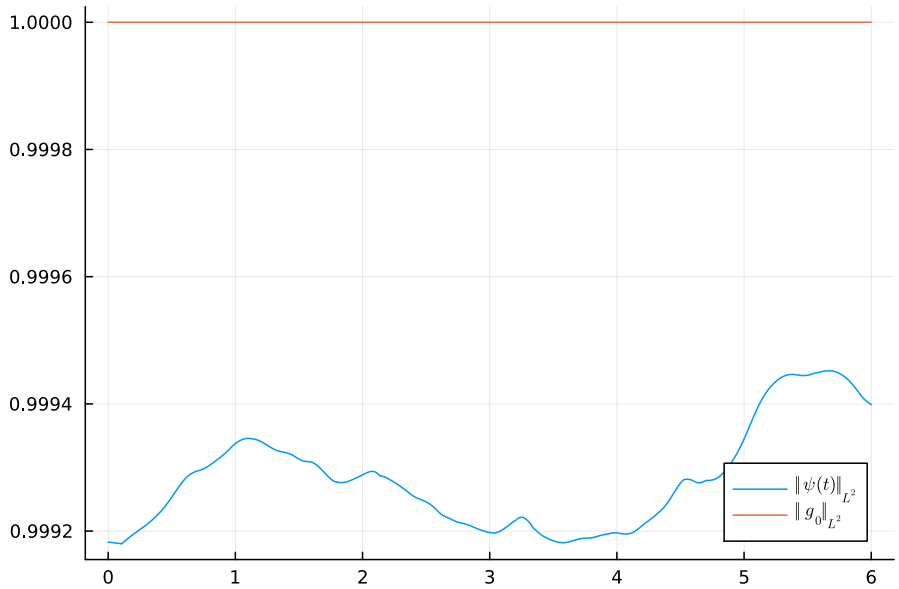

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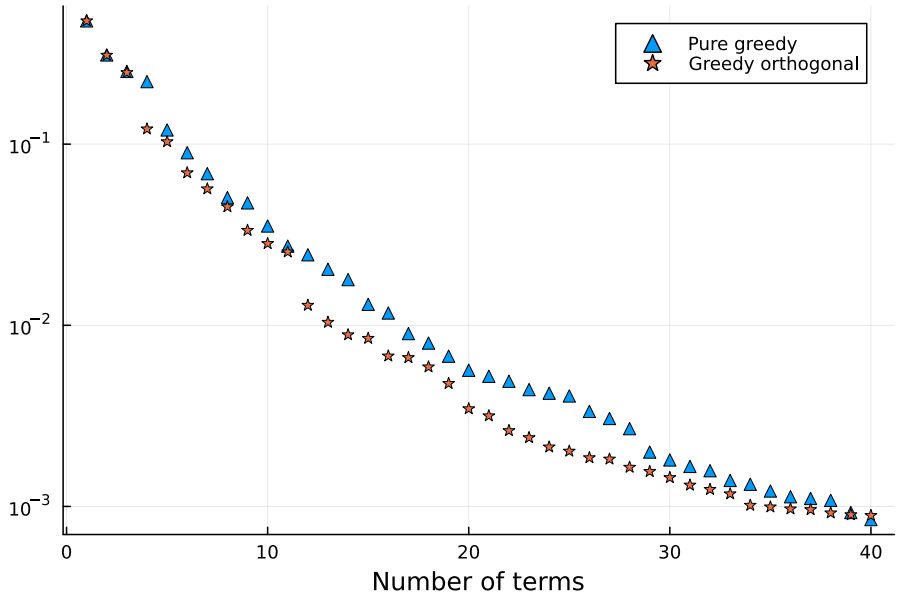

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Pros	**Cons**
Cheap parametrization	No proper initial guess
Sparsity	Sometimes takes a long time
Parallel in time	to converge

Alternative to space-time: The Dirac-Frenkel principle

- An alternative is to rely on the *Dirac-Frenkel variational principle*
- Define $\Sigma = \{g_1 + g_2 + ... + g_I \, : \, g_1, ..., g_I \in \mathcal{G}\}$ $(I \in \mathbb{N})$, and solve

$$\begin{cases} i\partial_t \varphi(t) = \pi_{T_{\varphi(t)}\Sigma} \left(\mathrm{e}^{-it\Delta} \, V \, \mathrm{e}^{it\Delta} \, \varphi \right), \\ \varphi(0) = \tilde{\psi}_0 \in \Sigma. \end{cases}$$

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- Advantage: Easy to implement, cheaper
- Problem: The Gram matrix of the g_j's may become ill-conditioned, leading to numerical instabilities

Conclusions and perspectives

- Fixing convergence issues
- Implementing a serious 3D example (Hydrogen...)
- Consider antisymmetric wave function ansatz

Thank you for your attention !