

MULTI-CONFIGURATION TIME DEPENDENT HARTREE THEORY

A TENSOR NETWORK PERSPECTIVE

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Outline

Discrete Variable Representation

Multi-Configuration Time Dependent Hartree

MCTDH vs. DMRG

DISCRETE VARIABLE REPRESENTATION

DISCRETE VARIABLE REPRESENTATION

PROBLEM DESCRIPTION

Non-Relativistic Multi-Dimensional Problem

- TDSE:

$$i\hbar \frac{\partial}{\partial t} |\Psi(\vec{q}, t)\rangle = H |\Psi(\vec{q}, t)\rangle$$

- TISE:

$$H |\Psi(\vec{q}, t)\rangle = E |\Psi(\vec{q}, t)\rangle$$

where

$$H = T + V$$

and $\vec{q} = (q_1, q_2, \dots, q_d)$.

DISCRETE VARIABLE REPRESENTATION

STANDARD PROCEDURE

Discretization: Spectral Method

Choose a set of basis functions: $\{|\Phi_I(\vec{q})\rangle\}_{I=1}^N$ and require

1. Orthonormal: $\langle\Phi_I|\Phi_J\rangle = \delta_{IJ}$
2. Complete: $\sum_{I=1}^n |\Phi_I\rangle\langle\Phi_I| = P \rightarrow 1, \quad n \rightarrow +\infty$

Therefore,

$$|\Psi\rangle \approx P|\Psi\rangle = \sum_I^n c_I |\Phi_I\rangle \quad \text{where} \quad c_I = \langle\Phi_I|\Psi\rangle$$

and

$$A \approx PAP = \sum_{IJ}^n |\Phi_I\rangle A_{IJ} \langle\Phi_J| \quad \text{where} \quad A_{IJ} = \langle\Phi_I|A|\Phi_J\rangle$$

1-D Problem

Standard procedure:

1. Choose a set of 1-D basis $\{|\varphi_i(x)\rangle\}_{i=1}^n$

2. Integrate

$$T_{ij} = \langle \varphi_i(x) | T | \varphi_j(x) \rangle = -\frac{\hbar}{2\mu} \int \varphi_i^*(x) \frac{d}{dx} \varphi_j(x) dx,$$

$$V_{ij} = \langle \varphi_i(x) | V | \varphi_j(x) \rangle = \int \varphi_i^*(x) V(x) \varphi_j(x) dx, \text{ and}$$

$$H_{ij} = T_{ij} + V_{ij}$$

3. Solve TDSE/TISE in matrix form

• TDSE:

$$i\hbar \dot{\mathbf{c}} = \sum_j H_{ij} c_j \quad \text{or} \quad i\hbar \dot{\mathbf{c}} = \mathbf{H} \mathbf{c}$$

• TISE:

$$\sum_j H_{ij} c_j = E c_i \quad \text{or} \quad \mathbf{H} \mathbf{c} = E \mathbf{c}$$

d -D Problem: $\vec{q} = (q_1, \dots, q_d)$

Standard procedure:

1. Choose a set of d -D basis $\{|\Phi_I(\vec{q})\rangle\}_{I=1}^N$
 - Use the direct product of d sets of 1-D basis

$$\bigotimes_{\kappa=1}^d \left\{ \left| \varphi_{i_\kappa}^{(\kappa)}(q_\kappa) \right\rangle \right\}_{i_\kappa=1}^{n_\kappa}$$

as the set of d -D basis, i. e.,

$$|\Phi_I(q_1, \dots, q_d)\rangle = \prod_{\kappa=1}^d \left| \varphi_{i_\kappa}^{(\kappa)}(q_\kappa) \right\rangle$$

where $I = (i_1, \dots, i_d)$.

d -D Problem: $\vec{q} = (q_1, \dots, q_d)$

Standard procedure:

1. Choose a set of d -D basis $\{|\Phi_I(\vec{q})\rangle\}_{I=1}^N$, where
$$|\Phi_I(\vec{q})\rangle = \prod_{\kappa=1}^d \left| \varphi_{i_{\kappa}}^{(\kappa)}(q_{\kappa}) \right\rangle$$
2. Integrate $H_{IJ} = T_{IJ} + V_{IJ}$
3. Solve TDSE/TISE in matrix form
 - TDSE: $i\hbar\dot{\mathbf{c}} = \mathbf{H}\mathbf{c}$
 - TISE: $\mathbf{H}\mathbf{c} = E\mathbf{c}$

DISCRETE VARIABLE REPRESENTATION

SELECTION OF BASIS

Motivation of DVR

- How to do the integral in the standard procedure?
- Kinetic energy

In Cartesian coordinate

$$T = -\frac{\hbar}{2} \sum_{\kappa=1}^d \frac{1}{\mu_{\kappa}} \frac{\partial^2}{\partial q_{\kappa}^2}$$

Therefore,

$$\mathbf{T} = \bigotimes_{\kappa=1}^d \mathbf{T}^{(\kappa)} \quad \text{where} \quad T_{ij}^{(\kappa)} = -\frac{\hbar}{2\mu_{\kappa}} \left\langle \varphi_i^{(\kappa)} \left| \frac{\partial^2}{\partial q_{\kappa}^2} \right| \varphi_j^{(\kappa)} \right\rangle$$

which can be integrated analytically once the set of basis is determined.

Motivation of DVR

- How to do the integration in the standard procedure?
 - Kinetic energy
 - Potential energy
Differs in different problems.
- Can we integrate some physical quantity analytically once the set of basis is determined, and transform to the potential matrix without integration?

Fact

If $\langle \Phi_I | \vec{x} | \Phi_J \rangle = \vec{x}_I \delta_{IJ}$ and $A = A(\vec{x})$,
 then $\langle \Phi_I | A | \Phi_J \rangle = A(\vec{x}_I) \delta_{IJ}$.

For a given set of basis functions $\{|\Xi_I(\vec{q})\rangle\}_{I=1}^N$, let
 $\vec{q}_{IJ} = \langle \Xi_I | \vec{q} | \Xi_J \rangle$, then there is a set of basis functions
 $\{|\Phi_I(\vec{q})\rangle\}_{I=1}^N$ s. t.

1. $|\Phi_J\rangle = \sum_{I=1}^N |\Xi_I\rangle U_{IJ}$, where \mathbf{U} is unitary, and
2. $\vec{\mathbf{Q}} = \mathbf{U}^\dagger \vec{\mathbf{q}} \mathbf{U}$, where $\vec{Q}_{IJ} := \langle \Phi_I | \vec{q} | \Phi_J \rangle = \vec{q}_{IJ} \delta_{IJ}$.

- $\{|\Xi_I(\vec{q})\rangle\}_{I=1}^N$: finite basis set representation
- $\{|\Phi_I(\vec{q})\rangle\}_{I=1}^N$: discrete variable representation

In DVR basis, $V_{IJ} = V(\vec{q}_I)\delta_{IJ}$.

- How to calculate $T|\Psi\rangle$?
 - Integrate in FBR and transform in DVR ($\mathcal{O}(N^2)$)
 - Use FFT and its inverse to calculate the differential ($\mathcal{O}(N\log N)$)

Example: (1-D) Sine-DVR Basis

Choose the n -th lowest eigenstates of 1-D infinite square well as FBR:

$$\xi_n(x) = \begin{cases} \sqrt{\frac{2}{L}} \sin(n\pi x/L), & \text{if } 0 \leq x \leq L \\ 0, & \text{else} \end{cases}$$

and in FBR, we have

$$T_{ij}^{(\text{FBR})} = -\frac{\hbar^2}{2\mu} \left(\frac{i\pi}{L} \right)^2 \delta_{ij}$$

Example: (1-D) Sine-DVR Basis

Let $q = \cos(\pi x/L)$, then \mathbf{q} could be diagonalized analytically:

$$U_{ij} = \sqrt{\frac{2}{n+1}} \sin\left(\frac{ij\pi}{n+1}\right)$$

$$Q_{ij} = q_i \delta_{ij} \quad \text{where} \quad q_i = \frac{iL}{n+1}$$

and $\mathbf{Q} = \mathbf{U}^\dagger \mathbf{q} \mathbf{U}$.

Example: (1-D) Sine-DVR Basis

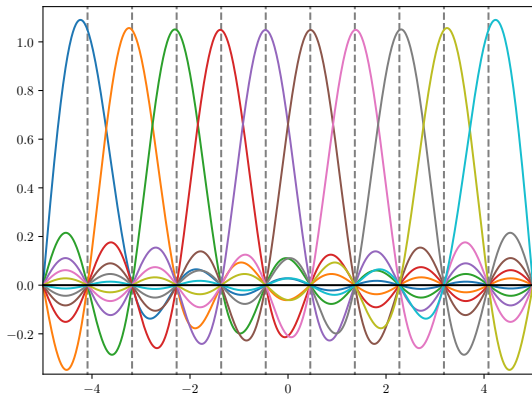


Figure 1: Sine-DVR basis functions ($L = 10$ a. u., $n = 10$)

Example: 1-D Double Well

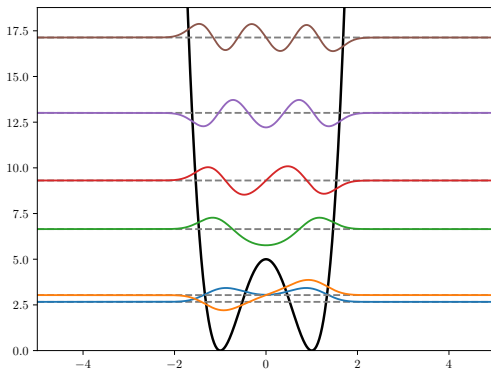


Figure 2: Potential curve and eigenstates

$$V = 5(x^2 - 1)^2, L = 10, n = 400 \text{ (in a. u.)}$$

Example: 1-D Double Well

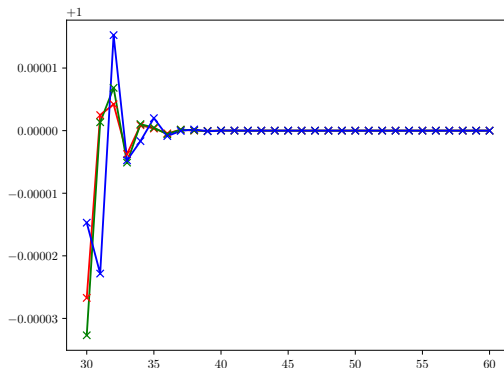


Figure 3: Relative energy to n

Red: ground state; green: 1st excited state; blue: 2nd excited state.

Relative to when $L = 10$, $n = 400$

Example: 1-D Double Well

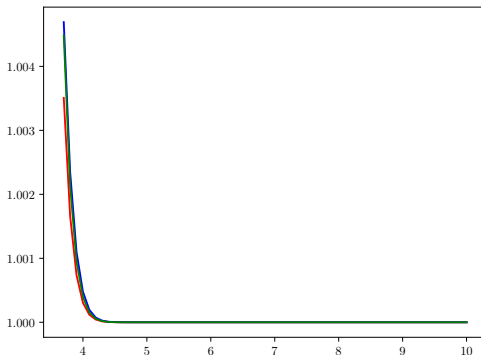


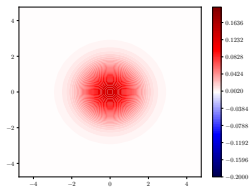
Figure 4: Relative energy to L

Red: ground state; green: 1st excited state; blue: 2nd excited state.
Relative to when $L = 10$, $n = 400$

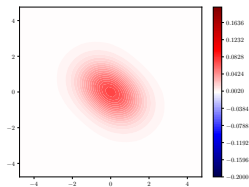
Example: 2-D harmonic oscillator

$$V = \frac{1}{2}x^2 + \frac{1}{2}y^2 + \frac{1}{4}xy, \quad n = 40, \quad L = 10 \quad (\text{in a. u.})$$

Start from the ground state when $V = \frac{1}{2}x^2 + \frac{1}{2}y^2$.



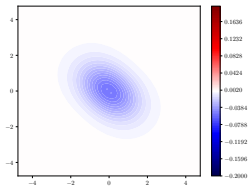
$t = 0.00$ a. u.



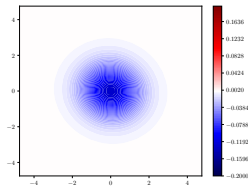
$t = 1.00$ a. u.

Figure 5: Wavefunctions of a 2-D harmonic oscillator at t (using RK45 as ODE solver, $\Delta t = 0.01$ a. u.)

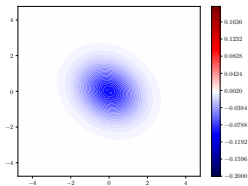
Example: 2-D harmonic oscillator (Cont'd)



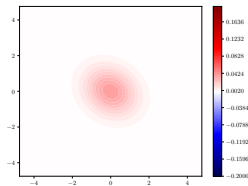
$t = 2.00$ a. u.



$t = 3.00$ a. u.



$t = 4.00$ a. u.



$t = 5.00$ a. u.

Example: 2-D harmonic oscillator

Autocorrelation function: $a(t) = \langle \Psi(0) | \Psi(t) \rangle$, and when initial state $|\Psi(0)\rangle$ is real, $a(t) = \langle \Psi^*(t/2) | \Psi(t/2) \rangle$

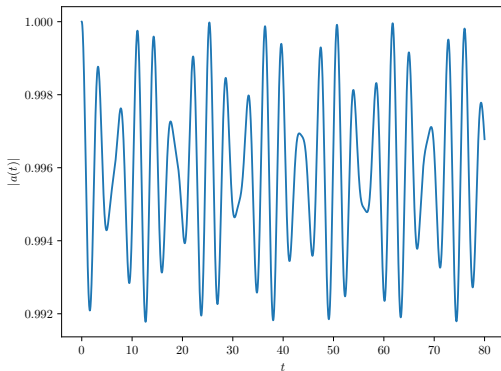


Figure 6: $|a(t)| - t$ (using RK45 as ODE solver, $\Delta t = 0.001$ a. u.)

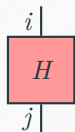
MULTI-CONFIGURATION TIME DEPENDENT HARTREE

Language: Tensor Network Notation

- Tensors



A blue circle labeled A with three external legs: one on the left labeled i , one on the right labeled j , and one on the bottom labeled k . To the right of the circle is the text $:= A_{ijk}$.



A red square labeled H with two external legs: one on the top labeled i and one on the bottom labeled j . To the right of the square is the text $:= H_{ij}$.

- Contraction



Diagram illustrating the contraction of two tensors A and B . On the left, tensor A (blue circle) has legs s (left), t (top), and i (bottom). Tensor B (blue circle) has legs v (top) and u (right). A horizontal line connects the bottom leg i of A to the top leg v of B . To the right of this is the text $:=$. Further right is the expression \sum_i followed by tensor A with legs s (left), t (top), and i (bottom), then a multiplication sign \times , and finally tensor B with legs i (left), v (top), and u (right).

MULTI-CONFIGURATION TIME DEPENDENT HARTREE

MOTIVATION

Express the Standard Procedure in TNN

- Wavefunction

$$\langle \Phi_I | \Psi \rangle = \text{Diagram of a blue circle labeled } A \text{ with } d \text{ incoming arrows labeled } i_1, i_2, \dots, i_d$$

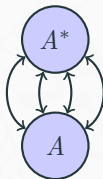
where $i_\kappa \in \{1, \dots, n_\kappa\}$, $\kappa = 1, \dots, d$.

Space for saving a wavefunction: $\prod_{\kappa=1}^d n_\kappa$ floats.

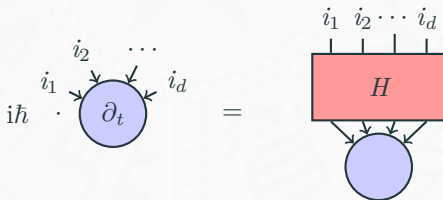
Express the Standard Procedure in TNN

- Normalization condition

$$1 = \langle \Psi | \Psi \rangle \approx \langle \Psi | P | \Psi \rangle =$$

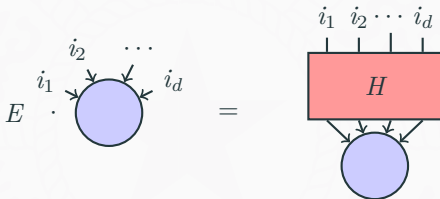


- TDSE



Express the Standard Procedure in TNN

- TISE



where $i_\kappa \in \{1, \dots, n_\kappa\}$, $\kappa = 1, \dots, d$.

Problem: $N = \prod_{\kappa=1}^d n_\kappa$ exponentially increase as d grows.

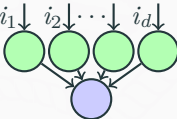
Motivation of MCTDH

Motivation: construct the wavefunction without form the whole space.

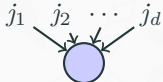
One simple way: Hartree method.

<http://www.pci.uni-heidelberg.de/tc/usr/mctdh/lit/introMCTDH.pdf>

Hartree method in TNN

$$\langle \Phi_I | \Psi \rangle =$$


where


$$\langle \Phi_J^{(1)} | \Psi \rangle =$$


and $j_\kappa \in \{1\}$, $\kappa = 1, \dots, d$.


Space for saving a wavefunction: $\sum_{\kappa=1}^d n_\kappa$ floats.

Hartree method in TNN

Extra normalization condition at the κ -th set of single particle functions (SPFs):

$$\delta_{ij} =$$


The diagram shows two green circles stacked vertically. The top circle contains a black asterisk (*). A vertical line with a downward arrow points from the label i to the top circle. A vertical line with an upward arrow points from the bottom circle to the label j . A double-headed vertical arrow connects the two circles.

$$P_{ij}^{(\kappa)} =$$


The diagram shows two green circles stacked vertically. The top circle is empty. A vertical line with a downward arrow points from the label i to the top circle. The bottom circle contains a black asterisk (*). A vertical line with an upward arrow points from the label j to the bottom circle.

Variational Principle

Solve TISE

$$\delta \left(\begin{array}{c} \text{Diagram 1} \\ \text{Diagram 2} \end{array} - \lambda \left(\begin{array}{c} \text{Diagram 3} \\ \text{Diagram 4} \end{array} - 1 \right) \right) = 0$$

The diagram illustrates the variational principle for solving the Time-Independent Schrödinger Equation (TISE). It shows the variation of the expectation value of the Hamiltonian H minus the energy λ (multiplied by the norm squared of the wavefunction minus 1) set to zero.

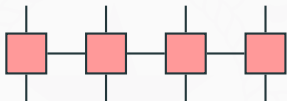
Diagram 1 (Left): A diagram representing the expectation value of the Hamiltonian H . It consists of a central red rectangle labeled H . Above the rectangle are four green circles, each containing an asterisk (*). Below the rectangle are four green circles. Arrows point from the four bottom green circles up to the four top green circles, and from each of these four top green circles up to a single purple circle at the top containing an asterisk (*). Arrows also point from each of the four bottom green circles down to a single purple circle at the bottom containing an asterisk (*).

Diagram 2 (Right): A diagram representing the norm squared of the wavefunction. It consists of a central purple circle at the top containing an asterisk (*). Below it are four green circles, each containing an asterisk (*). Below these are four more green circles. Arrows point from the four bottom green circles up to the four middle green circles, and from each of these four middle green circles up to the top purple circle. Arrows also point from each of the four bottom green circles down to a single purple circle at the bottom containing an asterisk (*).

Only special “structure” of \mathbf{H} can simplify the equation.

Structure of H

- Matrix product operators (MPOs)



- Summation of products of operators

$$\sum_{i=1}^t \left(\begin{array}{c} \text{red square} \\ \text{red square} \\ \text{red square} \\ \text{red square} \end{array} \right)$$

The diagram shows a summation symbol with a superscript t and a subscript $i=1$. Inside the parentheses of the summation are four red square blocks arranged vertically, each with a vertical line extending upwards and another extending downwards.

MULTI-CONFIGURATION TIME DEPENDENT HARTREE

DATA STRUCTURE

Structure of Wavefunction

$$\langle \Phi_I | \Psi \rangle = \begin{array}{c} i_1 \downarrow \quad i_2 \downarrow \quad \cdots \quad i_d \downarrow \\ \text{---} \circ \text{---} \circ \text{---} \circ \text{---} \circ \text{---} \\ \quad \quad \quad \swarrow \quad \downarrow \quad \searrow \\ \quad \quad \quad \circ \end{array}$$

where

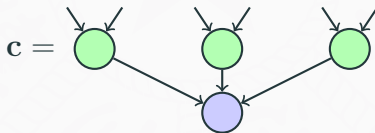
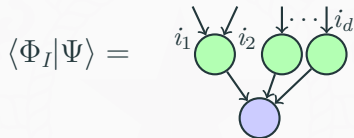
$$\langle \Phi_J^{(1)} | \Psi \rangle = \begin{array}{c} j_1 \quad j_2 \quad \cdots \quad j_d \\ \quad \swarrow \quad \downarrow \quad \searrow \\ \quad \quad \quad \circ \end{array}$$

Idea: $j_\kappa \in \{1, \dots, n_\kappa^{(1)}\}$, $\kappa = 1, \dots, d$, and $n_\kappa^{(1)} < n_\kappa$.

Space for saving a wavefunction: $\prod_{\kappa=1}^d n_\kappa^{(1)} + \sum_{\kappa=1}^d n_\kappa^{(1)} n_\kappa$ floats.

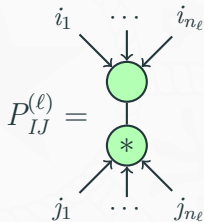
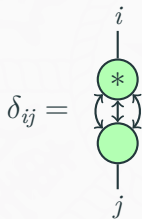
Mode Combination

Examples:



Normalization

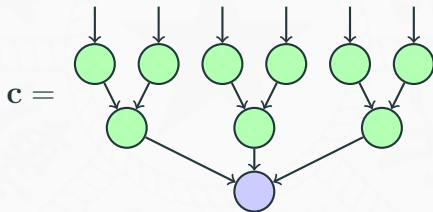
General normalization condition at the ℓ -th node:



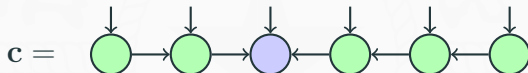
Multi-Layer

More nodes, but smaller ranks and smaller range of indices.

Example 1:



Example 2: matrix product states (MPSs) in DMRG



For a complete binary tree, the space for saving a wavefunction is $\mathcal{O}(dn^3)$ floats, if $n_\ell = \mathcal{O}(n)$ for all ℓ .

MULTI-CONFIGURATION TIME DEPENDENT HARTREE

ALGORITHM

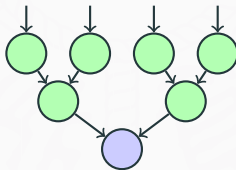
Solve TDSE

Use the same type of \mathbf{H} in Hartree method:

$$\sum_{i=1}^t \left(\begin{array}{c} \text{red square} \\ \text{vertical line} \end{array} \right)$$

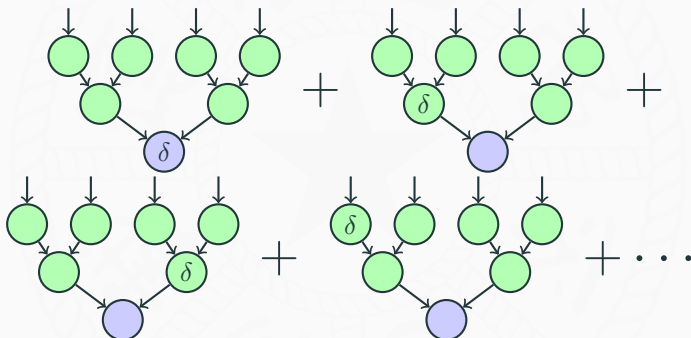
Dirac-Frenkel variational principle: $\langle \delta\Psi | i\hbar \frac{\partial}{\partial t} - H | \Psi \rangle = 0$.

Without loss of generality, let $\mathbf{c} =$



Solve TDSE

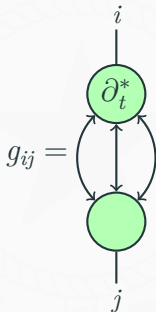
$$\delta \mathbf{c} =$$



Similarly, $\dot{\mathbf{c}}$.

Normalization

In order to hold all general normalization conditions during the propagation, one must have



where \mathbf{g} is a hermitian matrix. For simplicity, choose $\mathbf{g} = \mathbf{0}$.

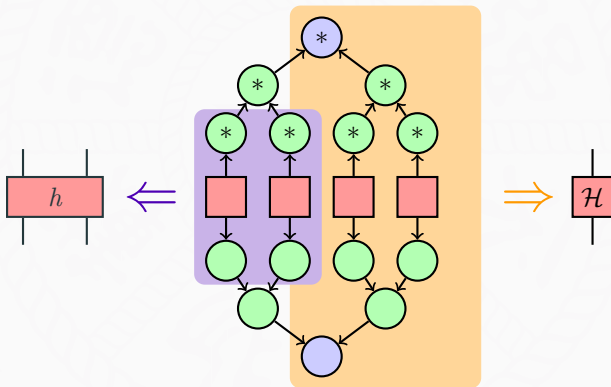
Equations of Motion

$$i\hbar \quad \text{[blue circle with } \partial_t \text{ and two incoming arrows]} = \text{[red box } h \text{ with two incoming arrows and a blue circle below it]}$$

$$i\hbar \quad \text{[green circle with } \partial_t \text{ and two incoming arrows]} = \text{[stack of boxes: } \mathcal{P} \text{ (yellow), } h \text{ (red), } \mathcal{H} \text{ (red), } \rho^{-1} \text{ (yellow)]}$$

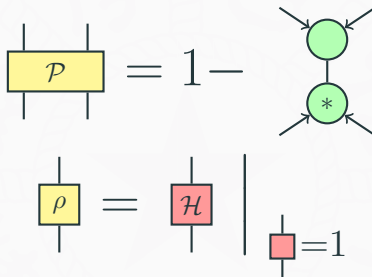
Equations of Motion (Cont'd)

where for all nodes, w. l. o. g., choose one term in \mathbf{H} and one node as an example:



Equations of Motion (Cont'd)

and



Time complexity: if $\text{rank} \leq p$ for all nodes, then a step of multiplication is of $\mathcal{O}(tpdn^{p+1})$;

Space complexity: $\mathcal{O}(tpdn^2 + dn^p)$.

Example: 2-D harmonic oscillator

$$V = \frac{1}{2}x^2 + \frac{1}{2}y^2 + \frac{1}{4}xy \text{ (all in a. u.)}$$

Start from the ground state when $V = \frac{1}{2}x^2 + \frac{1}{2}y^2$. Use Sine-DVR as the (primitive) basis set and $n = 40$, $L = 10$.

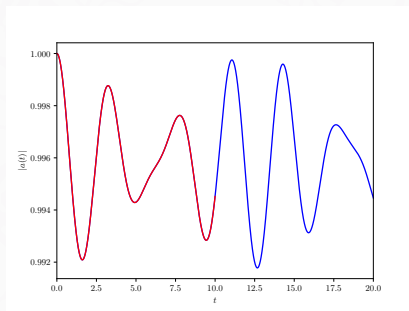


Figure 7: $|a(t)| - t$ (using RK45 as ODE solver, $\Delta t = 0.001$ a. u., renormalize all nodes during the propagation, $\Delta E < 10^{-6}$.)

Red: MCTDH; **blue:** Standard procedure.

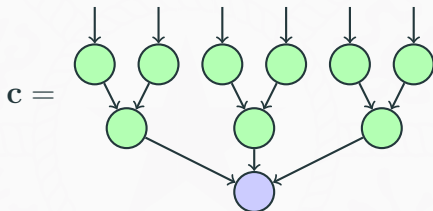
MCTDH vs. DMRG

MCTDH vs. DMRG

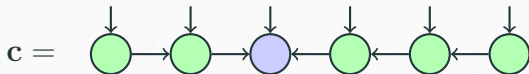
DATA STRUCTURE: A TN PERSPECTIVE

Structure of Wavefunction

- MCTDH



- DMRG



MCTDH vs. DMRG

ALGORITHM: DIFFERENT CHOICES

Algorithms

	TDSE	TISE
DMRG	Based on propergators	DMRG1, DMRG2
MCTDH	Based on DFVP	Self-consistent

- Other combinations?

Renormalization

The essence of sweeping over the tensor train.



Realized by (compressed) singular value decomposition (SVD).

- Use it in other TN-type wavefunctions.
 - Equations often get simpler at the root node.

Another Problem

- How to determine the TN structure of a wavefunction of a specific Hamiltonian *a priori*?

Conclusions

- The structure of wavefunctions in MCTDH and DMRG can be unified in tensor network theory;
- The algorithms used in traditional MCTDH and DMRG are interchangeable in principle;
- The proper structure of wavefunctions in a specific problem needs further studying.

Acknowledgments

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