Mathematical analysis of the Multi–Configuration Time Dependent Hartree–Fock equations for Coulomb Systems (MCTDHF)

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Physical context

▶ Multi-electrons (≤ 10) dynamics of a molecule interacting with intense laser field ($I \simeq 10^{12} W\, cm^{-2}$)

▶ $M \geq 1$ nuclei located at $R_1, \cdots, R_M \in \mathbb{R}^3$ with charge $z_1, \cdots, z_M > 0$, creating an electrostatic potential

$$V(x) := -\sum_{m=1}^{M} \frac{z_m}{|x - R_m|}$$

▶ N electrons of charge -1 a.u. represented quantum mechanically by a wave function $\Psi = \Psi(x_1, \ldots, x_N) \in L^2(\mathbb{R}^{3N}; \mathbb{C})$ normalized by $\int_{\mathbb{R}^{3N}} |\Psi|^2\, dx_1 \cdots dx_N = 1$

$|\Psi(x_1, ..., x_N)|^2 dx_1 \cdots dx_N$ is the density of probability for finding the N electrons at $x_1, ..., x_N$

Physical context

- **Indiscernability / Pauli principle for fermions** :

Ψ is antisymmetric, $\Psi \in L^2_{\rm as}(\mathbb{R}^{3N};\mathbb{C}) = \bigwedge_1^N L^2(\mathbb{R}^3;\mathbb{C})$

$$\Psi(x_1,\dots,x_i,\dots,x_j,\dots x_N) = -\Psi(x_1,\dots,x_j,\dots,x_i,\dots x_N)$$

- **Coulomb Hamiltonian for N electrons** :

$$H_N = \sum_{i=1}^N \left(-\frac{1}{2}\Delta_{x_i} + V(x_i)\right) + \sum_{1\le i<j\le N} \frac{1}{|x_i - x_j|}$$

$$H_N(t) = \sum_{i=1}^N \frac{1}{2}\big(-\nabla_{x_i} + A(t)\big)^2 + V(x_i) + \sum_{1\le i<j\le N} \frac{1}{|x_i - x_j|} + V_{\rm ext}(t)$$

H_N is self-adjoint on $L^2\left(\mathbb{R}^{3N}\right)$ with domain $H^2\left(\mathbb{R}^{3N}\right)$ and form-domain $H^1\left(\mathbb{R}^{3N}\right)$ [Kato, 1951]

- **Energy of the system** : $\mathcal{E}(\Psi) = \langle H_N\Psi, \Psi\rangle_{L^2(\mathbb{R}^{3N};\mathbb{C})}$

The Time-Dependent Schrödinger equation (TDSE)

The dynamics is governed by the linear TDSE

$$i\,\frac{\partial\,\Psi}{\partial t} = H_N\Psi, \quad \Psi(0) = \Psi^0, \quad t > 0$$

$$\Psi = \Psi(t, x_1, \ldots, x_N)$$

Global existence of a unique solution is known (Stone's theorem for self-adjoint operators)

- Continuous one-parameter unitary group : $\Psi(t) = e^{-i\,t\,H_N}\,\Psi^0$
- $\Psi^0 \in H^m(\mathbb{R}^{3N}) \;\Rightarrow\; \Psi(t) \in C^0\big(\mathbb{R}^+; H^m(\mathbb{R}^{3N})\big),\ m = 0, 1, 2$
- $\|\Psi^0\|_{L^2(\mathbb{R}^{3N})} = 1 \;\Rightarrow\; \|\Psi(t)\|_{L^2(\mathbb{R}^{3N})} = 1$ for all $t > 0$
- H_N self-adjoint and independent of time $\Longrightarrow$ energy is conserved : for all $t > 0$, $\mathcal{E}(\Psi(t)) := \langle H_N\Psi, \Psi\rangle = \mathcal{E}(\Psi(0))$

Time-Dependent Schrödinger equation (TDSE)

▶ The knowledge of the wavefunction gives all chemical properties of the molecule

▶TDSE is numerically untractable for more than two electrons

Example : For a molecule of water H_2O, $N = 10 = 8 + 2$. The reference space is $L^2(\mathbb{R}^{30})$. Discretization with $100 \times 100 \times 100$ grid points leads to vectors with 10^{60} components and $10^{60} \times 10^{60}$ matrices...

For dealing with atoms involving many electrons the accurate quantum theory, involving a solution of the wave equation in many-dimensional space, is far too complicated to be practical. One must therefore to approximate methods. [P.A.M. Dirac, 1930]

✘ Time-Dependent Hartree-Fock (TDHF)

✔ Multi-Configuration Time-Dependent Hartree-Fock hierarchy (MCTDHF)

The Hartree-Fock model (TDHF)

- Determinants are the simplest possible antisymmetric functions
- **Slater determinants** (or configurations or HF states) :

Let $\Phi = \big(\phi_1, \cdots, \phi_N\big) \in L^2(\mathbb{R}^3)^N$ with $\quad \int_{\mathbb{R}^3} \phi_i \, \overline{\phi}_j dx = \delta_{i,j}$,

$$\Psi_{\rm HF}(x_1, ..., x_N) = \frac{1}{\sqrt{N!}} \det\big(\phi_i(x_j)\big)_{1\leq i,j\leq N} := \phi_1 \wedge \cdots \wedge \phi_N$$

For $N = 2$, $(\phi_1 \wedge \phi_2)(x_1, x_2) = \frac{1}{\sqrt{2}}\Big(\phi_1(x_1)\phi_2(x_2) - \phi_2(x_1)\phi_1(x_2)\Big)$

Antisymmetrized product of N one-particle wavefunctions (or orbitals) ϕ_i

$\Psi_{\rm HF}$ is antisymmetric and $\|\Psi_{\rm HF}\|_{L^2(\mathbb{R}^{3N})} = 1$

- If the initial data Ψ^0 is a Slater determinant in TDSE, $\Psi(t)$ is not a Slater determinant at any time $t > 0$ because of the electron-electron interaction

$\Longrightarrow$ Force the evolution on the set of Slater determinants

The Hartree-Fock model (TDHF)

▶ Two unitary equivalent vectors Φ and $\Phi' = U\,\Phi$ with $U \in \mathbf{U}(N)$ give the same wavefunction $\Psi_{\rm HF}$ (and conversely). Up to a unitary transform, the Hartree–Fock system of equations reads: for every $1 \leq i \leq N$

$$i\frac{\partial}{\partial t}\phi_i = -\Delta\,\phi_i + V(x)\phi_i + \left(\sum_{j=1}^{N}|\phi_j|^2 \star \frac{1}{|x|}\right)\phi_i - \sum_{j=1}^{N}\left(\phi_i\overline{\phi}_j \star \frac{1}{|x|}\right)\phi_j$$

▶ Coupled system of $\mathbf{N}$ non-linear time-dependent PDEs of Schrödinger type

▶ Global-in-time well-posedness : existence, uniqueness, continuity w.r.t. initial data in the energy space $H^1(\mathbb{R}^3)^N$ and in $H^2(\mathbb{R}^3)^N$ [Chadam-Glassey, 1975]

✔ Numerical simulations are very powerful

✘ Does not fit with experimental datas for strong laser fields

Beyond Hartree-Fock approximation

▶ **Slater determinants span $\bigwedge_1^N \mathbf{L^2}(\mathbb{R}^3)$:** If $(\phi_i)_{i\geq 1}$ is an orthonormal basis of $L^2(\mathbb{R}^3)$, the family of all possible Slater determinants from this basis is an orthonormal basis of $\bigwedge_1^N L^2(\mathbb{R}^3)$. Any $\Psi \in \bigwedge_1^N L^2(\mathbb{R}^3)$ writes

$$\Psi = \sum_{1\leq i_1<i_2<\cdots<i_N} c_{i_1,\ldots,i_N}\, \phi_{i_1} \wedge \cdots \wedge \phi_{i_N}, \qquad \sum_{1\leq i_1<i_2<\cdots<i_N} |c_{i_1,\ldots,i_N}|^2 = \|\Psi\|^2$$

▶ **MC approximation :** Restrict to a finite number of Slater determinants or configurations. Let $K \geq N$ and

$$\Sigma_N^K = \Big\{\sigma : \{1,2,\ldots,N\} \longrightarrow \{1,\ldots,K\} \mid \sigma(1) < \ldots < \sigma(N)\Big\}$$

$$\mathcal{S}_{L^2(\mathbb{R}^3)^K} = \Big\{\Phi = (\phi_1,\ldots,\phi_K) \in L^2(\mathbb{R}^3)^K \mid \int_{\mathbb{R}^3} \phi_i\, \overline{\phi}_j\, dx = \delta_{i,j}\Big\}$$

$\#\Sigma_N^K = \binom{K}{N} := M$ number of all possible determinants

The multi-configuration ansatz

$$\Phi_\sigma(x_1,\dots,x_N)=\phi_{\sigma(1)}\wedge\cdots\wedge\phi_{\sigma(N)},\ \int_{\mathbb{R}^{3N}}\Phi_\sigma\,\overline{\Phi}_\tau=\delta_{\sigma,\tau}$$

$$\begin{array}{rcccl}\pi & : & S^{M-1}\times \mathcal{S}_{L^2(\mathbb{R}^3)^K} & \longrightarrow & \bigwedge_1^N L^2(\mathbb{R}^3)\\ & & (C,\Phi) & \longmapsto & \Psi_{\rm MC}:=\pi(C,\Phi)=\sum_{\sigma\in\Sigma_N^K} c_\sigma\,\Phi_\sigma\end{array}$$

with $\quad S^{M-1}=\left\{C=(c_\sigma)\in\mathbb{C}^M\ :\ \|C\|^2=\sum_{\sigma\in\Sigma_N^K}|c_\sigma|^2=1\right\}$

For $N=2$ and $K=4$, $M=6$, $\quad\Phi=(\phi_1,\phi_2,\phi_3,\phi_4)$, $C=(c_1,c_2,c_3,c_4,c_5,c_6)\in\mathbb{C}^6$

$$\pi(C,\Phi)=c_1\,\underbrace{\phi_1\wedge\phi_2}+c_2\,\underbrace{\phi_1\wedge\phi_3}+c_3\,\underbrace{\phi_1\wedge\phi_4}+c_4\,\underbrace{\phi_2\wedge\phi_3}+c_5\,\underbrace{\phi_2\wedge\phi_4}+c_6\,\underbrace{\phi_3\wedge\phi_4}$$

- the variables (C,Φ) lie on the $\mathcal{F}_N^K:=S^{M-1}\times\mathcal{S}_{L^2(\mathbb{R}^3)^K}$ (fibers)
- the corresponding wavefunction $\Psi=\pi(C,\Phi)$ lies on a subset $\mathcal{B}_N^K:=\left\{\pi(C,\Phi)\ :\ (C,\Phi)\in\mathcal{F}_N^K\right\}$ of $\left\{\Psi\in\bigwedge_1^N L^2(\mathbb{R}^3)\ :\ \|\Psi\|=1\right\}$(basis)

The multi-configuration ansatz

Multiconfiguration bridges the gap between HF states and general states.

- For $K = N$, $\quad \Sigma_N^K = \left\{ Id_{\{1,\cdots,N\}} \right\}$, $\quad \#\Sigma_N^K = 1$: single determinant states or Hartree-Fock approximation
- If $N \leq K \leq K'$,

$$\underbrace{\mathcal{B}_N^N}_{\text{HF states}} \subset \cdots \subset \mathcal{B}_N^K \subset \mathcal{B}_N^{K'} \subset \cdots \subset \underbrace{\left\{ \Psi \in \bigwedge_1^N L^2(\mathbb{R}^3) \ : \ \|\Psi\| = 1 \right\}}_{K \to +\infty}$$

- We now allow for time-dependent coefficients and orbitals ($\neq$ Galerkin approx.)

$$\Psi(t,x) = \sum_{1 \leq \sigma \leq M} c_\sigma(t)\, \Phi_\sigma(t,x)$$

and we want $\Psi(t)$ to evolve in time on $\mathcal{B}_N^K \Longrightarrow$ Structure of $\mathcal{B}_N^K$?

Two main issues

① **How to lift a path on $\mathcal{B}_N^K$ to a path on $\mathcal{F}_N^K$ in a smooth way ?**

As for HF states, different (C, Φ) define the same wavefunction.

Given $U \in \mathbf{U}(N)$, define $d(U) \in \mathbf{U}(M)$ by $d(U)^*_{\sigma,\tau} =$ det $(U_{\sigma(i),\tau(j)})$, if $\Phi = U \cdot \Phi'$ and $C = d(U) \cdot C'$ then $\Psi = \pi\big(C, \Phi\big) = \pi\big(C', \Phi'\big)$.

The fiber at Ψ $\pi^{-1}(\Psi)$ does not always have group orbit structure $\neq$ HF !

A given Slater determinant $\phi_1 \wedge \phi_2$ belongs to all approximation sets $\mathcal{B}_2^K$. If $K \geq 4$, the map π is not an homeomorphism in a neighborhood of $\phi_1 \wedge \phi_2$: for any orbitals $(\xi, \eta) \in \mathrm{Span}\{\phi_1, \phi_2\}^\perp$, $\phi_1 \wedge \phi_2 = \pi\big((1,0,0,0,0,0), (\phi_1, \phi_2, \xi, \eta)\big)$

➥ Under full-rank assumption, $\mathcal{B}_N^K$ is a smooth manifold (fiber bundle)

② **How to define evolution equations that preserve $\mathcal{B}_N^K$?**

TDSE does not preserve $\mathcal{B}_N^K$ because of electron-electron interactions:

$\Psi(t=0) \in \mathcal{B}_N^K \;\Rightarrow\; e^{-i\,t\,H_N}\,\Psi(t=0) \notin \mathcal{B}_N^K$ as soon as $t > 0$

➥ Dirac–Frenkel variational principle

$\mathcal{B}_N^K$ and finite rank density matrices

▶ **One-particle density matrices**: to $\Psi \in \bigwedge_1^N L^2(\mathbb{R}^3)$ one associates the self-adjoint operator γ_Ψ on $L^2(\mathbb{R}^3;\mathbb{C})$ with finite trace and kernel

$$\gamma_\Psi(x,y) = N \int_{\mathbb{R}^{3(N-1)}} \Psi(x,x_2,\dots,x_N)\overline{\Psi}(y,x_2,\dots,x_N)\,dx_2\dots dx_N$$

$$0 \leq \gamma_\Psi \leq \mathrm{Id}, \quad \mathrm{Tr}\gamma_\Psi = N$$

For $\Psi = \pi(C,\Phi) \in \mathcal{B}_N^K$, $\quad \gamma_\Psi(x,y) = \sum_{i,j=1}^{K} \gamma_{ij}\,\phi_i(x)\,\phi_j^*(y)$

then $\mathrm{Ran}(\gamma_\Psi) \subset \mathrm{Span}\{\phi_1,\cdots,\phi_K\}$, $\quad N \leq \mathrm{dimRan}\,\gamma_\Psi = \mathrm{rank}(\gamma_{ij}) \leq K$,
and conversely

▶ $\Psi \in \mathcal{B}_N^K$ (is a sum of at most $\binom{K}{N}$ determinants) $\Longleftrightarrow$ its one-particle density matrix is of finite rank $\leq K$ [Löwdin, 1955]

$\mathcal{B}_N^K$ and finite rank density matrices

- **For HF states** $\gamma_{\Psi_{\rm HF}}(x,y) = \sum_{i=1}^{N} \phi_i(x)\,\phi_i^*(y)$ is a projector of rank N

on Span$\{\phi_1, \cdots, \phi_N\}$. Note $(\gamma_{ij}) = I_N$

- **Full-rank hypothesis :**

$$d\mathcal{F}_N^K = \Big\{(C,\Phi) \in \mathcal{F}_N^K \ : \ \mathrm{rank}\gamma_{\pi(C,\Phi)} = K\Big\}, \quad d\mathcal{B}_N^K := \pi\big(d\mathcal{F}_N^K\big)$$

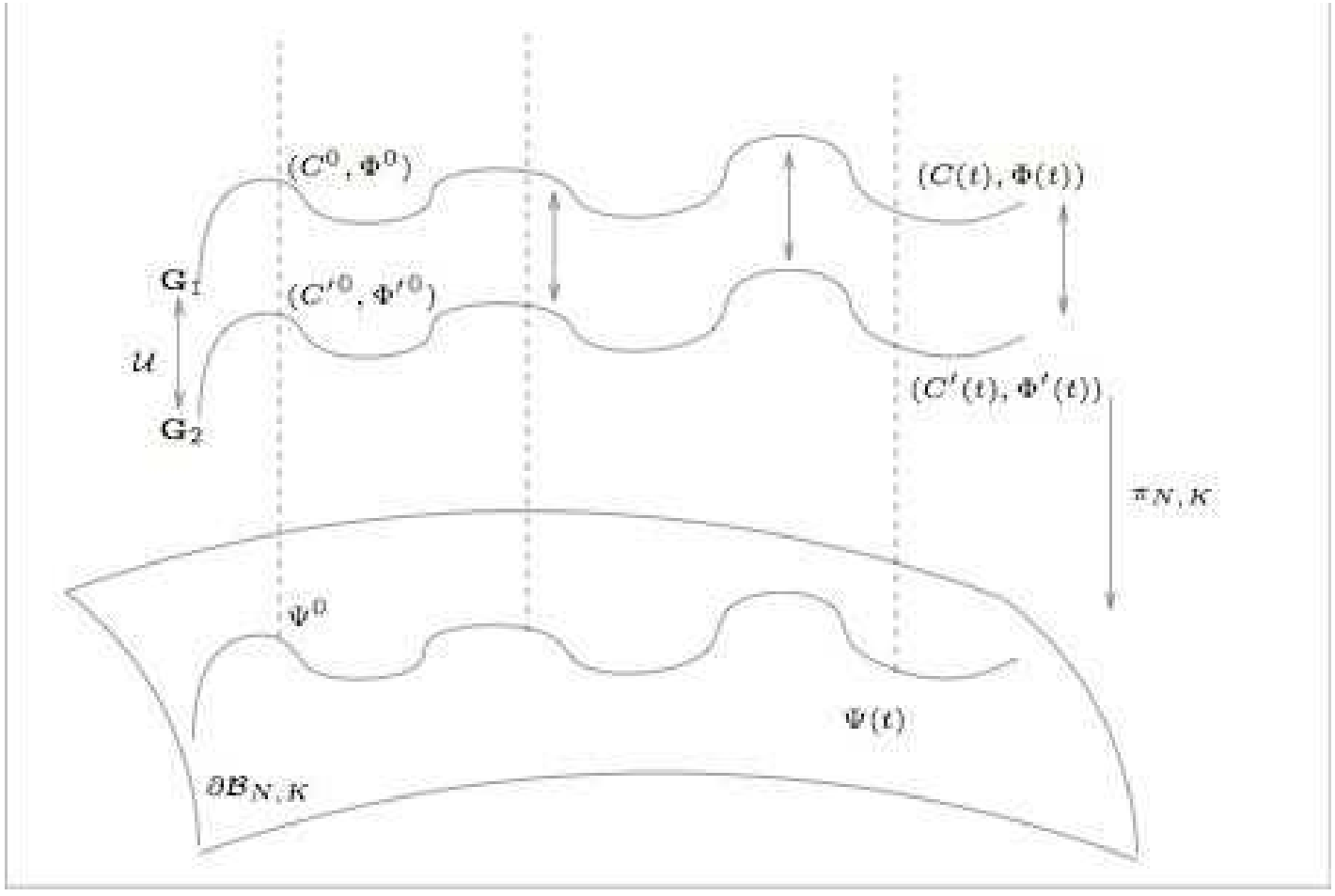
Given $\Psi = \pi\big(C,\Phi\big) = \pi\big(C',\Phi'\big)$ in $d\mathcal{B}_N^K$ there exists a unique couple of unitary transforms $(U, d(U)) \in \mathbf{U}(N) \times \mathbf{U}(M)$ such that

$$\Phi = U \cdot \Phi', \quad C = d(U) \cdot C'$$

π is a C^1 diffeomosphism on $d\mathcal{F}_N^K$.

Transitive unitary group action $\mathcal{G} \Rightarrow$ Fiber bundle : $d\mathcal{B}_N^K = d\mathcal{F}_N^K/\mathcal{G}$

Fiber Bundle Structure : summary



The Dirac-Frenkel Variational Principle

The **Dirac–Frenkel variational principle** forces the wavefunction $\Psi_{\rm MC}$ to evolve on $\mathrm{d}\mathcal{B}_N^K$ through [Dirac, 1930; Frenkel,1934; Lubich, 2004]

$$\frac{\partial \Psi}{\partial t} = \text{argmin}\left\{ \| -iH_N\,\Psi - \Theta\|_{L^2(0,T;L^2(\mathbb{R}^{3N}))} \ : \ \Theta \in \mathcal{T}_\Psi dB_N^K \right\}$$

or equivalently [McLachlan, 1964] $\left\langle i\,\dfrac{\partial}{\partial t}\Psi_{\rm MC} - H_N\Psi_{\rm MC}\,,\,\delta\Psi \right\rangle = 0$ for variations $\delta\Psi$ in $\mathcal{T}_\Psi dB_N^K$, the tangent space to the manifold $\mathrm{d}\mathcal{B}_N^K$ at $\Psi_{\rm MC}$

- With $\delta\Psi = i\,\Psi_{\rm MC}$ and H_N self-adjoint
  $\Rightarrow$ Orthonormality of orbitals and normalization of coefficients propagate
- With $\delta\Psi = \frac{\partial}{\partial t}\Psi_{\rm MC}$ and H_N independent of t
  $\Rightarrow \dfrac{d}{dt}\langle H_N\Psi_{\rm MC}, \Psi_{\rm MC}\rangle = 0 \Rightarrow$ Energy is kept constant by the flow

The MCTDHF equations

There are obtained by plugging the MC ansatz in the variational principle.

▶ **Under full rank assumption** a C^1-path $t \mapsto \Psi(t)$ on the basis $\mathrm{d}\mathcal{B}_N^K$ may be lifted to equivalent paths on expansion coefficients and orbitals $(C(t), \Phi(t)) \in \pi^{-1}(\Psi(t))$ up to time-dependent unitary transforms

▶ C^1-flow on unitary transforms is equivalent to a choice of a gauge i.e. of an arbitrary self-adjoint operator G on $L^2(\mathbb{R}^3)$ that demands

$$\forall\, t > 0,\ i \left\langle \frac{\partial}{\partial t}\phi_i, \phi_j \right\rangle = \left\langle \phi_i \middle| G \middle| \phi_j \right\rangle \qquad \text{for all } 1 \leq i \leq j \leq K,$$

A choice for $\mathbf{G}$ fixes a choice of lifting on $\mathrm{d}\mathcal{F}_N^K$. We focus on two choices

- Variational Equations $G = 0$: *Ad hoc* for Variational principle/Propagation of constraints/Energy conservation
- Working Equations $G = -\Delta + V$: *Ad hoc* for mathematical proofs of well-posedness of Cauchy problem

Variational equations

The Cauchy problem associated to the MCTDHF

$$
\begin{aligned}
& i\,\frac{dC}{dt} = \nabla_{\bar{C}} \big\langle H_N\,\Psi \,|\, \Psi \big\rangle = \sum_{\sigma,\tau} \langle H_N\,\Phi_\sigma \mid \Phi_\tau \rangle\, c_\tau \\
& i\,\mathbf{\Gamma}(t)\,\frac{\partial \Phi}{\partial t} = (I - \mathbf{P}_\Phi)\,\nabla_{\bar{\Phi}} \big\langle H_N\,\Psi \,|\, \Psi \big\rangle \\
& \big(C(t=0), \Phi(t=0)\big) = \big(C_0, \Phi_0\big) \;\in\; \mathrm{d}\mathcal{F}_N^K
\end{aligned}
$$

- $\mathbf{P}_\Phi(\cdot) = \sum_{i=1}^K \big\langle \cdot\,,\, \phi_i \big\rangle_{L^2(\mathbb{R}^3)}\, \phi_i$ projector on $\mathrm{Span}\{\phi\}_{i=1}^K$
- $\mathbf{\Gamma}(t) = (\gamma_{ij}(t))$: $K \times K$ Hermitian matrix (representation of the 1-pdm γ_Ψ in the vector space $\mathrm{Span}\{\phi\}_{i=1}^K$)

ODE's for the coefficients looks like Galerkin approximation but it is NOT since the orbitals are time-dependent

Variational equations

- By explicit calculations : $\mathbb{\Gamma}(t) = \mathbb{\Gamma}(C(t))$ only depends on C, and in a continuous manner

$\Rightarrow$ if $\mathbb{\Gamma}(C)$ is invertible at start, it remains so on small time interval

- If $\big(C(t), \Phi(t)\big)$ is a strong solution with $\mathbb{\Gamma}(t)$ invertible on $[0, T]$, then
  - $\Psi(t) = \pi\big(C(t), \Phi(t)\big)$ satisfies the Dirac–Frenkel variational principle
  - Orthonormality of orbitals and normalization of coefficients are preserved
  - Energy is conserved

- Mathematical analysis (existence, uniqueness, continuity w.r.t. initial data...) is performed on recasted equations in different gauge

Working equations

$$
\begin{aligned}
i\frac{dC}{dt} &= \mathbb{K}[\Phi]\, C, \\
i\,\mathbb{\Gamma}(t)\,\frac{\partial \Phi}{\partial t} &= \mathbb{\Gamma}(t)\,(-\mathbf{\Delta}+\mathbf{V})\,\Phi + (\mathbf{I}-\mathbf{P}_\Phi)\,\mathbb{W}[C,\Phi]\,\Phi, \\
\big(C(0),\Phi(0)\big) &= \big(C^0,\Phi^0\big) \in \mathrm{d}\mathcal{F}_N^K,
\end{aligned}
$$

- When $\mathbb{\Gamma}(t)$ invertible, $\quad i\left\langle \frac{\partial}{\partial t}\,\phi_i, \phi_j \right\rangle = \big\langle \phi_i \big| -\Delta + V \big| \phi_j \big\rangle$
- $\mathbb{K}[\Phi]$ and $\mathbb{W}[C,\Phi]$ are $M \times M$ and $K \times K$ resp. matrices
- Coupled system of $M = \binom{K}{N}$ linear ODEs for the expansion coefficients and K PDEs of Schrödinger type with non-local non-linearities

Working equations

$$\mathbb{K}[\Phi]_{\sigma,\tau} = \frac{1}{2} \sum_{\substack{i,j\in\tau,\ k,l\in\sigma \\ \tau\setminus\{i,j\}\equiv\sigma\setminus\{k,l\}}} (-1)^{\tau}_{i,j}\ (-1)^{\sigma}_{k,l} \int_{\mathbb{R}^6} \frac{\phi_i(x)\bar{\phi}_k(x)\phi_j(y)\,\bar{\phi}_l(y)}{|x-y|} dx\,dy$$

$$\mathbb{W}[C,\Phi]_{ij} = 2 \sum_{k,l=1}^{K} \gamma_{ijkl}(C)\ \overline{\phi}_k\ \phi_l \star \frac{1}{|x|}$$

γ_{ijkl} are the expansion coefficients of the 2-body density matrix in $\mathrm{Span}\{\phi_1,\cdots,\phi_K\}$ depend only on C and quadratically

Without electron-electron interaction, $\mathbb{K}=0$ and $\mathbb{W}=0$; then C is independent of time, and so does $\mathbb{\Gamma}$. The system reduces to

$$i\,\frac{\partial\Phi}{\partial t} = (-\Delta + V)\,\Phi$$

Well-posedness of working equations

THEOREM. [C. Bardos, I. C., N. Mauser, S. Trabelsi, ARMA, 2010]

Let $m \in \{1,2\}$ and $(C^0,\Phi^0) \in \mathcal{F}_N^K$ with $\Phi^0 \in H^m(\mathbb{R}^3)^K$ and $\mathbb{\Gamma}(C^0)$ invertible. Then, there exists a maximal existence time $T^\star > 0$ (possibly $+\infty$, independent of m) s.t. the MCTDHF equations admit a unique solution (C,Φ) with

$$C \in C^1\big([0,T^\star);\mathbb{C}^M\big),\ \Phi \in C^0\big([0,T^\star);H^m(\mathbb{R}^3)\big)\cap C^1\big([0,T^\star);H^{m-2}(\mathbb{R}^3)\big)^K$$

and that depends continuously on the initial data (C^0,Φ^0). On $[0,T^\star]$:

(i) $\big(C(t),\Phi(t)\big) \in \mathrm{d}\mathcal{F}_N^K$ and $\mathbb{\Gamma}\big(C(t)\big)$ is invertible.

(ii) Energy is conserved : $\mathcal{E}\Big(\pi(C(t),\Phi(t)\Big) = \mathcal{E}\Big(\pi(C(0),\Phi(0)\Big)$

(iii) Dirac–Frenkel variational principle is satisfied.

(iv) If $T^\star < +\infty$, $\limsup_{t\nearrow T^\star} \|\mathbb{\Gamma}(C(t))^{-1}\| = +\infty$: $\mathbb{\Gamma}(t)$ singular at $T^\star$

Strategy of proof

If $\mathbb{\Gamma}$ invertible (true at start, expected for t small)

$$\begin{cases} i\,\dfrac{dC(t)}{dt} = \mathbb{K}[\Phi]\,C \\ i\,\dfrac{\partial\Phi(t)}{\partial t} = (-\mathbf{\Delta}+\mathbf{V})\,\Phi(t) + \mathbb{\Gamma}\big(t\big)^{-1}\,(\mathbf{I}-\mathbf{P}_{\Phi})\mathbb{W}[C,\Phi]\,\Phi(t), \\ C(0)=C^0, \quad \Phi(0)=\Phi^0, \quad \mathbb{\Gamma}\big(C^0\big) \text{ invertible}\,. \end{cases}$$

In a neighborhood of (C^0,Φ^0) the non-linearities are locally bounded and locally Lipschitz in $\mathbb{C}^M\times\left(H^m(\mathbb{R}^3)^K\right)$ for $m=1$ or $=2$

$\Rightarrow$ local-in-time well-posedness for a "mild solution" in integral form in both spaces (Segal's Theorem) (as in [Chadam-Glassey, JMP, 1975] for TDHF)

Strategy of proof

- for H^2 initial data, equations are satisfied in strong sense.
- As long as $\mathbb{\Gamma}(t)$ invertible by unitary transforms the mild solution is also solution to the "variational system" :
  - Dirac–Frenkel principle is satisfied
  - $\mathcal{F}_N^K$ is preserved by the MCTDHF flow : $\|C(t)\| = 1$ and $\Phi(t)$ are orthonormal orbitals
  - Energy is conserved $\Longrightarrow$ the H^1-norm stays locally bounded (detailed below)
- By density of H^2 in H^1 and continuity with respect to initial data also true for H^1 initial data

Strategy of proof

With $\Psi(t) = \pi\big(C(t), \Phi(t)\big)$, the energy writes

$$\langle H_N \Psi(t)\,,\, \Psi(t)\rangle = \langle \mathbb{\Gamma}(t)\,(-\mathbf{\Delta} + V)\,\Phi(t)\,;\,\Phi(t)\rangle + \underbrace{\langle \mathbb{W}[C,\Phi]\,\Phi(t), \Phi(t)\rangle}_{\geq 0}$$

$$\geq \mathrm{cst}\,\|\sqrt{\mathbb{\Gamma}}\,\Phi(t)\|^2_{(H^1(\mathbb{R}^3))^K}$$

by Kato's inequality and positivity of electron-electron potential. If

$$\mathbb{\Gamma}(t) \sim \mathrm{diag}\big(\gamma_1(t), \cdots, \gamma_K(t)\big)$$

$$\Big(\min_{1\leq i\leq K} \gamma_i\Big) \sum_{i=1}^K \|\phi_i\|^2_{H^1} = \sum_{i=1}^K \gamma_i\,\|\phi_i\|^2_{H^1} \sim \|\sqrt{\mathbb{\Gamma}}\Phi(t)\|^2_{(H^1(\mathbb{R}^3))^K} \leq \mathcal{E}(\Psi^0)$$

Full-rank assumption implies $\gamma_1(t) = \min_{1\leq i\leq K}\gamma_i$ stays away from zero

$$\|\Phi(t)\|_{(H^1(\mathbb{R}^3))^K} \lesssim \|\mathbb{\Gamma}(t)^{-1}\|^{1/2} = \frac{1}{\sqrt{\gamma_1(t)}}$$

Strategy of proof

- The maximal time of existence is finite if the norm $\|\mathbb{\Gamma}(t)^{-1}\|$ explodes in finite time i.e. the smallest eigenvalue of $\mathbb{\Gamma}(t))$ tends to 0 when $t \to T^\star$. $\mathbb{\Gamma}(t)$ becomes singular at T^*
- **When $\mathbf{K} = \mathbf{N}$**, we recover Chadam and Glassey's results on TDHF as a special case. The matrix $\mathbb{\Gamma}(t)$ turns to be the identity, thus invertible, for all time. In that case, $T^\star = +\infty$. Solutions are global-in-time.
- A well-posedness result for MCTDH model (few interacting distinguishable nuclei) with $V \equiv 0$, bounded regular binary interaction, and initial data in H^2, was obtained in [Koch & Lubich, M2AN,2007] by different methods. Algebra for MCTDH is more difficult (no symmetry assumption), analysis is the same. Same limitative full-rank assumption.

Should one expect global-in-time solutions ? Any numerical evidence ?

First example of global-in-time solutions

- **Standing waves for TDSE :**

if $\lambda \in \mathbb{R}$ is an eigenvalue of H_N with corresponding eigenstate $\mathbf{\Psi}$, then $\Psi(t,x) = e^{-i\lambda\, t}\, \mathbf{\Psi}(x)$ is a solution of TDSE

➥ $\mathbf{\Psi}$ is a critical point of $\Big\langle H_N\, \Psi\, ,\, \Psi \Big\rangle$ under constraint $\|\Psi\| = 1$, with Lagrange multiplier λ

- Existence of infinitely many critical points of the energy functional $\Big\langle H_N\, \pi(C,\Phi)\, ,\, \pi(C,\Phi) \Big\rangle$ on $d\mathcal{F}_N^K$ [Le Bris,AIHP 94; Friesecke, ARMA 03; Lewin, ARMA 04]

Let $\mathbf{\Psi} = \pi(C,\Phi)$ be a critical point of with $\mathbb{\Gamma}(C)$ invertible

$$\nabla_{\bar{C}} \Big\langle H_N\, \mathbf{\Psi}\, ,\, \mathbf{\Psi} \Big\rangle = \lambda\, C$$

$$\nabla_{\bar{\Phi}} \Big\langle (-\Delta + V)\, \mathbf{\Psi}\, ,\, \mathbf{\Psi} \Big\rangle = \mathbb{\Gamma}(C)\, H_N\, \Phi + \mathbb{W}[C,\Phi]\, \Phi = \Lambda \cdot \Phi$$

with $\lambda \in \mathbb{R}$ and Λ Hermitian matrix.

First example of global-in-time solutions

Let $\big(U(t), d(U(t))\big) \in \mathbf{U}(M) \times \mathbf{U}(N)$ and

$$\tilde{C}(t) = d(U(t))e^{-i\lambda\, t}\, C, \quad \tilde{\Phi}(t) = U(t)\Phi$$

then

$$\Psi(t) = \pi(\tilde{C}(t), \tilde{\Phi}(t)) = e^{-i\lambda\, t}\, \pi(C, \Phi)$$

give standing wave solutions

$\Longrightarrow$ there exists infinitely many global solutions solutions with full-rank

Global well-posedness: A sufficient condition

For $K \geq N$ define $\quad I(K) = \inf_{(C,\Phi)\in\mathcal{F}_N^K} \mathcal{E}(\pi(C,\Phi)).$

$K \mapsto I(K)$ is non-increasing, $\quad I(N) = I^{HF}, \quad \lim_{K\to\infty} I(K) = \inf \sigma(H_N)$

THEOREM. [C. Bardos, I. C., N. Mauser, S. Trabelsi, ARMA, 2010]

Let the initial data $(C^0,\Phi^0) \in \mathcal{F}_N^K$ *with* $\mathbb{\Gamma}(C^0)$ *invertible.*
If $\mathcal{E}(\pi(C^0,\Phi^0)) < I(K-1)$ *then the solution is global in time.*

- It always holds $I_K \leq I_{K-1}$
- For positive ions ($\sum_{m=1}^M z_m \geq N$) $\quad I_K < I_{K-2}$ [Friesecke, ARMA, 2003]. Then such initial data exist either for K or $K-1$.
- First criteria in the literature for global-in-time existence for such multiconfiguration models

Idea of the proof

For $0 < t < T^\star$, use a choice of gauge $(C, \Phi) \mapsto (C', \Phi')$ that diagonalizes the operator $\mathbb{\Gamma}(t)$:

$$\pi(C, \Phi) = \pi(C', \Phi'), \quad \mathbb{\Gamma}(C') = U \cdot \mathbb{\Gamma}(C) \cdot U^* = \mathrm{diag}\Big(\gamma_1, \cdots, \gamma_K\Big)$$

with $0 \leq \gamma_1 \leq \cdots \leq \gamma_K \leq 1$. Assume :

$$\text{For } \ t_n \longrightarrow T^*, \qquad \lim_{n \to \infty} \|\mathbb{\Gamma}\big(t_n\big)^{-1}\| = +\infty$$

$$0 \leq \underbrace{\gamma_1(t_n) \leq \cdots \leq \gamma_m(t_n)}_{\substack{n \to +\infty \\ \longrightarrow 0}} < \beta \leq \gamma_{m+1}(t_n) \leq \cdots \leq \gamma_K(t_n) \leq 1$$

$\beta > 0$ $\qquad$ $m \geq 1$ $\qquad$ loss of rank .

Idea of the proof

Let $(C^n, \Phi^n) := (C(t_n), \Phi(t_n)) \in \mathcal{F}_N^K$. For all i, $\sqrt{\gamma_i^n}\phi_i^n$ is bounded in H^1 by energy conservation

$$\text{for } i \geq m+1,\ \gamma_i^n > \beta > 0, \quad \{\phi_i^n\}_{n\geq 1} \text{ is bounded in } H^1$$

$$\text{for } 1 \leq i \leq m, \begin{cases} \lim_{n\to+\infty} \|\sqrt{\gamma_i^n}\,\phi_i^n\|_{L^2(\mathbb{R}^3)} = 0 \\ \gamma_i^n = \sum_{i\in\sigma} |{c_\sigma}^n|^2 \quad \Longrightarrow \quad c_\sigma^n \longrightarrow 0 \quad \text{if } \sigma \cap \{1, \cdots, m\} \neq \emptyset \end{cases}$$

(C^n, Φ^n) weakly converges to $(C^\star, \Phi^\star)$ in $\mathbb{C}^{\binom{K}{N}} \times L^2(\mathbb{R}^3)^K$ and $\pi(C^n, \Phi^n)$ converges to

$$\Psi^* := \sum_{\sigma\cap\{1,2,\ldots,m\}=\emptyset} c_\sigma\,\Phi_\sigma = \pi(C^*, \Phi^*) \quad \in \mathrm{d}\mathcal{B}_N^{K-m}$$

Idea of the proof

$$\begin{aligned}\mathcal{E}(\pi(C^0,\Phi^0)) &= \lim_{n\to+\infty}\mathcal{E}(\pi(C^n,\Phi^n))\\ &\geq \mathcal{E}(\pi(C^*,\Phi^*))\\ &\geq I(K-m)\geq I(K-1)\end{aligned}$$

The above proof works on bounded domains

Skipped difficulties on $\mathbb{R}^3$:

- orthonormality constraints do not pass to weak limit
- the energy functional is not weakly lower semi continuous

Regularization of the density matrix

Regularization of the density matrix are used for numerics [Scrinzi et al, PRA 2005; Beck et al., PR 2000]

$$\mathbb{\Gamma}^\varepsilon = \mathbb{\Gamma} + \varepsilon\, I_K \quad \text{or} \quad \mathbb{\Gamma}^\varepsilon = \mathbb{\Gamma} + \varepsilon\, \exp(-\mathbb{\Gamma}/\epsilon), \qquad 0 < \varepsilon << 1.$$

$$\begin{aligned} i\frac{dC}{dt} &= \mathbb{K}[\Phi]\, C, \\ i\,\frac{\partial \Phi}{\partial t} &= (-\mathbf{\Delta} + \mathbf{V})\, \Phi + \mathbb{\Gamma}^\varepsilon(C)^{-1}\, (\mathbf{I} - \mathbf{P}_\Phi)\, \mathbb{W}[C, \Phi]\, \Phi, \\ \big(C(0), \Phi(0)\big) &= \big(C^0, \Phi^0\big) \in \mathrm{d}\mathcal{F}_N^K, \end{aligned}$$

The perturbed system preserves $\mathcal{F}_N^K$ but does not conserve energy !

$\Rightarrow$ L^2 global well-posedness for MCTDHF and regularized MCTDHF equations based on Strichartz estimates [N. Mauser, S. Trabelsi, M3As, 2010]

Convergence of the regularized problem

THEOREM.[C, Bardos, I.C., N. Mauser, S. Trabelsi, ARMA, 2010]

Let $(C_0, \Phi_0) \in \mathcal{F}_N^K$ *with* $\Phi_0 \in H^1(\mathbb{R}^3)^K$. *Assume that the solution* $(C(t), \Phi(t))$ *to the working equations is well-defined on* $[0, T]$ *and is such that*

$$\sup_{0 \leq t \leq T} \|\mathbb{\Gamma}(t)^{-1}\| \leq M < +\infty.$$

Then, on the same time interval it is the limit for $\epsilon \to 0$ *of the solution* $(C_\epsilon, \Phi_\epsilon)$ *to the regularized problem with the same initial data.*

Numerical experiments

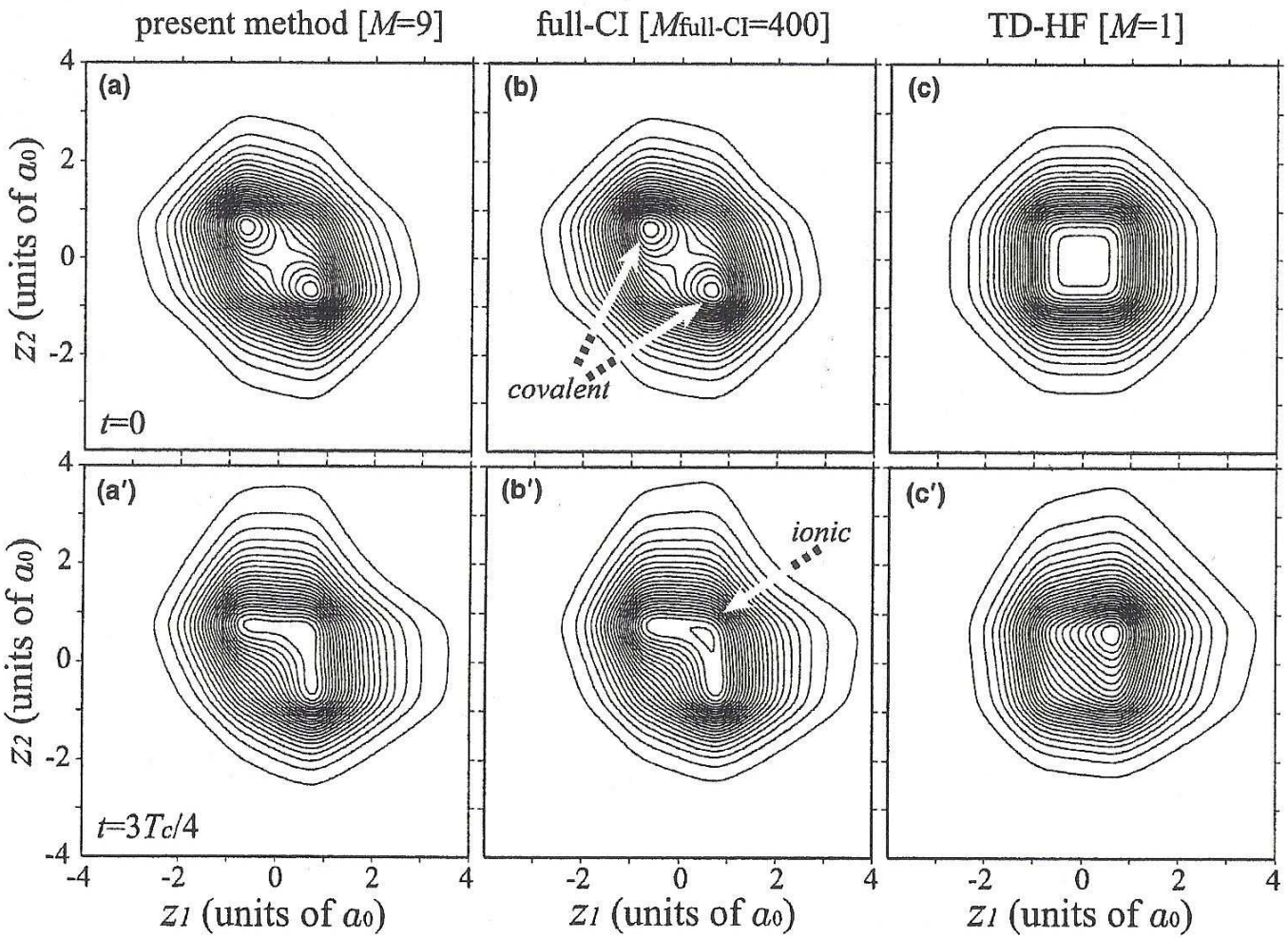


Fig. 2. Contour plots of the reduced density functions: (a)–(c) the ground state density functions at $t = 0$ and (a′)–(c′) the snapshots at $t = 3T_c/4$. We compare the results of the present method (a and a′) with those of the full-CI (b and b′) and of the TDHF (c and c′). The length of the determinantal expansion of the wave function M is depicted for each method. The contour lines in the six panels are plotted at the same intervals.

Extensions and Open questions

- **Laser fields** : Everything works for

$$\mathbf{H}(t) := (i\nabla + A(t))^2 + \omega(t)\, V(x)$$

with $\omega(t)$ and $A(t)$ real continuous, $A(0) = 0$. A typical example is $A(t) = A_0 \exp\big(-(t/\tau)^2\big) \sin(\alpha t)$, $A_0 > 0$

- What happens beyond finite T^*?
- Convergence to solutions to TDSE when $K \longrightarrow +\infty$
  Problem: might be that $T^*(K) \xrightarrow[K\to+\infty]{} 0$
  In any case $\gamma_1(t) \leq \frac{N}{K}$ thus $\|\mathbb{\Gamma}^{-1}\| \geq \frac{K}{N}$ goes to infinity with K.

Happy

Birthday

Claude !