Tensorization of Electronic Schrödinger Equation and Second Quantization

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Introduction

Electronic Schrödinger Equation
Quantum mechanics

**Goal:** Calculation of physical and chemical properties on a microscopic (atomic) length scale.

- E.g. atoms, molecules, clusters, solids
- E.g. chemical behaviour, bonding energies, ionization energies, conduction properties, essential material properties
Electronic structure calculation

Reduction of the problem to the computation of an electronic wave function $\Psi$ for given fixed nuclei.

The electronic Schrödinger equation, describes the stationary nonrelativistic behaviour of system of $N$ electrons in an electrical field

$$\hat{H}\psi = E\psi.$$

Electronic structure determines e.g.

- bonding energies, reactivity
- ionization energies
- conductivity,
- in a wider sense molecular geometry, -dynamics,...

of atoms, molecules, solids etc.
Electronic Schrödinger equation

\( N \) nonrelativistic electrons + Born Oppenheimer approximation

\[
H\psi = E\psi
\]

The Hamilton operator in atomic units

\[
H = -\frac{1}{2} \sum_i \Delta_i - \sum_i \sum_{\nu=1}^K \frac{Z_\nu}{|x_i - a_\nu|} + \frac{1}{2} \sum_{i\neq j}^N \frac{1}{|x_i - x_j|}
\]

acts on \textit{anti-symmetric} wave functions (Pauli principle)

\[
\psi(x_1, s_1, \ldots, x_N, s_N) \in \mathbb{R}, \quad x_i = (x_i, s_i) \in \mathbb{R}^3 \times \{\pm \frac{1}{2}\},
\]

\[
\psi(\ldots; x_i, s_i; \ldots; x_j, s_j; \ldots) = -\psi(\ldots; x_j, s_j; \ldots; x_i, s_i; \ldots)
\]
Variational formulation

**Input:** position $a_\nu$ of the $\nu$’s atom, nuclear charge $Z_\nu$, number of electrons $N$.

**Output:** We are mainly interested in the ground-state energy, i.e. the lowest eigenvalue in the configuration space

$$\psi \in \mathcal{V} = H^1(\mathbb{R}^3 \times \{\pm \frac{1}{2}\})^N \cap \bigwedge_{i=1}^N L^2(\mathbb{R}^3 \times \{\pm \frac{1}{2}\}).$$

$$E_0 = \min_{\langle \psi, \psi \rangle = 1} \langle H\psi, \psi \rangle, \quad \psi = \arg\min_{\langle \psi, \psi \rangle = 1} \langle H\psi, \psi \rangle$$

Basic Problem - Curse of dimensionality

- linear eigenvalue problem, but extremely high-dimensional
- + anti-symmetry constraints + lack of regularity.
- traditional approximation methods (FEM, Fourier series, polynomials, MRA etc.): approximation error in $\mathbb{R}^1$: $\lesssim n^{-s}$, $s$-regularity $\sim$, $\mathbb{R}^{3N'}$: $\lesssim n^{3sN'}$, ($s < \frac{5}{2}$) with $n$ DOFs
- Nondeterministic methods: Quantum Monte Carlo methods have problems with fermions

For large systems $N' >> 1$ ($N' > 1$) the electronic Schrödinger equation seems to be intractable! But 70 years of impressive progress has been awarded by the Nobel price 1998 in Chemistry: Kohn, Pople.
Slater determinants
and Full CI method
Anti-symmetric tensor products - Slater determinants

Approximation by sums of anti-symmetric tensor products:

\[ \psi = \sum_{k=1}^{\infty} c_k \psi_k \]

\[ \psi_k(\mathbf{x}_1, s_1; \ldots; \mathbf{x}_{N'}, s_{N'}) = \varphi_{1, k} \wedge \ldots \wedge \varphi_{N', k} = \frac{1}{\sqrt{N'!}} \det(\varphi_{i,k}(\mathbf{x}_j, s_j)) \]

with \( \varphi_{i,k} \in \{\varphi_j : j = 1, \ldots\} \), w.l.o. generality

\[ \langle \varphi_i, \varphi_j \rangle = \sum_{s=\pm \frac{1}{2}} \int_{\mathbb{R}^3} \varphi_i(\mathbf{x}, s) \overline{\varphi_j(\mathbf{x}, s)} d\mathbf{x} = \delta_{i,j} . \]

A Slater determinant: \( \psi_k \) is an (anti-symmetric) product of \( N' \) orthonormal functions \( \varphi_i \), called spin orbital functions

\[ \varphi_i : \mathbb{R}^3 \times \{\pm \frac{1}{2}\} \rightarrow \mathbb{R} , \; i = 1, \ldots, N, \]

For present applications it is sufficient to consider real valued functions \( \psi, \varphi \).
Single and two particle operators

Abbreviations:

a) Single particle operators

\[ A = h = \sum_{i=1}^{N'} h_i = \sum_{i=1}^{N'} \left( \frac{1}{2} \Delta_i + \sum_{j=1}^{M} \frac{-Z_j}{|x_i - R_j|} \right) = \sum_{i=1}^{N'} \left( \frac{1}{2} \Delta_i + V_{\text{core}}(x_i) \right), \quad h_i = h_j, \]

\[ \langle i | h | j \rangle := \langle \varphi_i, h_j \varphi_j \rangle \]

a) Two particle operators

\[ G = \sum_{i}^{N'} \sum_{j>i}^{N'} \frac{1}{|x_i - x_j|}, \quad H = h + G \]

\[ \langle a, b | i, j \rangle := \sum_{s, s'} = \pm \frac{1}{2} \int \int \varphi^*_a(x, s) \varphi^*_b(x', s') \varphi_i(x, s) \varphi_j(x', s') \frac{dxdx'}{|x - x'|} \]

\[ \langle a, b || i, j \rangle := \langle a, b | i, j \rangle - \langle a, b | j, i \rangle. \]
Single and two particle operators

Abbreviations:

a) Single particle operators $A = h = \sum_{i=1}^{N'} h_i = \sum_{i=1}^{N'} \left( \frac{1}{2} \Delta_i + \sum_{j=1}^{M} \frac{-Z_j}{|x_i - R_j|} \right) = \sum_{i=1}^{N'} \left( \frac{1}{2} \Delta_i + V_{\text{core}}(x_i) \right), \; h_i = h_j,$

$$\langle i | h | j \rangle := \langle \varphi_i, h \varphi_j \rangle$$

a) Two particle operators $G = \sum_{i}^{N'} \sum_{j>i}^{N'} \frac{1}{|x_i - x_j|}, \; H = h + G$

$$\langle a, b | i, j \rangle := \sum_{s, s' = \pm \frac{1}{2}} \int \int \frac{\varphi_a^*(x, s) \varphi_b^*(x', s') \varphi_i(x, s) \varphi_j(x', s')}{|x - x'|} \; dx \; dx'$$

$$\langle a, b || i, j \rangle := \langle a, b | i, j \rangle - \langle a, b | j, i \rangle.$$
Slater-Condon Rules

Single particle operators: $\psi^1 = \psi_{SL} = \psi[\ldots, \nu_i, \nu_j, \ldots]$

1) $\psi^1 = \psi^2 =: \psi$ 
   $\psi^1 = \psi[\ldots, \nu_i, \nu_j, \ldots]$ 
   $\psi^2 = \psi[\ldots, \nu_i, \nu_j, \ldots]$ 
   $\langle \psi, h\psi \rangle = \sum_{l=1}^N \langle l | h | l \rangle$

2) $\psi^2 = X^a_j \psi^1$ 
   $\psi^1 = \psi[\ldots, \nu_i, \nu_j, \ldots]$ 
   $\psi^2 = \psi[\ldots, \nu_i, \nu_j, \ldots]$ 
   $\langle \psi^2, h\psi^1 \rangle = \langle a | h | j \rangle$

3) $\psi^1 = X_{i,j}^{a,b} \psi^2$ 
   or higher excitations 
   $\psi^1 = \psi[\ldots, \nu_i, \nu_j, \ldots]$ 
   $\psi^2 = \psi[\ldots, \nu_a, \nu_b, \ldots]$ 
   $\langle \psi^2, h\psi^1 \rangle = 0$

Proof *exercise Hint: Leibniz formula +

$$\langle \psi^2, h_i \psi^1 \rangle = \int \varphi_a(x_i)(h_i\varphi_i)(x_i) \cdot \int \varphi_b(x_k)\varphi_j(x_k) \int \varphi_m(x_1)\varphi_m(x_1) \ldots = \langle a | h | l \rangle \delta_{b,j}$$
Slater-Condon Rules for two particle operators:

1) \( \psi_1 = \psi^2 \)
   \[ \psi^1 = \psi[\ldots, \nu_i, \nu_j, \ldots] \]
   \[ \psi^2 = \psi[\ldots, \nu_i, \nu_j, \ldots] \]
   \[ \langle \psi^1, G\psi^1 \rangle = \frac{1}{2} \sum_{i=1}^{N} \sum_{j=1}^{N} \langle ij||ij \rangle \]

2) \( \psi^2 = X^a_j \psi^1 \)
   \[ \psi^1 = \psi[\ldots, \nu_i, \nu_j, \ldots] \]
   \[ \psi^2 = \psi[\ldots, \nu_i, \nu_a, \ldots] \]
   \[ \langle \psi^2, G\psi^1 \rangle = \sum_{i=1}^{N} \langle ji||ai \rangle \]

3) \( \psi^1 = X^{a,b}_{i,j} \psi^2 \)
   \[ \psi^1 = \psi[\ldots, \nu_i, \nu_j, \ldots] \]
   \[ \psi^2 = \psi[\ldots, \nu_a, \nu_b, \ldots] \]
   \[ \langle \psi^2, G\psi^1 \rangle = \langle ij||ab \rangle \]

3) \( \psi^1 = X^{a,b,c}_{i,j,l} \psi^2 \)
   \[ \psi^1 = \psi[\ldots, \nu_i, \nu_j, \nu_l, \ldots] \]
   \[ \psi^2 = \psi[\ldots, \nu_a, \nu_b, \nu_c, \ldots] \]
   \[ \langle \psi^2, G\psi^1 \rangle = 0 \]

or higher excitations

with

\[ \langle a, b|i, j \rangle := \sum_{s, s' = \pm \frac{1}{2}} \int \int \frac{\varphi_a^*(x, s)\varphi_b^*(x', s')\varphi_i(x, s)\varphi_j(x', s')}{|x - x'|} \ dxdx' \]

\[ \langle a, b||i, j \rangle := \langle a, b|i, j \rangle - \langle a, b|j, i \rangle. \]
Spin functions and spatial orbitals

Spin orbital function: $\varphi(x, s) \in \mathbb{R}, (\mathbb{C}), x \in \mathbb{R}^3 \ s = \pm \frac{1}{2}$, spin functions $\chi$ and spatial orbital functions $\phi$:

$$\varphi(x, s) = \phi_\alpha(x) \chi_\alpha(s) + \phi_\beta(x) \chi_\beta(s)$$

with spin functions $\chi_\alpha(\frac{1}{2}) = 1, \chi_\alpha(-\frac{1}{2}) = 0, \chi_\beta(s) = 1 - \chi_\alpha(s)$

$$\varphi^\alpha(x, s) = \phi_\alpha(x) \chi_\alpha(s)$$

UHF (Unrestricted Hartree Fock, single spin state orbitals) :

$$\phi_{\alpha,i}, \phi_{\beta,j}, \ N' = N_\alpha + N_\beta.$$  

Closed shell RHF (Restricted Hartree Fock) :

$$\phi_{\alpha,i} = \phi_{\beta,i} = \phi_i, \ i = 1, \ldots, N = \frac{N'}{2}.$$
CI Configuration Interaction Method

Approximation space for (spin) orbitals $(x_j, s_j) \rightarrow \varphi(x_j, s_j)$

$$\mathcal{X}_h := \text{span} \{ \varphi_i : i = 1, \ldots, N \} , \quad \langle \varphi_i, \varphi_j \rangle = \delta_{i,j}$$

E.g. Canonical orbitals $\varphi_i, i = 1, \ldots, N$ are eigenfunctions of the discretized single particle operator (e.g Fock operator)

$$\mathcal{F} := \mathcal{F}_h = \sum_{k=1}^{N} F_k : \mathcal{V}_{FCI} \rightarrow \mathcal{V}_{FCI} ,$$

$$\langle F \varphi_i - \lambda_i \varphi_i, \phi_h \rangle = 0 \forall \phi_h \in \mathcal{X}_h$$

Full CI (for benchmark computations $\leq N = 18$) is a Galerkin method w.r.t. the subspace

$$\mathcal{V}_{FCI} = \bigwedge_{i=1}^{N} \mathcal{X}_h = \text{span}\{ \psi_{SL} = \psi[\nu_1, \ldots \nu_N] = \frac{1}{N!} \text{det}(\varphi_{\nu_i}(x_j, s_j))_{i,j=1}^{N} \}$$
CI Configuration Interaction Method

\( \mathcal{N} \) (discrete) eigenfunction \( \varphi_i \), \( \langle F \varphi_i - \lambda_i \varphi_i, \phi_h \rangle = 0 \ \forall \phi_h \in \mathcal{X}_h \)

The first \( N \) eigenfunctions \( \varphi_i \) are called \textbf{occupied orbitals} the others are called \textbf{unoccupied orbitals} (traditionally)

\[ \varphi_1, \cdots, \varphi_N, \varphi_{N+1}, \cdots, \varphi_N \]

Galerkin ansatz: \( \Psi = c_0 \Psi_0 + \sum_{\nu \in J} c_\nu \Psi_\nu \)

\[ \mathbf{H} = \langle \psi_\nu', H \psi_\nu \rangle, \quad \mathbf{Hc} = E \mathbf{c} \quad , \dim \mathcal{V}_h = \binom{N}{N} \]

The matrix coefficients of \( \mathbf{H} = \langle \psi_\nu', H \psi_\nu \rangle \) can be computed by Slater-Condon-rules. (sparse matrix)
Configuration Interaction Method

**Assumption:** For any $\varphi \in H^1$ there exist $\varphi_h \in \mathcal{X}_h$ such that $\|\varphi_h - \varphi\|_{H^1} \to 0$, if $h \to 0$ (roughly: $\lim_{h \to 0} \mathcal{X}_h = H^1(\mathbb{R}^3 \times \{\pm \frac{1}{2}\})$)

**Theorem**

Let $E_0$ be a single eigenvalue and $H\psi = E_0\psi$ and $E_{0,h}$, $\psi_h \in \mathcal{V}_h \subset \mathcal{V}_{FCI}$ be the Galerkin solution, then for $h < h_0$ hold

$$\|\psi - \psi_h\|_{\mathcal{V}} \leq c \inf_{\phi_h \in \mathcal{V}_h} \|\psi - \phi_h\|_{\mathcal{V}}$$

$$E_{0,h} - E_0 \leq C \inf_{\phi_h \in \mathcal{V}_h} \|\psi - \phi_h\|^2_{\mathcal{V}}.$$

Since $\dim \mathcal{V}_h = \mathcal{O}(N^N)$, (curse of dimension), the full CI method is infeasible for large $N$ or $N$!!!!
Binary tensorization,
Fock spaces and second quantization
Configuration Space

Consider a (complete) orthonormal basis \( \{ \varphi_i : i = 1, \ldots, d \} \)
(e.g. by MCSCF)

\[
\mathcal{X}_h := \text{span} \{ \varphi_i : i = 1, \ldots, d \} \subset \mathcal{X} = H^1(\mathbb{R}^3, \pm \frac{1}{2})
\]

ONB of antisymmetric functions by Slater determinants

\[
\psi_{SL}[k_1, \ldots, k_N](x_1, s_1; \ldots; x_N, s_N) := \frac{1}{\sqrt{N!}} \det(\varphi_{k_i}(x_j, s_j))_{i,j=1}^N
\]

Curse of dimensionality \( \dim \mathcal{V}_{FCl}^N = \binom{d}{N} ! \)

For a discrete operator \( H : \mathcal{V}_{FCl}^N =: \mathcal{V} \to \mathcal{V}' \)

\[
H\psi = \sum_{\nu', \nu} \langle \psi_{\nu'}, H\psi_{\nu} \rangle c_{\nu} \psi_{\nu'} = \sum_{\nu'} (Hc)_{\nu'} \psi_{\nu'}. 
\]
Fock space

Let $\Psi_\mu := \Psi_{SL}[\Phi_{k_1}, \ldots, \Phi_{k_N}] = \Psi[k_1, \ldots, k_N]$ basis Slater det.
Labeling of indices $\mu \in \mathcal{I}$ by an binary string of length $d$

$$\mu = (0, 0, 1, 1, 0, \ldots) =: \sum_{i=0}^{d-1} \mu_i 2^i, \ \mu_i = 0, 1$$

- $\mu_i = 1$ means $\Phi_i$ is (occupied) in $\Psi[\ldots]$.
- $\mu_i = 0$ means $\Phi_i$ is absent (not occupied) in $\Psi[\ldots]$.

(discrete) Fock space $\mathcal{F}_d$ is of dim $\mathcal{F}_d = 2^d$, ($\mathbb{K} := \mathbb{C}, \mathbb{R}$)

$$\mathcal{F}_d := \bigoplus_{N=0}^{d} \mathcal{V}^N_{FCI} = \{\Psi : \Psi = \sum_{\mu} c_{\mu} \Psi_{\mu}\}$$

$$\mathcal{F}_d \simeq \{\mathbf{c} : \mu \mapsto \mathbf{c}(\mu_0, \ldots, \mu_{d-1}) = c_{\mu} \in \mathbb{K}, \ \mu_i = 0, 1\} = \bigotimes_{i=1}^{d} \mathbb{K}^2$$

This is a basis dependent formalism $\Rightarrow$ : Second Quantization
Second quantization

Second quantization: annihilations operators:

\[ a_j \Psi[j, 1, \ldots, N] = \Psi[1, \ldots, N] \]

and \( = 0 \) if \( j \) not apparent in \( \Psi[\ldots] \).

Sign-normalization: \( j \) appears in the first place in \( \Psi[j, 1, \ldots, N] \).

The adjoint of \( a_b \) is a creation operator \( a_b^\dagger \)

\[ a_b^\dagger \Psi[1, \ldots, N] = \Psi[b, 1, \ldots, N] = (-1)^N \Psi[1, \ldots, N, b] \]

Lemma

\[ a_k a_l = -a_l a_k , \quad a_k^\dagger a_l^\dagger = -a_l^\dagger a_k^\dagger , \quad a_k^\dagger a_l + a_l a_k^\dagger = \delta_{k,l} \]
Discrete annihilation and creation operators

\[ A := \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix}, \quad A^T = \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} \]

In order to obtain the correct phase factor, we define

\[ S := \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \]

and the discrete annihilation operator

\[ a_p \sim A_p := S \otimes \ldots \otimes S \otimes A_{(p)} \otimes I \otimes \ldots \otimes I \]

where \( A_{(p)} \) means that \( A \) appears on the \( p \)-th position in the product.

The creation operator

\[ a_p^\dagger \sim A_p^T := S \otimes \ldots \otimes S \otimes A_{(p)}^T \otimes I \otimes \ldots \otimes I \]
Schrödinger operator

One and two electron integrals

\[
h^p_q := \langle \phi_q, (\frac{-1}{2} \Delta - V_{\text{core}}) \phi_p \rangle, \quad p, q, r, s = 1, \ldots, d,
\]

\[
g^{p,q}_{r,s} := \frac{1}{2} \langle \phi_r(x, s_1) \phi_s(y, s_2), \frac{\phi_p(x, s_1) \phi_q(y, s_2)}{|x-y|} \rangle
\]

Theorem (Slater -Condon, (HRS (2010)))

The Galerkin matrix \( H \) of electronic Schrödinger Hamiltonian is sparse and can be represented by

\[
H = \sum_{p,q=1}^{d} h^q_p A^T_p A_q + \sum_{p,q,r,s=1}^{d} g^{p,q}_{r,s} A^T_r A^T_s A_p A_q.
\]

Modification, treating spin explicitly, (spin symmetries and other symmetries could be enforced (Legeza et al. ))

\[
\mathcal{F}_d = \bigotimes_{i=1}^{d} K^4, \quad (K = \mathbb{C}, \mathbb{R})
\]
Particle number operator and Schrödinger eqn.

\[ P := \sum_{p=1}^{d} a_p^\dagger a_p , \quad \simeq \quad P := \sum_{p=1}^{d} A_p^T A_p . \]

The space of \( N \)-particle states is given by

\[ \mathcal{V}^N := \{ \mathbf{c} \in \bigotimes_{i=1}^{d} \mathbb{K}^2 : P\mathbf{c} = N\mathbf{c} \} . \]

Variational formulation of the Schrödinger equation

\[ \mathbf{c} = (c(\mu)) = \arg\min \{ \langle H\mathbf{c}, \mathbf{c} \rangle : \langle \mathbf{c}, \mathbf{c} \rangle = 1, P\mathbf{c} - N\mathbf{c} = 0 \} . \]

Approximate

\[ \mathbf{c} \in \bigotimes_{i=1}^{d} \mathbb{K}^2 \simeq \mathbf{c}_e \in \mathcal{M}_r (\mathbb{T}_r) \]

in hierarchical tensor format (e.g. MPS) by means of e.g. (M)ALS \( \Rightarrow \) QC DMRG
(quantum chemistry) QC DMRG
Separation into (two) subsystems $A_i, B_i$

$\mathcal{F}_{A_i} = \text{generated by } \{\varphi_1 \ldots \varphi_i\}, \quad \mathcal{F}_{B_i} = \text{generated by } \{\varphi_{i+1} \ldots \varphi_d\}$

SVD factorization of $U_{x_1,\ldots,x_i}^{x_{i+1}}$ (E. Schmidt). $r_i$ is a measure for the entanglement of the two subsystems $A_i, B_i, i = 1, \ldots, d - 1$. If $A_i, B_i$ are independent $\rightarrow r_i = 1$ - size consistency.

Working assumption: $r_i$ moderate, and truncation error small!

Deficiencies:

1. representation depends on the ordering of the basis functions (variables)
2. representation depends on the choice basis functions (molecular, natural, localized orbitals)
3. scaling $O(r^2)$. 
Discussion

Consequences having an MPS approximation:

1. represents a FCI wave function
2. permeates the full FCI space
3. complexity (storage): $2 \sum_{i} d_{i} r_{i-1} r_{i} = O(r^2 d)$
4. electron density and reduced density matrices can be computed $\Rightarrow$ gradients, forces, ...
5. subspaces, mixed states, (e.g. multiple eigenvalues, degeneracy and near degeneracy) can be computed as well
6. size consistency and black box (yes and no)

I cannot (will not) answer the open questions:

1. for which systems the working assumption holds?
2. are there better tensor formats (e.g. HT or tensor networks)?
(of course $r_i < r_c$ usually $r_i << r_c$)

Instead we consider the questions

1. is the TT or MPS parametrization stable? (yes) can we remove redundancy completely? (yes)
2. how to compute the MPS approximation of the FCI wave function?
QC-DMRG and TT resp, MPS approximations

In courtesy of O Legeza (Hess & Legeza & ..)

LiF dissociation, 1st + 2nd eigenvalue
No approximation so far except FCI! How to extract essential information of a large space dim $\sim 2^d$ ($d >> 10$)?

1. HF wave function: Let $\mathbb{K}^2 = \text{span}\{e^0, e^1\}$, a single Slater determinant is rank 1 in $\bigotimes_{i=1}^{d} \mathbb{K}^2$, e.g.

$$\psi[1, \ldots, N] \simeq e^1_{(0)} \otimes \ldots \otimes e^1_{(N)} \otimes e^0_{(N+1)} \otimes \ldots \otimes e^0_{(d)} \in \mathcal{V}^N.$$ 

2. sparsity: most determinants could be neglected

$$\{\mu : c_\mu \approx 0\}, \text{ number of nonzero amplitudes } NNZ << 2^d,$$

$\rightarrow$ canonical rank $r_c \leq NNZ$.

3. exponential parametrization (CC), using

$$\text{excitation operators } X^v_o = a^{\dagger}_v a_o.$$ 

with $NNZ << 2^d$.

4. Tensor product approximation In particular in TT format (MPS)
Separation into (two) subsystems $A_i, B_i$

\[ F_{A_i} \text{ generated by } \{\varphi_1 \ldots \varphi_i\}, \quad F_{B_i} \text{ generated by } \{\varphi_{i+1} \ldots \varphi_d\} \]

SVD factorization of $U_{x_1,\ldots,x_i}^{x_{i+1}}$ (E. Schmidt). $r_i$ is a measure for the entanglement of the two subsystems $A_i, B_i, i = 1, \ldots, d - 1$. If $A_i, B_i$ are independent $\rightarrow r_i = 1$ - size consistency.

Working assumption: $r_i$ moderate, and truncation error small!

Deficiencies:
1. representation depends on the ordering of the basis functions (variables)
2. representation depends on the choice basis functions (molecular, natural, localized orbitals)
3. scaling $\mathcal{O}(r^2)$. 
II.

Optimization with TT tensors or MPS
- DMRG algorithm
results
Optimization problems

Tensor optimization
Approximation of $W \in \mathcal{H}$: by a TT tensor of rank $r$

$$U = \arg\min \left\{ F(V) = \frac{1}{2} \langle V - W, V - W \rangle : V \in T_r \right\} .$$

Linear elliptic equations

$$U = \arg\min \left\{ F(V) = \frac{1}{2} \langle AV, V \rangle - \langle B, V \rangle : V \in T_r \right\} .$$

eigenvalue problems

$$U = \arg\min \left\{ F(V) = \langle AV, V \rangle : \langle V, V \rangle = 1 : V \in T_r \right\} .$$
**Optimization Problems in Matrix Product States (MPS)**

TT- tensor $U = \sum_k \bigotimes_{\nu=1}^d u_{k,\nu}^{\nu-1}$

Tensor optimization

Approximation of $W \in \mathcal{H}$: by MPS of rank $r$

$$U = \arg\min \{ F(V) = \frac{1}{2} \langle V - W, V - W \rangle : V \in \mathcal{T}_r, (P - N)V = 0 \}.$$  

Linear elliptic equations

$$U = \arg\min \{ F(V) = \frac{1}{2} \langle AV, V \rangle - \langle B, V \rangle : V \in \mathcal{T}_r, (P - N)V = 0 \}.$$ 

eigenvalue problems

$$U = \arg\min \{ F(V) = \langle AV, V \rangle : \langle V, V \rangle = 1 : V \in \mathcal{T}_r, (P - N)V = 0 \}.$$
Optimization by relaxation in TT format ALS

Alternating Linear Scheme - ALS

Relaxation (see e.g. Gauss-Seidel, ALS):
For $j = 1, \ldots, d$:

1. fix all component tensors $U_\nu$, $\nu \in \{1, \ldots, d\}\setminus \{j\}$, except index $j$.

\[ P_{i,1,U} : \begin{array}{c}
\begin{array}{ccc}
\text{r}_2 & \text{r}_3 \\
n_3
\end{array}
\end{array} \rightarrow \begin{array}{c}
\begin{array}{cccc}
\text{U}_1 & \text{U}_2 & \text{U}_3 & \text{U}_4 & \text{U}_5 \\
n_1 & n_2 & n_3 & n_4 & n_5
\end{array}
\end{array} \]

2. Optimize $U_j(k_{j-1}, x_j, k_j)$, and orthogonalize left
Repeat the relaxation procedure (in the opposite direction.)

Relaxation scheme $U(x_\nu)^{k_\nu}_{k_{\nu}-1}$

This is NOT exactly the DMRG algorithm!
A modified relaxation algorithm MALS

Disadvantage of simple relaxation:
1. $U_j, \ldots$ must be (left)-orthogonalized,
2. $r = (r_1, \ldots r_d)$ is fixed, but we want an adaptive rank selection

Modified Alternating Linear Scheme MALS (DMRG)

Idea: optimize comprised components

$$U_{i,i+1} =: \left[ W(k_{j-1}, x_j, x_{j+1}, k_{j+1}) \right] \approx \sum_{k_j=1}^{r_j} \left[ U(x_j) \right]_{k_j}^{k_j} \left[ V(x_{j+1}) \right]_{k_j}^{k_{j+1}} \in \mathbb{R}^{r_{i-1} n_i n_{i+1} r_{i+1}}.$$
Retractions, extensions and density matrices

Let $W_i(t) \in \mathbb{R}^{r_i-1 \cdot n_i \cdot r_i}$ be of the size of a component tensor.
The extension or dressing operator

$$E_i(t) := E_i(U(t)) = E_{i,1}(t),$$

extending $W_i$ to a tensor in $\mathcal{H} = \bigotimes_{i=1}^{d} \mathbb{R}^{n_i}$, is defined by

$$W_i(t) \mapsto E_i(t)W_i(t) := U_1(t) \cdots W_i(t) \cdots U_d(t).$$

The adjoint operator is $E_i^T$. The left inverse $E_i^\dagger$ applied to the
tensor $U = U_1 \cdots U_i \cdots U_d \in \mathbb{T}_r$ is given by

$$U_i = E_i^\dagger U = (E_i^T E_i)^{-1} E_i^T U.$$

If $U_i, i = 1, \ldots, d - 1$, are left orthogonal, the $i$–th density matrix will be defined by

$$I \otimes D_i := (E_i^T E_i)^{-1}. $$
Left part tensor $L_i(k_{i-1})$ (resp. $R_i$ right part) is pointwise given by

$$[L_i(k_{i-1})](x_1, \ldots, x_{i-1}) = \sum_{(k_1 \ldots k_{i-2})=1}^{r_1 \ldots r_{i-2}} U_1(k_0, x_1, k_1) \cdots U_{i-1}(k_{i-2}, x_{i-1}, k_{i-1}).$$

$$U = \sum_{k_{i-1}=1}^{r_{i-1}} \sum_{k_i=1}^{r_i} L_i(k_{i-1}) \otimes U_i(k_{i-1}, x_i, k_i) \otimes R_i(k_i),$$

For given tensor $U \in \bigotimes_{i=1}^d \mathbb{R}^{n_i}$ We define a linear operator

$$E_{i,j}^T := E_{i,j}(U) : \bigotimes_{i=1}^d \mathbb{R}^{n_i} \rightarrow \mathbb{R}^{r_{i-1}n_i} \otimes \mathbb{R}^{n_i+1} \otimes \cdots \otimes \mathbb{R}^{n_{i+j-1}r_{i+j-1}}.$$

by setting

$$E_{i,j}^T := L_i \otimes I_{n_i} \otimes \cdots \otimes I_{n_{i+j-1}} \otimes R_{i+j} \underbrace{\otimes \cdots \otimes \otimes}_{j\text{--times}} R_{i+j}$$

The adjoint operator extends a component $V_i$ to the tensor

$$E_{i,1} V_i = U_1 \cdots U_{i-1} V_i U_{i+1} \cdots U_d.$$
Relaxation TT-ALS algorithm

**Require:** Functional : \( \mathbb{R}^{n_1 \times \cdots \times n_d} \rightarrow \mathbb{R} \), component starting vector \((U_1, \ldots, U_d)\).

Right-orthogonalise the components \((U_2, \ldots, U_d)\).

while termination criterion is not fulfilled do
  for \( i = 1, \ldots, d - 1 \) do
    Find \( \hat{U}_i = \arg\min J \circ P_{i,1} : \mathbb{R}^{r_{i-1} \times n_i \times r_i} \rightarrow \mathbb{R} \).
    Set \( U_i, U_{i+1} \leftarrow \hat{U}_i, U_{i+1} \), by applying an orthogonality method.
  end for
  for \( i = d, \ldots, 2 \) do
    Find \( \hat{U}_i = \arg\min P_{i,1} \circ J : \mathbb{R}^{r_{i-1} \times n_i \times r_i} \rightarrow \mathbb{R} \).
    Set \( U_i, U_{i-1} \leftarrow \hat{U}_i, U_{i-1} \), by applying an orthogonality method.
  end for
end while
Modified relaxation TT- MALS algorithm (DMRG).

**Require:** Functional : \( \mathbb{R}^{n_1 \times \ldots \times n_d} \rightarrow \mathbb{R} \), component starting vector \((U_1, \ldots, U_d)\).
Right-orthogonalise the components \((U_2, \ldots, U_d)\).

**while** termination criterion is not fulfiled **do**

for \(i = 1, \ldots, d-1\) **do**

(Optimization step) Find

\([k_{i-1}, x_i, x_{i+1}, k_{i+1}] \mapsto W_i(k_{i-1}, x_i, x_{i+1}, k_{i+1})\) where

\[ W_i = \arg\min J \circ P_{i,2} : \mathbb{R}^{r_{i-1} \times n_i n_{i+1} \times r_{i+1}} \rightarrow \mathbb{R}. \]

(Decimation step) Approximate with low rank \(r_i\)

\[
[W_{x_{i+1}, k_{i+1}}^{k_{i-1}, x_i}] \approx [U_{k_{i-1}, x_i}^{k_i}] [V_{x_{i+1}, k_{i+1}}^{k_i}].
\]

end for

Repeat with reverse order

**end while**
ALS - Stationary conditions (equations)

For fixed $j$, and $Ax = B$ the stationary condition is

\[ \tilde{A}_j U_j = E_{j,1}^T A E_{j,1} U_j = b_j = E_{j,1} B. \]

For the eigenvalue problem

\[ \tilde{A}_j U_j = E_{j,1}^T A E_{j,1} U_j = \lambda \tilde{M}_j U_j = \lambda E_{j,1}^T E_{j,1} U_j. \]

For MALS we obtain

\[ \tilde{A}_j = E_{j,2}^T A E_{j,2}, \quad \tilde{M}_j = E_{j,2}^T E_{j,2}. \]

Lemma (Rohw. & Holtz & S.)

*If* $U_j$, $j < i$ are left-, and $U_j$, $i < j$ are right-orthogonal *then*

\[ \text{cond} \tilde{A}_i \leq \text{cond} A \text{ and } \tilde{M}_i = I! \]

Remark: det $A_i \neq 0$ can hold, for some $A$ when det $A = 0$ (-RIP, compressed sensing)
ALS - Solution scheme for linear equations $Ax = b$

- Proceeding in “sweeps”: Solution of successive linear systems
- additional orthogonalisation

**sweep**

1st MIS

\[ P_{1,1}^T A_{1,1} u = P_{1,1}^T b \]

2nd MIS

\[ P_{2,1}^T A_{2,1} u = P_{2,1}^T b \]

\[ P_{2,1}^T A_{2,1} u = P_{2,1}^T b \]

\[ P_{d,1}^T A_{d,1} u = P_{d,1}^T b \]

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\[ P_{d,1}^T A_{d,1} u = P_{d,1}^T b \]
MALS - Solution scheme for linear equations $Ax = b$ (inspired by the DMRG algorithm)

- Proceeding in “sweeps”: Solution of successive linear systems for comprised components $U_{i,i+1} \in \mathbb{R}^{r_i-1 \times n_i \times n_{i+1} \times r_{i+1}}$, other components fixed
- Subsequent SVDs restore orthogonal TT-format
Example: Eigenvalue problem in QTT

in courtesy of B. Khoromskij, I. Oseledets, *QTT: Toward bridging high-dimensional quantum molecular dynamics and DMRG methods*,

\[ H\psi = \left(-\frac{1}{2}\Delta + V\right)\psi = E\psi \]

with potential energy surface given by Henon-Heiles potential

\[ V(q_1, \ldots, q_f) = \frac{1}{2} \sum_{k=1}^{f} q_k^2 + \lambda \sum_{k=1}^{f-1} \left( q_k q_{k+1} - \frac{1}{3} q_k^3 \right). \]

Dimensions \( f = 4, \ldots, 256; \) 1-D grid size \( n = 128 = 2^7 = 2^d; \) \( \sim \) QTT-tensors \( \in \bigotimes_{i=1}^{7f} \mathbb{R}^2 = \mathbb{R}^{\underbrace{2 \times \ldots \times 2}_{7f=1792}}. \)
Thank you
for your attention.

References:


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