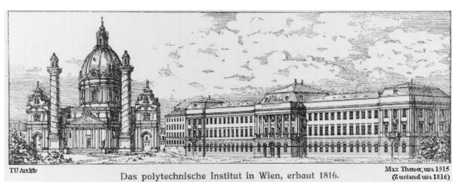

Time Propagation of the MCTDHF Equations in Electron Dynamics

Othmar Koch and Christian Lubich

Supported by the Austrian Academy of Sciences, APART Program

MCTDHF

Multi-configuration time-dependent Hartree–Fock method: The Time-Dependent Schrödinger Equation for a system of unbound fermions interacting by Coulomb forces,

$$\mathrm{i}\frac{\partial\psi}{\partial t}(x_1,\dots,x_f,t) = H\psi(x_1,\dots,x_f,t), \quad \psi(0)=\psi_0,$$

where

$$H = T + V = \sum_{k=1}^{f}\left(-\frac{1}{2}\Delta^{(k)} + \sum_{l<k}\frac{1}{|x_k - x_l|}\right)$$

is approximated by $u \in \mathcal{M}$,

$$\psi(x_1,\dots,x_f,t) \approx u := \sum_J a_J(t)\Phi_J(x,t)$$
$$= \sum_{j_1,\dots,j_f} a_{j_1,\dots,j_f}(t)\phi_{j_1}(x_1,t)\otimes\dots\otimes\phi_{j_f}(x_f,t).$$

Pauli exclusion principle: (a_J) is antisymmetric.
Dirac–Frenkel variational principle:

$$\left\langle \delta u \left| \mathrm{i}\frac{\partial u}{\partial t} - Hu \right.\right\rangle = 0 \quad \forall \text{ variations } \delta u \in \mathcal{T}_u\mathcal{M}.$$

Additional constraints for uniqueness:

$$\langle\phi_j|\phi_k\rangle = \delta_{j,k}, \quad \left\langle \phi_j \left| \frac{\partial\phi_k}{\partial t}\right.\right\rangle = -\mathrm{i}\,\langle\phi_j\,|\,T\,|\,\phi_k\rangle.$$

This yields the equations of motion

$$\mathrm{i}\frac{da_J}{dt} = \sum_K \langle\Phi_J\,|V|\,\Phi_K\rangle\, a_K =: \mathcal{A}_V(\phi)a,$$
$$\mathrm{i}\frac{\partial\phi_j}{\partial t} = T\phi_j + (I-P)\sum_k\sum_l \rho_{j,l}^{-1}\overline{V}_{l,k}\phi_k$$
$$=: T\phi + \mathcal{B}_V(a,\phi),$$

where

$\psi_j := \langle\phi_j|u\rangle$, … single hole functions,
$\rho_{j,l} := \langle\psi_j|\psi_l\rangle$, … density matrix,
$\overline{V}_{l,k} := \langle\psi_l\,|V|\,\psi_k\rangle$, … meanfield operator matrix,
$P := \sum_j|\phi_j\rangle\langle\phi_j|$, … orthogonal projector.

Existence Result

Theorem 1: Consider the MCTDHF equations of motion. If the initial data for ϕ_j is in the Sobolev space H^2, then there exists a unique classical solution of the MCTDHF equations satisfying

$$a_J \in C^2([0,t^*),\mathbb{C}), \quad \phi_j \in C^1([0,t^*),L^2)\cap C([0,t^*),H^2),$$

where either $t^*=\infty$ or the density matrix ρ becomes singular for $t=t^*$.

Outline of Proof

- Sobolev spaces H^k: The set of all functions in the Hilbert space L^2 of square integrable functions which have weak derivatives up to order $\le k$. Equipped with the norm

$$\|u\|_{H^k} := \left(\sum\|\partial^\alpha u\|_{L^2}^2\right)^{1/2},$$

where the sum is over all derivatives up to order k.
- $\mathcal{A}_V$, $\mathcal{B}_V$ contain terms

$$\left\langle \phi_1(x)\left|\frac{1}{|x-y|}\right|\phi_2(x)\phi_3(y) - \phi_3(x)\phi_2(y)\right\rangle =: \mathcal{F}_1(y),$$
$$\left\langle \phi_1(x)\tilde\phi_1(y)\left|\frac{1}{|x-y|}\right|\phi_2(x)\tilde\phi_2(y) - \tilde\phi_2(x)\phi_2(y)\right\rangle =: \mathcal{F}_2,$$

taking into account the anti-symmetry for fermions.
- Hardy, Hölder and Sobolev inequalities imply that bounds for $|\mathcal{F}_2|$ and the L^2, H^1 and H^2 norms of $\mathcal{F}_1$ only depend linearly on $\|\phi_j\|_{H^2}$.
- It is well known that T generates a strongly continuous group of unitary operators on H^2.
- Thus, by a classical theorem, a solution in H^2 exists locally in time.
- This satisfies the Dirac-Frenkel variational principle, whence energy conservation implies bounded H^1 norm.
- An integral representation associated with the variation of constant formula and the Gronwall lemma finally imply exponentially bounded H^2 norm.
- Thus, a solution exists in H^2 when ρ is nonsingular. □
- **Remark:** A different proof to show global existence in H^1 is given in **[Bardosetal08]**. This work includes a criterion for global invertibility of ρ.

Time Propagation: Variational Splitting

Algorithm: One step $t_0 \mapsto t_0 + \Delta t$

- Compute $u_{1/2}^- \in \mathcal{M}$ as the solution at time $t_0 + \frac{1}{2}\Delta t$ of

$$\left\langle \delta u \left| \mathrm{i}\frac{\partial}{\partial t} - T \right| u \right\rangle = 0 \quad \forall\delta u \in \mathcal{T}_u\mathcal{M}, \tag{1}$$

with initial value $u(t_0) = u_0$.
- Compute $u_{1/2}^+ \in \mathcal{M}$ as the solution at time $t_0 + \Delta t$ of

$$\left\langle \delta u \left| \mathrm{i}\frac{\partial}{\partial t} - V \right| u \right\rangle = 0 \quad \forall\delta u \in \mathcal{T}_u\mathcal{M},$$

with initial value $u(t_0) = u_{1/2}^-$.
- Compute $u_1 \in \mathcal{M}$ as the solution at time $t_0 + \Delta t$ of (1) with initial value $u(t_0 + 1/2\Delta t) = u_{1/2}^+$.

Remarks:

- Originally proposed by C. Lubich for quantum molecular dynamics with bounded potential V **[Lubich04]**.
- Equivalent to the classical second-order Strang splitting for the equations of motion into

$$\hat{T} := -\mathrm{i}(0,T)^T, \qquad \hat{V} := -\mathrm{i}(\mathcal{A}_V,\mathcal{B}_V).$$

- Equations of motion uncouple into single-particle (linear) Schrödinger equations with Hamiltonian T and computationally expensive computations for the particle-particle potential V
- These can be treated differently in both discretization method and time-stepping $\Longrightarrow$ increase in efficiency

Convergence Theorem

Theorem 2: Consider the numerical approximation of the MCTDHF equations of motion given by time semi-discretization based on variational splitting with step size Δt, $u_j \mapsto u_{j+1} = \mathcal{S}_{\Delta t}u_j$, $j = 0,1,\dots$. Then the convergence estimates

$$\|u_n - u(t_n)\|_{H^1} \le \text{const}\,\Delta t, \qquad \text{for } t_n = n\Delta t,$$
$$\|u_n - u(t_n)\|_{L^2} \le \text{const}\,(\Delta t)^2,$$

hold if the exact solution satisfies $u \in H^2$ for the first bound and $u \in H^3$ for the second.

Preparation: Lie Derivatives

For a (nonlinear) vector field F, denote by φ_F^t the flow of the differential equation

$$\dot\psi = F(\psi),$$

so that $\varphi_F^t(\psi_0)$ is the solution at time t of

$$\dot\psi = F(\psi), \quad \psi(0) = \psi_0.$$

If G is another vector field, define the *Lie derivative* D_F by

$$(D_F G)(\psi) = \frac{d}{dt}\Big|_{t=0} G(\varphi_F^t(\psi)) = G'(\psi)F(\psi)$$

for a vector field G on H^1 and $\psi \in H^1$. Now define

$$\big(\exp(tD_F)G\big)(\psi) := G(\varphi_F^t(\psi)).$$

Manipulation rules:

$$\frac{d}{dt}\exp(tD_F)G(\psi) = (D_F\exp(tD_F)G)(\psi)$$
$$= (\exp(tD_F)D_F G)(\psi),$$
$$[D_F, D_G] = D_{[G,F]},$$

for the *commutator* $[D_F, D_G] := D_F D_G - D_G D_F$ of the Lie derivatives of two vector fields F and G.

See **[Hairer etal02]**.

Example: The commutator $[\hat{T},\hat{V}]$ is computed using

$$[\mathrm{i}\Delta, \mathrm{i}\mathcal{B}_V](\phi) = \Delta\mathcal{B}_V(\phi) + \mathrm{i}\mathcal{B}_V{}'(\phi)(\Delta\phi), \quad \text{where}$$
$$\mathrm{i}\mathcal{B}_V{}'(\phi)(\Delta\phi) = \frac{d}{d\tau}\Big|_{\tau=0}\mathcal{B}_V\left(e^{\mathrm{i}\tau\Delta}\phi\right).$$

It is found that it contains terms of the forms

$$\left\langle \phi_1(x)\left|\nabla_y\frac{1}{|x-y|}\right|\phi_2(x)\nabla_y\phi_3(y) - \phi_3(x)\nabla_y\phi_2(y)\right\rangle,$$
$$\left\langle \phi_1(x)\left|\nabla_x\frac{1}{|x-y|}\right|\nabla_x\phi_2(x)\phi_3(y) - \nabla_x\phi_3(x)\phi_2(y)\right\rangle,$$

which can be estimated using Hardy, Hölder and Sobolev inequalities as in **Theorem 1**.

Outline of Proof

- First, stability in the H^1 norm is shown: If

$$\|u\|_{H^1} \le M_1, \qquad \|v\|_{H^1} \le M_1,$$

then we have

$$\|\mathcal{S}_{\Delta t}(u) - \mathcal{S}_{\Delta t}(v)\|_{H^1} \le e^{c_1\Delta t}\|u - v\|_{H^1},$$

with a constant $c_1 = c_1(M_1)$.
- We then estimate the local error in H^1. Let $u \in H^2$ and for some constant $M_2 > 0$,

$$\|u\|_{H^2} \le M_2,$$

then

$$\|\mathcal{S}_{\Delta t}(u_0) - u(\Delta t)\|_{H^1} \le c_2(\Delta t)^2,$$

with a constant $c_2 = c_2(M_2)$.
In this argument, a bound for the commutator $\|[\hat{T},\hat{V}](u)\|_{H^1} = \|[\mathrm{i}T,\mathrm{i}\mathcal{B}_V](u)\|_{H^1}$ is used. This depends on the H^2-norm of u.
- Combining stability and consistency in H^1, a standard argument yields convergence in H^1,

$$\|u_n - u(t_n)\|_{H^1} \le \text{const}\,\Delta t, \qquad \text{for } t_n = n\Delta t.$$

- Boundedness of the numerical solution in H^1,

$$\|u_n\|_{H^1} \le \text{const}\,,$$

now follows inductively.
- Next, stability in L^2 is investigated. It is found that

$$\|\mathcal{S}_{\Delta t}(u) - \mathcal{S}_{\Delta t}(v)\|_{L^2} \le e^{c_3\Delta t}\|u - v\|_{L^2},$$

with a constant $c_3 = c_3(M_1)$.
- Then, the local error in L^2 is considered. We estimate the L^2-norm of the double commutator $\|[\hat{T},[\hat{T},\hat{V}]](u)\|_{L^2} = \|[\mathrm{i}T,[\mathrm{i}T,\mathrm{i}\mathcal{B}_V]](u)\|_{L^2}$. The bound depends on $M_3 = \|u\|_{H^3}$. We thus conclude

$$\|\mathcal{S}_{\Delta t}(u_0) - u(\Delta t)\|_{L^2} \le c_4(\Delta t)^3,$$

where $c_4 = c_4(M_3)$.
- Since we had previously concluded that $\|u_n\|_{H^1}$ is bounded, we conclude convergence:

$$\|u_n - u(t_n)\|_{L^2} \le \text{const}\,(\Delta t)^2.$$

- Finally we conclude that the numerical approximation u_n is in H^2 by showing boundedness of $u_{1/2}^+$. This follows using the Gronwall Lemma. □

Remark: A similar analysis is given for the cubic nonlinear Schrödinger equation in **[Lubich08]**.

Outlook

Incorporation of a nuclear attractive potential $U = \sum_{k=1}^{f}\frac{1}{|x_k|}$:

- $U \ll \Delta \Rightarrow$ **Theorem 1** ✓
- Variational splitting — work in progress.

Incorporation of a drift term, $T^{(k)} \leftarrow \frac{1}{2}\left(-\mathrm{i}\nabla^{(k)} - A(t)\right)^2$ (motivated by applications in laser physics **[Caillatetal05]**):

- $\nabla \ll \Delta \Rightarrow$ **Theorem 1** ✓
- Variational splitting — no new terms appear in commutators $\Rightarrow$ **Theorem 2** ✓

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