

MULTI-CONFIGURATION TIME DEPENDENT HARTREE THEORY

A TENSOR NETWORK PERSPECTIVE

Xinxian Chen

September 28, 2018

Department of Chemistry, Tsinghua University

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

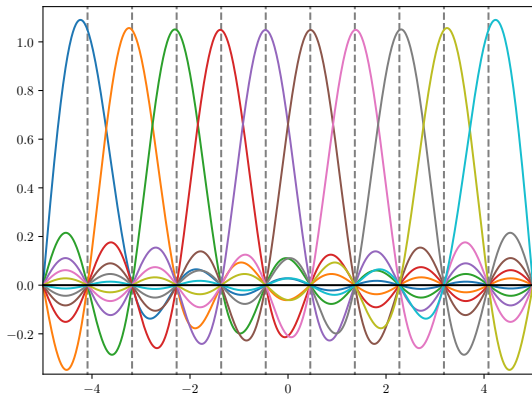


Figure 1: Sine-DVR basis functions ($L = 10$ a. u., $n = 10$)

Example: 1-D Double Well

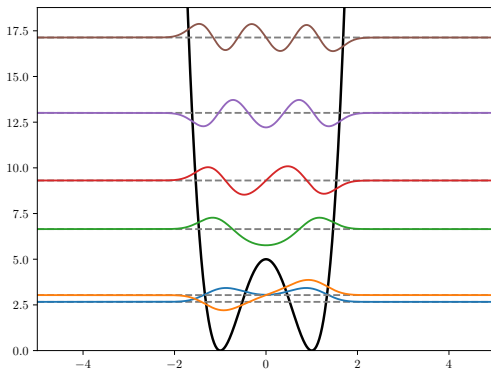


Figure 2: Potential curve and eigenstates

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

Example: 1-D Double Well

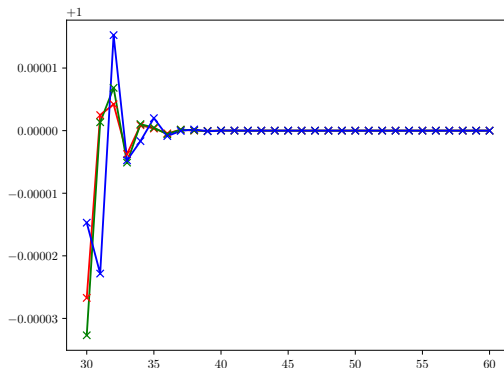


Figure 3: Relative energy to n

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

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

Example: 1-D Double Well

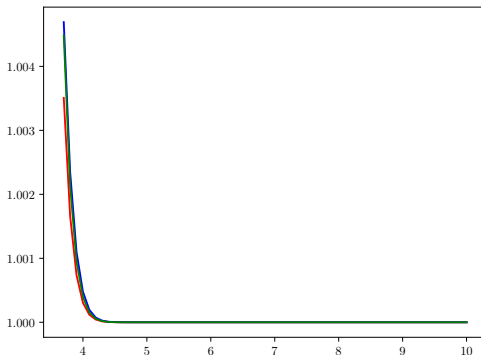


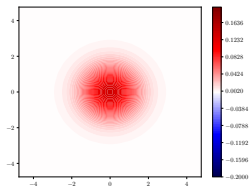
Figure 4: Relative energy to L

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

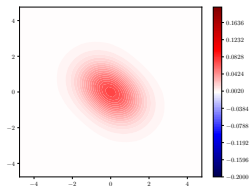
Example: 2-D harmonic oscillator

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

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



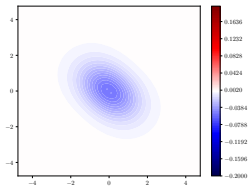
$t = 0.00$ a. u.



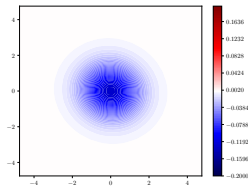
$t = 1.00$ a. u.

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

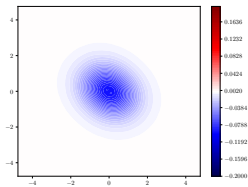
Example: 2-D harmonic oscillator (Cont'd)



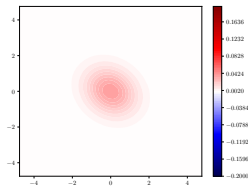
$t = 2.00$ a. u.



$t = 3.00$ 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, $a(t) = \langle \Psi^*(t/2) | \Psi(t/2) \rangle$

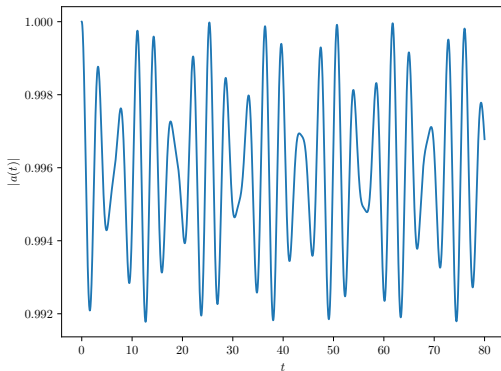


Figure 6: $|a(t)| - t$ (using RK45 as ODE solver, $\Delta t = 0.001$ a. u.)

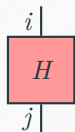
MULTI-CONFIGURATION TIME DEPENDENT HARTREE

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), u (right), and i (bottom). The two bottom legs, both labeled i , are connected by a horizontal line, indicating a contraction. To the right of this diagram is the text $:=$. Further right is the expression \sum_i followed by the same two tensors A and B as on the left, but now they are separate and not connected. The bottom leg of A is labeled i and the bottom leg of B is labeled i , with a multiplication sign \times between them.

MULTI-CONFIGURATION TIME DEPENDENT HARTREE

MOTIVATION

Express the Standard Procedure in TNN

- Wavefunction

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

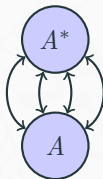
where $i_\kappa \in \{1, \dots, n_\kappa\}$, $\kappa = 1, \dots, d$.

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

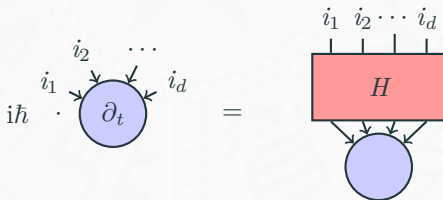
Express the Standard Procedure in TNN

- Normalization condition

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

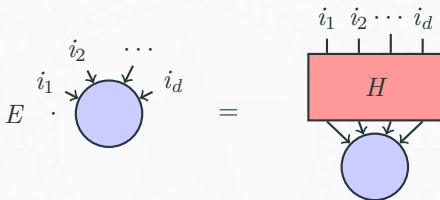


- TDSE



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.

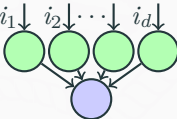
Motivation of MCTDH

Motivation: construct the wavefunction without form the whole space.

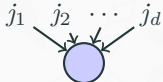
One simple way: Hartree method.

<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):

$$\delta_{ij} =$$


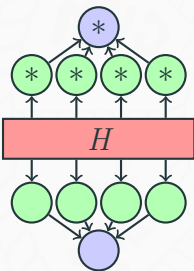
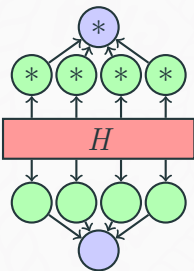
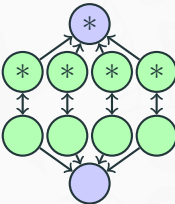
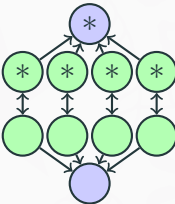
The diagram shows two green circles stacked vertically. The top circle contains a black asterisk (*). A vertical line with a downward arrow points from the label i to the top circle. A vertical line with an upward arrow points from the bottom circle to the label j . A double-headed vertical arrow connects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

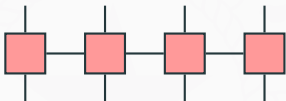
Solve TISE

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





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

Structure of H

- Matrix product operators (MPOs)



- Summation of products of operators

$$\sum_{i=1}^t \left(\begin{array}{c} \text{MPO}_1 \\ \text{MPO}_2 \\ \text{MPO}_3 \\ \text{MPO}_4 \end{array} \right)$$

A diagram showing the summation of products of operators. It features a summation symbol $\sum_{i=1}^t$ followed by a large left parenthesis. Inside the parenthesis are four red square blocks, each with a vertical line extending upwards and another extending downwards, representing individual MPOs. A large right parenthesis follows the blocks.

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

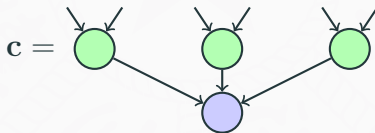
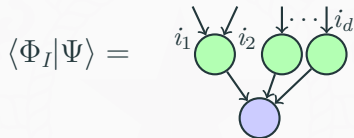
$$\langle \Phi_J^{(1)} | \Psi \rangle = \begin{array}{c} j_1 \quad j_2 \quad \cdots \quad j_d \\ \quad \swarrow \quad \downarrow \quad \searrow \\ \quad \quad \quad \circ \end{array}$$

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.

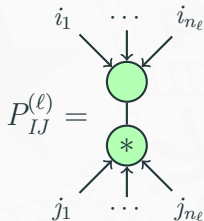
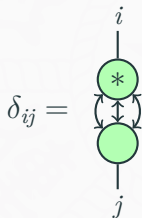
Mode Combination

Examples:



Normalization

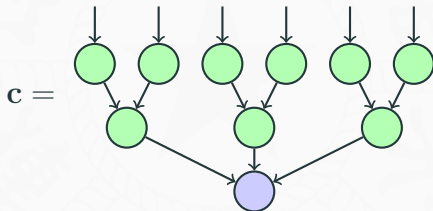
General normalization condition at the ℓ -th node:



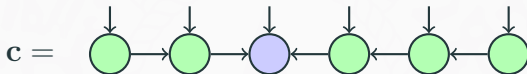
Multi-Layer

More nodes, but smaller ranks.

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 .

Generally, if $\text{rank} \leq p$ for all nodes, the space for saving a wavefunction is of $\mathcal{O}(dn^p)$.

MULTI-CONFIGURATION TIME DEPENDENT HARTREE

ALGORITHM

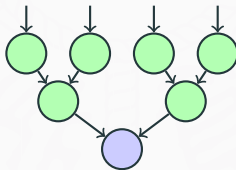
Solve TDSE

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

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

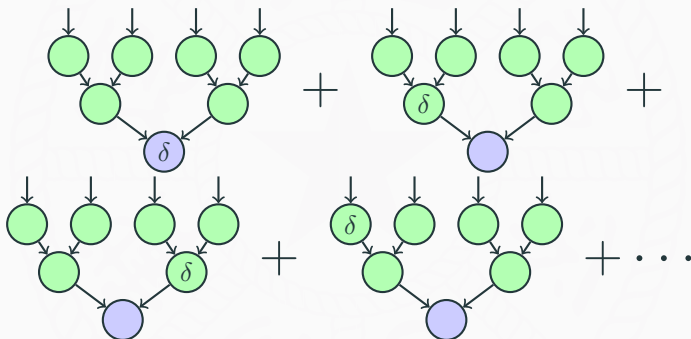
Dirac-Frenkel variational principle: $\langle \delta\Psi | i\hbar \frac{\partial}{\partial t} - H | \Psi \rangle = 0$.

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



Solve TDSE

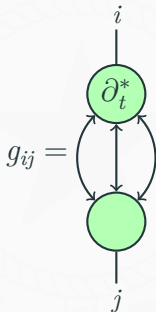
$$\delta \mathbf{c} =$$



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 $\mathbf{g} = \mathbf{0}$.

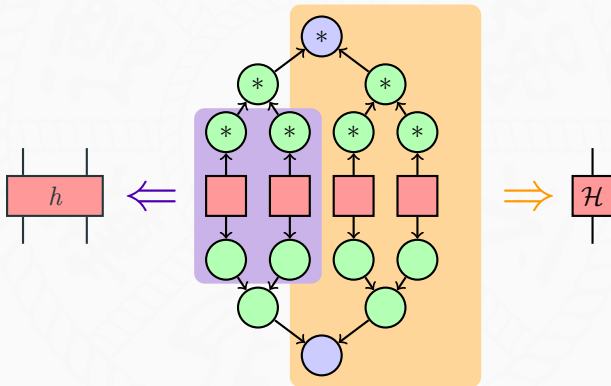
Equations of Motion

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

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

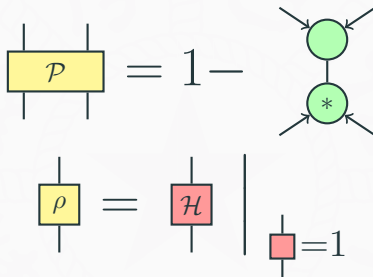
Equations of Motion (Cont'd)

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 $n = 40$, $L = 10$.

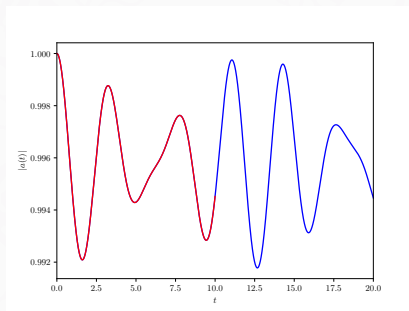


Figure 7: $|a(t)| - t$ (using RK45 as ODE solver, $\Delta t = 0.001$ a. u., renormalize all nodes during the propagation, $\Delta E < 10^{-6}$.)

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

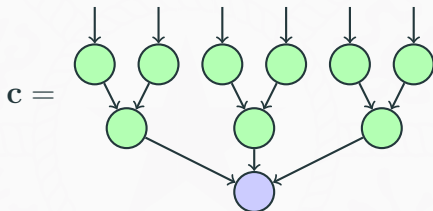
MCTDH vs. DMRG

MCTDH vs. DMRG

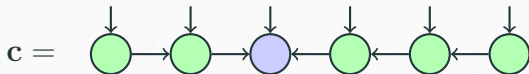
DATA STRUCTURE: A TN PERSPECTIVE

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

Thank “Tsinghua Undergraduate Overseas Training Support Program” for the financial support;

Thank Professor Haobin Wang for his patient help in DVR and (ML-)MCTDH theory;

Thank Jiajun Ren for discussions about DMRG theory.

Thank you for your listening!