

# 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

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# DISCRETE VARIABLE REPRESENTATION

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## 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

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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

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## 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

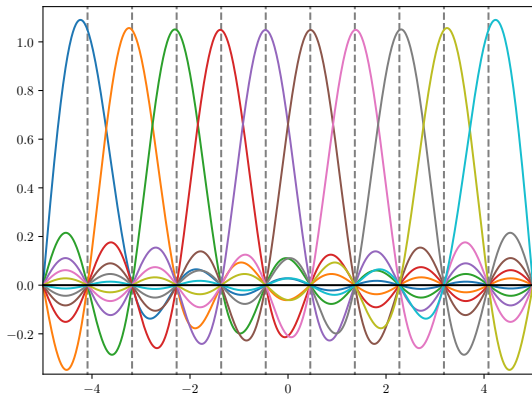
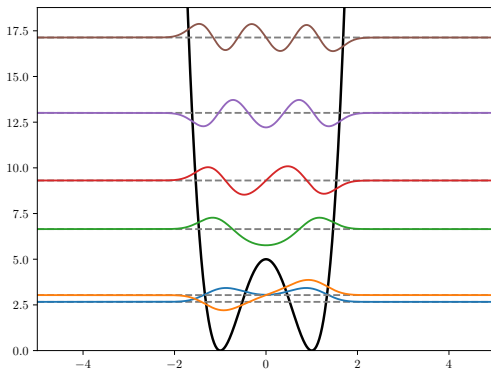


Figure 1: Sine-DVR basis functions ( $L = 10$  a. u.,  $n = 10$ )

# 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

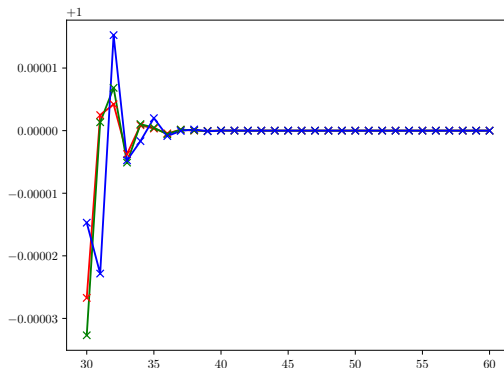


Figure 3: Relative energy to  $n$

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

# Example: 1-D Double Well

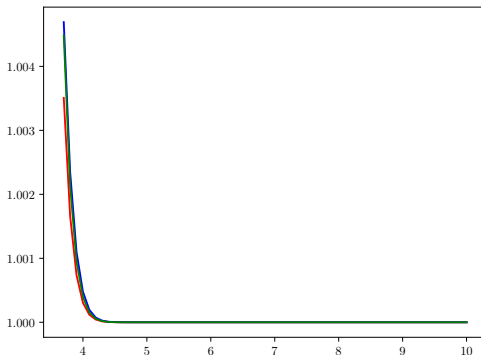


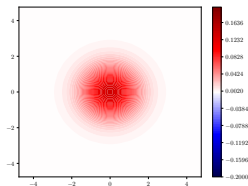
Figure 4: Relative energy to  $L$

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

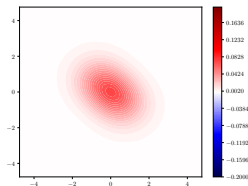
## Example: 2-D harmonic oscillator

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

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



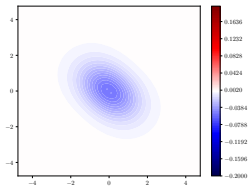
$t = 0.00$  a. u.



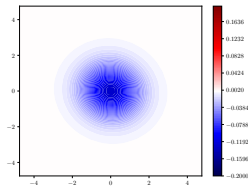
$t = 1.00$  a. u.

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

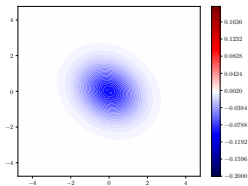
# Example: 2-D harmonic oscillator (Cont'd)



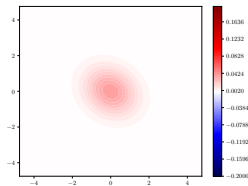
$t = 2.00$  a. u.



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

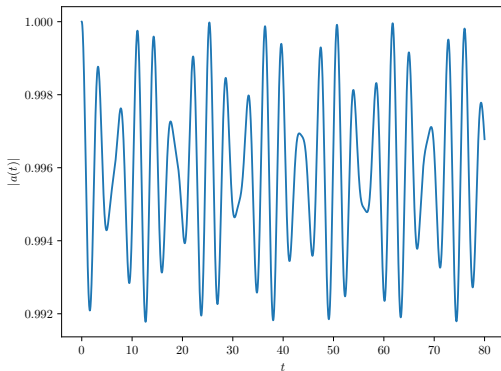


Figure 6:  $|a(t)| - t$  (using RK45 as ODE solver,  $\Delta t = 0.001$  a. u.)



# MULTI-CONFIGURATION TIME DEPENDENT HARTREE

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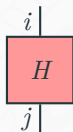
# Language: Tensor Network Notation

- Tensors



A blue circle labeled  $A$  with three external legs: one on the left labeled  $i$ , one on the top right labeled  $j$ , and one on the bottom labeled  $k$ .

$$:= A_{ijk}$$



A red square labeled  $H$  with two external legs: one on the top labeled  $i$  and one on the bottom labeled  $j$ .

$$:= H_{ij}$$

- Contraction



Two blue circles labeled  $A$  and  $B$  connected by a horizontal line.  $A$  has legs  $s$  (left),  $t$  (top), and  $i$  (bottom).  $B$  has legs  $v$  (top),  $u$  (right), and  $i$  (bottom). The bottom legs of both are labeled  $i$ .

$$:= \sum_i \text{ (Diagram of } A \text{ with legs } s, t, i \text{)} \times \text{ (Diagram of } B \text{ with legs } v, u, i \text{)}$$

# MULTI-CONFIGURATION TIME DEPENDENT HARTREE

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## MOTIVATION

# Express the Standard Procedure in TNN

- Wavefunction

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

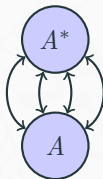
where  $i_\kappa \in \{1, \dots, n_\kappa\}$ ,  $\kappa = 1, \dots, d$ .

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

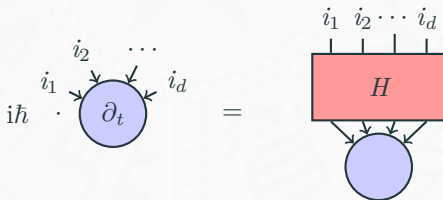
# Express the Standard Procedure in TNN

- Normalization condition

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

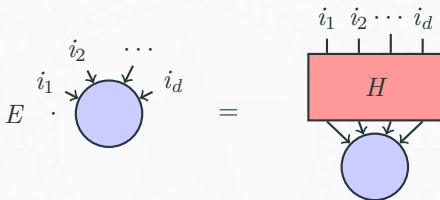


- TDSE



# 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

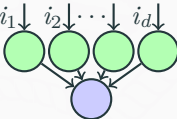
Motivation: construct the wavefunction without form the whole space.

One simple way: Hartree method.

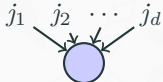
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[http://www.pci.uni-heidelberg.de/tc/usr/mctdh/lit/intro\\_MCTDH.pdf](http://www.pci.uni-heidelberg.de/tc/usr/mctdh/lit/intro_MCTDH.pdf)

# 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  $\kappa$ -th set of single particle functions (SPFs):

$$\delta_{ij} = \begin{array}{c} i \\ | \\ \textcircled{*} \\ \updownarrow \\ \textcircled{\phantom{*}} \\ | \\ j \end{array} \quad P_{ij}^{(\kappa)} = \begin{array}{c} i \\ \downarrow \\ \textcircled{\phantom{*}} \\ | \\ \textcircled{*} \\ \uparrow \\ j \end{array}$$

# Variational Principle

Solve TISE

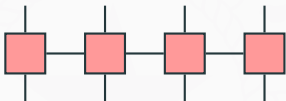
$$\delta \left( \begin{array}{c} \text{Diagram 1} \\ \text{Diagram 2} \end{array} - \lambda \left( \begin{array}{c} \text{Diagram 3} \\ \text{Diagram 4} \end{array} - 1 \right) \right) = 0$$

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  $H$  with respect to a trial wavefunction, constrained to have a norm of 1. The diagrams consist of nodes (green circles with asterisks, blue circles with asterisks, and empty green circles) and arrows representing the structure of the wavefunction and the Hamiltonian operator.

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 squares, each with a vertical line extending upwards and another extending downwards, representing individual MPOs. A large right parenthesis follows the squares.

# MULTI-CONFIGURATION TIME DEPENDENT HARTREE

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DATA STRUCTURE

# Structure of Wavefunction

Diagram illustrating the inner product of two states,  $\langle \Phi_I | \Psi \rangle$ . The circuit consists of four green circles (qubits) and one purple circle (qubit). The green circles are labeled  $i_1, i_2, \dots, i_d$  above them, indicating inputs. The purple circle is labeled  $i_{d+1}$  below it, indicating an output. The green circles are connected to the purple circle by arrows, representing the inner product operation.

where

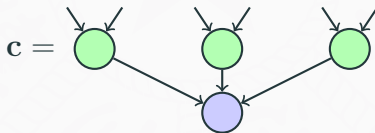
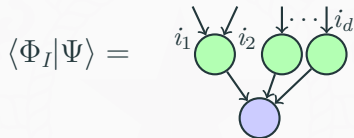
$$\langle \Phi_J^{(1)} | \Psi \rangle = \begin{array}{c} j_1 \quad j_2 \quad \cdots \quad j_d \\ \swarrow \quad \downarrow \quad \searrow \\ \text{blue circle} \end{array}$$

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.

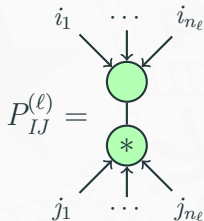
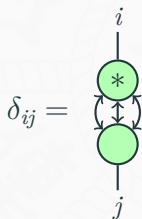
# Mode Combination

Examples:



# Normalization

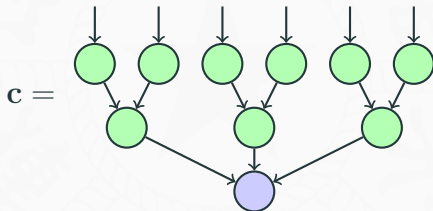
General normalization condition at the  $\ell$ -th node:



# Multi-Layer

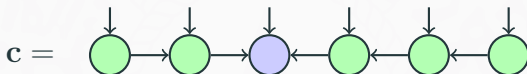
More nodes, but smaller ranks.

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  $\ell$ .

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

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ALGORITHM

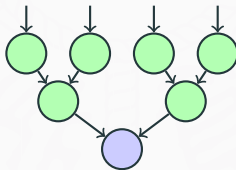
# Solve TDSE

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

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

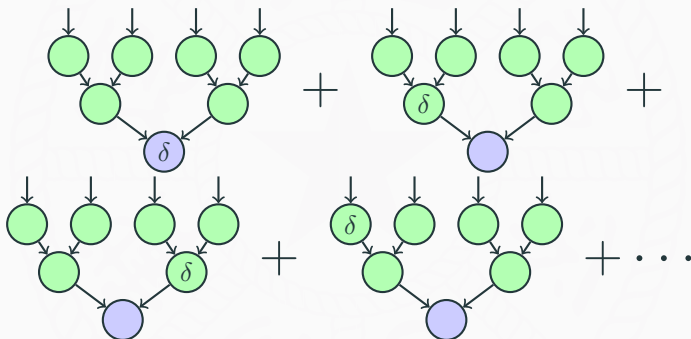
Dirac-Frenkel variational principle:  $\langle \delta\Psi | i\hbar \frac{\partial}{\partial t} - H | \Psi \rangle = 0$ .

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



# Solve TDSE

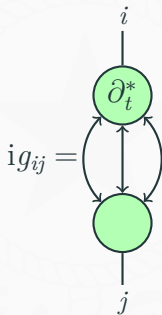
$$\delta \mathbf{c} =$$



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

# Normalization

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



where  $\mathbf{g}$  is Hermitian. For simplicity, choose  $\mathbf{g} = 0$ .

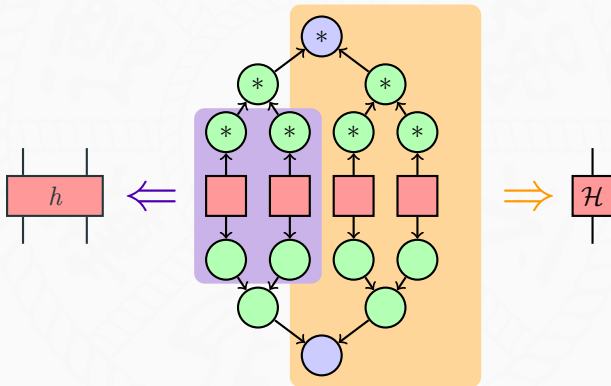
# Equations of Motion

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

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

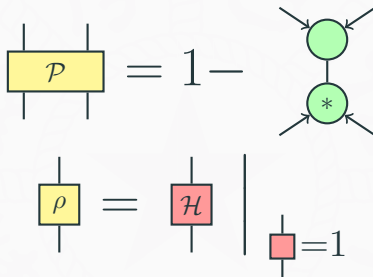
# Equations of Motion (Cont'd)

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  $\mathcal{O}(tpdn^{p+1})$ ;

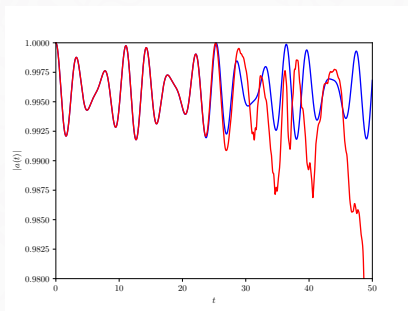
Space complexity:  $\mathcal{O}(tpdn^2 + dn^p)$ .



# 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.)

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

# MCTDH vs. DMRG

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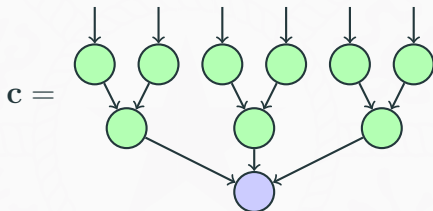
# MCTDH vs. DMRG

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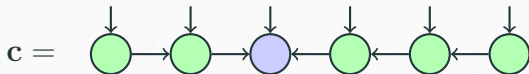
DATA STRUCTURE: A TN PERSPECTIVE

# Structure of Wavefunction

- MCTDH



- DMRG



# MCTDH vs. DMRG

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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.

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