On the MCTDH method

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Setting

Full quantum dynamics: Schrödinger equation

$$i\dot{\psi} = H\psi, \qquad \psi = \psi(x_1, \dots, x_N, t)$$

$$\uparrow$$

Intermediate models: via Dirac–Frenkel variational principle

Born–Oppenheimer, MCTDH, Gaussian wavepackets, ...

$$\downarrow$$

Classical molecular dynamics: Newtonian eqs. of motion

$$M\ddot{x} = -\nabla V(x), \qquad x = (x_1(t), \dots, x_N(t))$$

Time-dependent Schrödinger equation

$$i \, \frac{\partial \psi}{\partial t} = H\psi$$

N-particle wavefunction: $\psi = \psi(x, t)$

for $x = (x_1, \dots, x_N) \in \mathbb{R}^{3N}, \; x_n \in \mathbb{R}^3, \; t > 0$

Hamiltonian operator: $H = T + V$

with the kinetic and potential energy operators

$$T = -\sum_{j=1}^{N} \frac{1}{2m_j} \Delta_{x_j} \quad \text{and} \quad V = V(x)$$

(smooth and bounded)

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high-dimensional linear PDE

Outline

The Dirac–Frenkel variational approximation principle

The MCTDH method, or dynamical low-rank approximation

Modelling error of MCTDH

Splitting integrators for MCTDH

Outline

Dirac-Frenkel principle: abstract formulation

$\mathcal{M} \subset L^2(\mathbb{R}^{3N})$ approximation manifold

$T_u\mathcal{M}$ tangent space at $u \in \mathcal{M}$, *complex* linear

approximate wavefunction $u = u(\cdot, t) \in \mathcal{M}$ determined from

$$\langle i\dot{u} - Hu,\ v\rangle = 0 \qquad \text{for all} \quad v \in T_u\mathcal{M}$$

Galerkin method on a state-dependent approximation space

Dirac 1930, Frenkel 1934 (TDHF)

Orthogonal projection

$$\mathit{Re}\,\langle \dot{u} - \tfrac{1}{i} Hu,\ v\rangle = 0 \qquad \text{for all} \quad v \in T_u\mathcal{M}$$

equivalent to minimum defect condition:

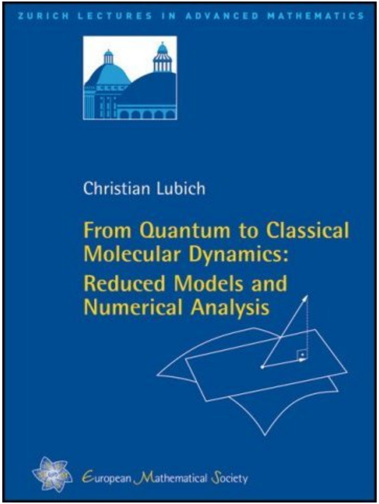
determine approximation $t \mapsto u(t) \in \mathcal{M}$ such that

$$\dot{u} = \vartheta \in T_u\mathcal{M} \quad \text{with} \quad \|\vartheta - \tfrac{1}{i} Hu\| = \min!$$

orthogonal projection $\dot{u} = P(u)\frac{1}{i}Hu$

Frenkel 1934 $\rightarrow$ *Dirac*

Cover illustration: tangent space projection



$$\dot{u} = P(u)\frac{1}{i}Hu$$

2008

Symplectic projection

$$\mathit{Im}\, \langle \dot{u} - \tfrac{1}{i} Hu,\ v \rangle = 0 \qquad \text{for all} \quad v \in T_u\mathcal{M}$$

- symplectic 2-form on $L^2(\mathbb{R}^{3N})$: $\quad \omega(\xi, \eta) = -2\, \mathit{Im}\, \langle \xi, \eta \rangle$
- Hamiltonian function: $\quad \mathcal{H}(u) = \langle u, Hu \rangle$

Hamiltonian system on the manifold $\mathcal{M}$:

$$\omega(\dot{u}, v) = d\mathcal{H}(u)v \qquad \text{for all} \quad v \in T_u\mathcal{M}$$

Consequences:

- energy conservation: $\mathcal{H}(u(t)) = \text{Const.}$
- symplecticity of the flow: for $\xi(t), \eta(t)$ tangent vectors propagated by the linearized flow along $u(t)$,

$$\omega(\xi(t), \eta(t)) = \text{Const.}$$

Approximation error

- A posteriori error bound

$$\|u(t) - \psi(t)\| \le \int_0^t \mathrm{dist}\big(\tfrac{1}{i} Hu(\tau), T_{u(\tau)}\mathcal{M}\big)\, d\tau$$

- Quasi-optimality

$$\|u(t) - \psi(t)\| \le d(t) + Ce^{c\kappa t} \int_0^t d(\tau)\, d\tau$$

with $d(t) = \mathrm{dist}(\psi(t), \mathcal{M})$ the best-approximation error,

κ bound of the curvature of $\mathcal{M}$

L. 2005

Outline

MCTDH

Multi-Configuration Time-Dependent Hartree method

Time-dependent Hartree method

Hartree product:

$$\psi(x_1,\ldots,x_N,t) \approx \phi^{(1)}(x_1,t)\cdot\ldots\cdot\phi^{(N)}(x_N,t)$$

Equations of motion for single-particle functions $\phi^{(n)}(x_n,t)$ by Dirac–Frenkel reduction to

$$\mathcal{M} = \left\{\phi^{(1)}\otimes\ldots\otimes\phi^{(N)} \,\middle|\, \phi^{(n)}\in L^2(\mathbb{R}^3)\right\}$$

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separation of variables, rank-1 approximation

MCTDH

Multi-Configuration Time-Dependent Hartree method

Approximate by linear combination of Hartree products

$$\psi(x,t) \approx \sum_{j_1=1}^{r_1} \dots \sum_{j_N=1}^{r_N} a_{j_1 \dots j_N}(t)\, \phi_{j_1}^{(1)}(x_1,t) \cdot \ldots \cdot \phi_{j_N}^{(N)}(x_N,t)$$

- mutually orthogonal single-particle functions $\phi_1^{(n)}, \dots, \phi_{r_n}^{(n)}$
- core tensor $(a_{j_1 \dots j_N})$ of full multilinear rank $(r_1, \dots, r_N)$

Tucker format, not canonical format
also hierarchical Tucker format

H.D. Meyer et al. 1990 - , MCTDH book 2009

MCTDH equations of motion

from Dirac–Frenkel variational principle on MCTDH manifold:
coupled system of ODEs and low-dimensional nonlinear PDEs

$$
\begin{aligned}
i\,\frac{da_J}{dt} &= \sum_L \langle \Phi_J \,|\, H \,|\, \Phi_L \rangle\, a_L \\
i\rho^{(n)}\,\frac{\partial \phi^{(n)}}{\partial t} &= \left(I - P^{(n)}\right) \langle H \rangle_{\neg x_n} \phi^{(n)}
\end{aligned}
$$

with $\Phi_J(x,t) = \prod_{n=1}^{N} \phi_{j_n}^{(n)}(x_n,t)$ for multi-indices $J = (j_1, \ldots, j_N)$

Meyer, Manthe & Cederbaum 1990

existence and regularity: *Koch & L. 2007*

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Convergence of MCTDH to Schrödinger solution???

Naive expectation: Taking more and more terms in the linear combination of Hartree products yields an ever better accuracy

Obstructions:

- approximation properties of basis of Hartree products $\Phi_J(\cdot, t)$?
- ill-conditioned coefficient tensor (a_J)
  $\leftrightarrow$ ill-conditioned density matrices $\rho^{(n)} = A_{(n)} A_{(n)}^*$

Quasi-optimality of variational approximation

$u(t)$ MCTDH approximation, $\psi(t)$ exact wave function

MCTDH error bounded by best-approximation error:

$$\|u(t) - \psi(t)\| \le d(t) + C_\kappa \, e^{c\kappa t} \int_0^t d(\tau)\, d\tau$$

with $d(t) = \text{dist}(\psi(t), \mathcal{M})$ and κ curvature of manifold $\mathcal{M}$

but $\kappa \sim \text{cond}(\rho^{(n)})^{1/2} \to \infty$ as number of configurations $\to \infty$

L. 2005

Objectives

- error bounds in the case of ill-conditioned density matrices
- convergence as number of configurations $\to \infty$

under appropriate assumptions

Conte & L., M2AN 2010

Approximability assumption

Exact wavefunction can be written as

$$\psi(t) = v(t) + e(t)$$

where $v(t) \in \mathcal{M}$ has small defect:

$$\left\| i\frac{\partial v}{\partial t}(\cdot, t) - Hv(\cdot, t) \right\| \leq \varepsilon$$

Small error $e(t)$ and small defect (ε) for some linear combination of Hartree products

Assumptions on the coefficient tensor

$B_{(n)}(t)$ nth unfolding of coefficient tensor $(b_{j_1 \dots j_N}(t))$ of $v(t) \in \mathcal{M}$, for $n = 1, \dots, N$

- Bound of the pseudo-inverse (large: small δ!):

$$\|B_{(n)}^{\dagger}(t)\|_2 = \frac{1}{\sigma_{r_n}(B_{(n)}(t))} \le \delta^{-1}$$

- Small time derivatives of the coefficient tensor in components that correspond to small singular values:

$$\left\| B_{(n)}^{\dagger}(t)\, \dot{B}_{(n)}(t) \right\|_2 \le c,$$

Error bound

$$\|u(t) - \psi(t)\| \le \|e(t)\| + 2t\varepsilon \qquad \text{for} \quad t = O(\delta/\varepsilon)$$

Bound independent of δ (ill-conditioning)

for fixed rank, can achieve $\delta \sim \varepsilon$ by perturbing $v(t)$: $t = O(1)$

Convergence: if we *assume* $\delta \sim \varepsilon$, we can admit arbitrary number of configurations: $\varepsilon \to 0$

Proof of the error bound

use estimates for the tangent space projection $P(u)$ to show a quadratic differential inequality

$$\frac{d}{dt}\|u - v\| \leq \frac{C}{\delta}\|u - v\|^2 + \varepsilon$$

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Variational splitting: the formal game

variational approximation: $H = T + V$

$$\langle i\dot{u} - Hu,\ v\rangle = 0 \qquad \text{for all} \quad v \in T_u\mathcal{M}$$

time step $u_n \to u_{n+1}$ via

1. half-step with V: $u^-_{n+1/2}$ solution at $\Delta t/2$ of

$$\langle i\dot{u} - Vu,\ v\rangle = 0 \qquad \text{for all} \quad v \in T_u\mathcal{M}$$

2. full step with T: $u^+_{n+1/2}$ solution at Δt of

$$\langle i\dot{u} - Tu,\ v\rangle = 0 \qquad \text{for all} \quad v \in T_u\mathcal{M}$$

3. half-step with V: u_{n+1} solution at $\Delta t/2$ of

$$\langle i\dot{u} - Vu,\ v\rangle = 0 \qquad \text{for all} \quad v \in T_u\mathcal{M}$$

Variational splitting for MCTDH

- Step with T: since $Tu \in T_u\mathcal{M}$ for $u \in \mathcal{M}$, integration step with T decouples into $\dot{a}_J = 0$ and

$$i \, \frac{\partial \phi_j^{(k)}}{\partial t}(x_k, t) = -\frac{1}{2m_k} \, \Delta_{x_k} \phi_j^{(k)}(x_k, t)$$

  low-dimensional free Schrödinger equations $\to$ FFT

- Step with V: MCTDH for Hamiltonian V instead of H
  $\to$ larger time steps independently of the space discretization

  work in preparation (with I. Oseledets): integration scheme that does not suffer from ill-conditioning of the matrices $\rho^{(k)}$

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explicit, unconditionally stable scheme *L. 2004*

Error of variational splitting for MCTDH

second order error bound

$$\|u_n - u(t_n)\| \le C\,\Delta t^2 \,\max \|u(t)\|_{H^2}$$

same as for Strang splitting for the linear Schrödinger equation

basic tool: Lie-commutator bounds for vector fields corresponding to T and V

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