3.4.2 The MCTDH Method

Generalization of the aforementioned model reductions for the finite-dimensional case to the infinite-dimensional case applicable to the multi-particle Schrödinger equation.

Approximation $\psi \approx u \in \mathcal{M}$,

$$\mathcal{M} = \{ u \,;\, u = \sum_{j_1=1}^{N_1} \cdots \sum_{j_f=1}^{N_f} a_{j_1 \cdots j_f}(t)\, \phi_{j_1}^{(1)}(x_1, t) \cdot \ldots \cdot \phi_{j_f}^{(f)}(x_f, t) \}$$

We impose the "Dirac-Frenkel Variational Principle" [6]

$$\langle \delta u \,|\, \dot{u} - \tfrac{1}{i} H u \rangle = 0 \quad \forall \, \delta u \in T_u \mathcal{M} \qquad (1)$$

δu ... "variations", i.e. tangent space to $\mathcal{M}$ in u.

This procedure is referred to as "Multi-Configuration Time-Dependent Hartree Method".

For fermions (i.e., electronic Schrödinger equation), an antisymmetrized version for indistinguishable particles is used ("Multi-Config Time-Dep. Hartree-Fock")

$$\mathcal{M} = \{ u \,;\, u = \sum_{j_1=1}^{N} \cdots \sum_{j_f=1}^{N} a_{j_1 \cdots j_f}(t)\, \phi_{j_1}(x_1, t) \cdot \ldots \cdot \phi_{j_f}(x_f, t),$$

$a_J = a_{j_1 \cdots j_f}$ is antisymmetric under exchange of any two indices $\}$

Formally analogous to the dynamical low rank approximation of matrices and tensors, it is possible to derive differential equations for the coefficient tensor a_J and the "orbitals" or "single-particle functions" $\phi_{j_k}^{(k)}$: If in addition to the variational principle (1) from p. 45 the "gaugeing" or "orthogonality constraints"

$$\langle \phi_{j_k}^{(k)} | \phi_{l_k}^{(k)} \rangle = \delta_{jl} \quad , \forall j, l, k$$

$$\langle \phi_{j_k}^{(k)} | \dot{\phi}_{l_k}^{(k)} \rangle = 0$$

are imposed, the resulting equations of motion (also referred to as "working equations") are

$$i \dot{a}_J = \sum_K \langle \Phi_J | H | \Phi_K \rangle a_K \qquad \forall J = (j_1, \dots, j_f)$$

$$i \dot{\phi}_{j_k}^{(k)} = (Id - P^{(k)}) \sum_{m_k=1}^{N_k} \sum_{l_k=1}^{N_k} \left[\rho^{(k)} \right]^{-1}_{j_k, m_k} \cdot$$

$$\cdot \langle \psi_{m_k}^{(k)} | H | \psi_{l_k}^{(k)} \rangle \phi_{l_k}^{(k)}$$

where

$$\Phi_J(x_1, \dots, x_f, t) := \phi_{j_1}^{(1)}(x_1) \cdot \ldots \cdot \phi_{j_f}^{(f)}(x_f),$$

$$\psi_{j_k}^{(k)}(x_1, \dots, x_{k-1}, x_{k+1}, \dots, x_f; t) = \langle \phi_{j_k}^{(k)} | u \rangle = \%$$

$$(\varphi_{j_k}^{(k)}) = \sum_{j_1=1}^{N_1} \cdots \sum_{j_{k-1}}^{N_{k-1}} \sum_{j_{k+1}}^{N_{k+1}} \cdots \sum_{j_f=1}^{N_f} a_{j_1 \cdots j_f}(t)\, \phi_{j_1}^{(1)}(x_1,t) \cdot \ldots \cdot \phi_{j_{k-1}}^{(k-1)}(x_{k-1},t) \cdot$$

$$\cdot\, \phi_{j_{k+1}}^{(k+1)}(x_{k+1},t) \cdot \ldots \cdot \phi_{j_f}^{(f)}(x_f,t)$$

$$\rho^{(k)} = (\rho_{i_k,j_k}^{(k)})_{i_k,j_k=1}^{N_k} \quad \text{with} \quad \rho_{i_k,j_k}^{(k)} = \langle \varphi_{i_k}^{(k)} | \varphi_{j_k}^{(k)} \rangle$$

$P^{(k)}$ ... orthogonal projector onto the space spanned by $\phi_{j_k}^{(k)}$, i.e.

$$P^{(k)} \varphi = \sum_{j_k=1}^{N_k} \phi_{j_k}^{(k)} \langle \phi_{j_k}^{(k)} | \varphi \rangle .$$

All inner products above are meant w.r.t. all variables present in both arguments $\langle \cdot | \cdot \rangle$.

A more convenient form of the working equations may be obtained by imposing a different gauging:

Note that orthonormality $\langle \phi_i | \phi_j \rangle = \delta_{ij}$ implies $\frac{d}{dt}\langle \phi_i | \phi_j \rangle = \langle \dot{\phi}_i | \phi_j \rangle + \langle \phi_i | \dot{\phi}_j \rangle \equiv 0$.

So alternatively

$$\langle \phi_{j_k}^{(k)} | \dot{\phi}_{\ell_k}^{(k)} \rangle = -i \langle \phi_{j_k}^{(k)} | G^{(k)} | \phi_{\ell_k}^{(k)} \rangle$$

can be required, with any self-adjoint operators $G^{(k)}$.

Particularly, with the choice $G^{(k)} = T^{(k)} = -\frac{1}{2M_k}\Delta^{(k)}$,

the working equations become

$$i\dot{a}_j = \sum_K \langle \Phi_j | V | \Phi_K \rangle a_K \qquad =: d_V(\phi)a, \forall j$$

$$i\dot{\phi}^{(k)}_{j_k} = T^{(k)}\phi^{(k)}_{j_k} + (\mathrm{Id} - P^{(k)}) \sum_{l_k=1}^{N_k}\sum_{m_k=1}^{N_k} (\rho^{(k)})^{-1}_{j_k, m_k} \langle \psi^{(k)}_{m_k} | V | \psi^{(k)}_{l_k} \rangle \phi^{(k)}_{l_k} \qquad =: T\phi + B_V(a,\phi),$$

$$j_k = 1, \ldots, N_k, \; k = 1, \ldots, f.$$

The method was introduced in [29] and comprehensively described in [30], where particularly the gauging topics above are treated. A more mathematical discussion is given in [31].

Quasi-Optimality of Variational Approximations

We have now introduced several variational approximations on particular manifolds $\mathcal{M}$, and seek to improve the very general a posteriori error bound from 32 by means of particular assumptions on the problem and on $\mathcal{M}$. These correspond to properties which could be indeed verified in the finite-dimensional case in the context of the low rank approximation of time-dependent matrices, see 44.

Assumptions:

1) $H = A + B$, A, B ... self-adjoint,
2) $e^{-itA} u \in \mathcal{M} \;\forall u \in \mathcal{M} \Leftrightarrow Au \in T_u\mathcal{M} \;\forall u \in \mathcal{M} \cap D(A)$
3) $\|B\varphi\| \leq \beta \|\varphi\| \quad \forall \varphi \in \mathcal{H}$
4) $T_u\mathcal{M}$ ... complex linear, $u \in T_u\mathcal{M} \;\forall u \in \mathcal{M}$
5) Curvature bounds for $u, v \in \mathcal{M}$, $\varphi \in \mathcal{H}$

$$\|(P(u) - P(v))\varphi\| \leq \varkappa \|u - v\| \, \|\varphi\|$$

$$\|P^\perp(v)(u-v)\| \leq \varkappa \|u - v\|^2$$

$P(u)$ ... ortho. proj. onto $T_u\mathcal{M}$, $P^\perp := I - P$.

6) $\mathcal{P}(u(t))\varphi \in C^1$ for $u \in C^1$, $\varphi \in \mathcal{H}$.

7) $\psi(0) \in \mathcal{M}$, $\|\psi(0)\| = 1$

8) $\operatorname{dist}(\psi(t), \mathcal{M}) \leq \frac{1}{2\varkappa}$, $0 \leq t \leq t^*$

9) $\psi, u \in D(H)$,

$\|H\psi(t)\| \leq \mu$, $\|Hu(t)\| \leq \mu$, $\|Au(t)\| \leq \mu$.

10) $\operatorname{dist}(H\psi(t), T_{v(t)}\mathcal{M}) \leq \delta$,

$\operatorname{dist}(Hu(t), T_{u(t)}\mathcal{M}) \leq \delta$,

$v \in \mathcal{M}$ ... best approximation, $\|v(t) - \psi(t)\| = \operatorname{dist}(\psi(t), \mathcal{M})$.

Remark: The critical assumption 3) is not satisfied for the Coulomb potential. For the necessary modifications, see [13].

Under the assumptions 1) – 10) above, we can prove the following Theorem:

Theorem (Quasi-optimality of variational approx.):
Under the assumptions 1) – 10), the error satisfies

$$\|u(t) - \psi(t)\| \leq d(t) + C \cdot e^{\gamma t} \int_0^t d(s)\, ds, \quad 0 \leq t \leq t^*,$$

$d(t) := \operatorname{dist}(\psi(t), \mathcal{M})$, $\gamma = 2\varkappa\delta$, $C = \beta + 3\varkappa\mu$.

Proof:

a) v is characterized by

$$P(v)(v-\psi)=0. \qquad (*)$$

$$\Rightarrow P(v)(\dot{v}-\dot{\psi})+[P'(v)\cdot(v-\psi)]\dot{v}$$

(note: $[P'(v)\cdot\varphi]\dot{v}=\frac{d}{dt}P(v(t))\varphi$)

$\dot{v}\in T_v M \Rightarrow P(v)\dot{v}=\dot{v} \leadsto$

$$(I+P'(v)\cdot(v-\psi))\dot{v}=P(v)\dot{\psi}. \qquad (1)$$

By 5) and 8) we thus have

$$\|P'(v)\cdot(v-\psi)\|\leq \varkappa\|v-\psi\|\leq\tfrac{1}{2}$$

$\Rightarrow I+P'$ in (1) is invertible, and

$$\dot{v}=P(v)\dot{\psi}+r(v,\psi),\quad \|r(v,\psi)\|\leq 2\varkappa\mu\|v-\psi\|,$$

using 9), $\|\dot{\psi}\|=\|H\psi\|\leq\mu$.

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

In the following, assume $v(t)\in D(H)=D(A)$.

(This is an assumption on the regularity of the best approximation!)

b) $\dot{u} - \dot{v} = \mathcal{P}(u)\frac{1}{i}Hu - \mathcal{P}(v)\dot{\psi} - r(v,\psi) \quad |\cdot(u-v)$

$$\langle u-v | \dot{u}-\dot{v}\rangle = \langle u-v | \mathcal{P}(u)\tfrac{1}{i}Hu - \mathcal{P}(v)\dot{\psi} - r(v,\psi)\rangle$$

$$\operatorname{Re}\langle u-v | \dot{u}-\dot{v}\rangle = \operatorname{Re}\langle u-v | \mathcal{P}(u)\tfrac{1}{i}Hu - \mathcal{P}(v)\dot{\psi} - r(v,\psi)\rangle = (*)$$

$$\| $$

$$\frac{1}{2}\frac{d}{ds}\|u-v\|^2 = \|u-v\|\frac{d}{ds}\|u-v\|$$

$(*) = \mathrm{I} + \mathrm{II} + \mathrm{III}$

$$\mathrm{I} = -\operatorname{Re}\langle u-v | \mathcal{P}(u)iHu - \mathcal{P}(v)iHv\rangle$$

$$\mathrm{II} = -\operatorname{Re}\langle u-v | \mathcal{P}(v)iH(v-\psi)\rangle$$

$$\mathrm{III} = -\operatorname{Re}\langle u-v | r(v,\psi)\rangle$$

c) $Au \in T_u M \Rightarrow \mathcal{P}^{\perp}(v)iAv = 0$, $H = A+B$ selfadjoint $\Rightarrow$

$$\mathrm{I} = \operatorname{Re}\langle u-v | \mathcal{P}^{\perp}(u)iHu - \mathcal{P}^{\perp}(v)iHv\rangle$$

(since $\operatorname{Re}\langle u-v | iHu - iHv\rangle = \operatorname{Re}(i\underbrace{\langle u-v|H|u-v\rangle}_{\in\mathbb{R}})$ $= 0$)

$$= \operatorname{Re}\langle u-v | \mathcal{P}^{\perp}(u)iHu\rangle - \operatorname{Re}\langle u-v | \mathcal{P}^{\perp}(v)iBv\rangle.$$

$$\mathrm{II} = -\operatorname{Re}\langle u-v | \mathcal{P}(v)iA(v-\psi)\rangle - \operatorname{Re}\langle u-v | \mathcal{P}(v)iB(v-\psi)\rangle.$$

Using Assumption 4) $\Rightarrow \mathcal{P}(v)v = v$.

$(*)$ on p. 51 implies thus

$$v = \mathcal{P}(v)\psi, \quad v-\psi = \mathcal{P}^{\perp}(v)(v-\psi) = -\mathcal{P}^{\perp}(v)\psi.$$

$$\langle v | P(v) iA(v-\psi)\rangle = -\langle v | P(v) iAP^\perp(v)\psi\rangle =$$
$$= \langle P^\perp(v) iAv | \psi\rangle = 0$$

(by Assumption 2))

Similarly,

$$\langle u | iAP^\perp(u)(v-\psi)\rangle = 0.$$

Together, we have

$$\langle u-v | P(v) iA(v-\psi)\rangle =$$
$$= \langle u | iA(v-\psi)\rangle - \langle u-v | P^\perp(v) iA(v-\psi)\rangle$$
$$= -\langle u | iA(P^\perp(u) - P^\perp(v))(v-\psi)\rangle +$$
$$+ \langle u-v | P^\perp(v) iA\psi\rangle =$$
$$= -\langle iAu | (P(u) - P(v))(v-\psi)\rangle +$$
$$+ \langle P^\perp(v)(u-v) | P^\perp(v) iH\psi\rangle -$$
$$- \langle u-v | P^\perp(v) iB\psi\rangle.$$

Thus,

$$I + II = \mathrm{Re}\langle P^\perp(u)(u-v) | P^\perp(u) iHu\rangle -$$
$$- \mathrm{Re}\langle u-v | iB(v-\psi)\rangle +$$
$$+ \mathrm{Re}\langle iAu | (P(u) - P(v))(v-\psi)\rangle -$$
$$- \mathrm{Re}\langle P^\perp(v)(u-v) | P^\perp(v) iH\psi\rangle.$$

With the assumptions,

$$|\mathrm{I}+\mathrm{II}| \leq \varkappa \|u-v\|^2 \delta + \|u-v\| \cdot \beta \|v-\varphi\| +$$
$$+ \mu \varkappa \|u-v\| \|v-\varphi\| + \varkappa \|u-v\|^2 \delta =$$
$$= 2 \varkappa \delta \|u-v\|^2 + (\beta + \varkappa\mu) \|u-v\| \|v-\varphi\|.$$

$$\Rightarrow \frac{d}{dt} \|u-v\| \leq \gamma \|u-v\| + C \|v-\varphi\|,$$

$\gamma := 2 \varkappa \delta$, $C := \beta + 3 \varkappa \mu$.

Gronwall lemma $\Rightarrow$

$$\|u(t)-v(t)\| \leq C \cdot e^{\gamma t} \int_0^t \|v(s)-\varphi(s)\| ds$$

Triangle inequality for $u-\varphi = (u-v)+(v-\varphi)$ yields the proposition of the Theorem. //