

# Density matrix renormalization group with long-range interactions and its applications

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Jinhua, 2012.08.14-18

## Outline:

- 1) Density matrix renormalization group with long-range interaction
- 2) The  $t$ - $J$ - $V$ - $W$  model realized in polar molecule systems
- 3) Some preliminary results

# Fundamental issue in quantum chemistry calculation

How to solve the Schrodinger equation of molecules  
**accurately** and **efficiently** ?

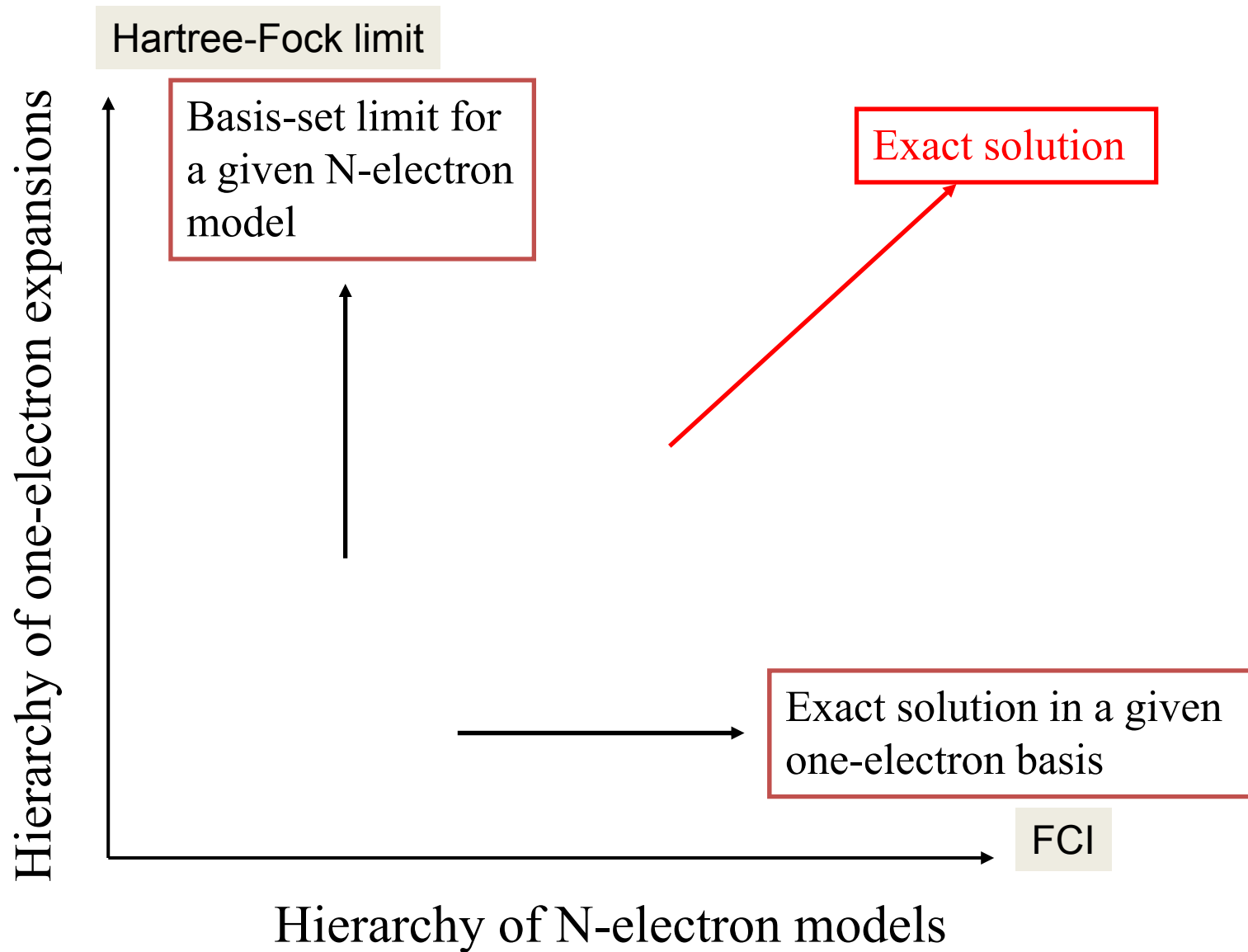
$$i\hbar \frac{\partial}{\partial t} \psi(r, R) = \left[ -\sum_i \frac{\hbar^2}{2m_e} \nabla_i^2 + V(r, R) \right] \psi(r, R)$$

$$V(r, R) = -\sum_{il} \frac{Z_l e^2}{|r_i - R_l|} + \frac{1}{2} \sum_{i \neq j} \frac{e^2}{|r_i - r_j|}$$

*“The underlying physical laws necessary for the mathematical theory of a large part of physics and the whole of chemistry are thus completely known, and the difficulty is only that the exact application of these laws leads to equations much too complicated to be soluble.”*

--- P. A. M. Dirac, Proc. R. Soc. Lond. Ser. A123, 714 (1929)

# Two errors in solving for the Schrodinger equation



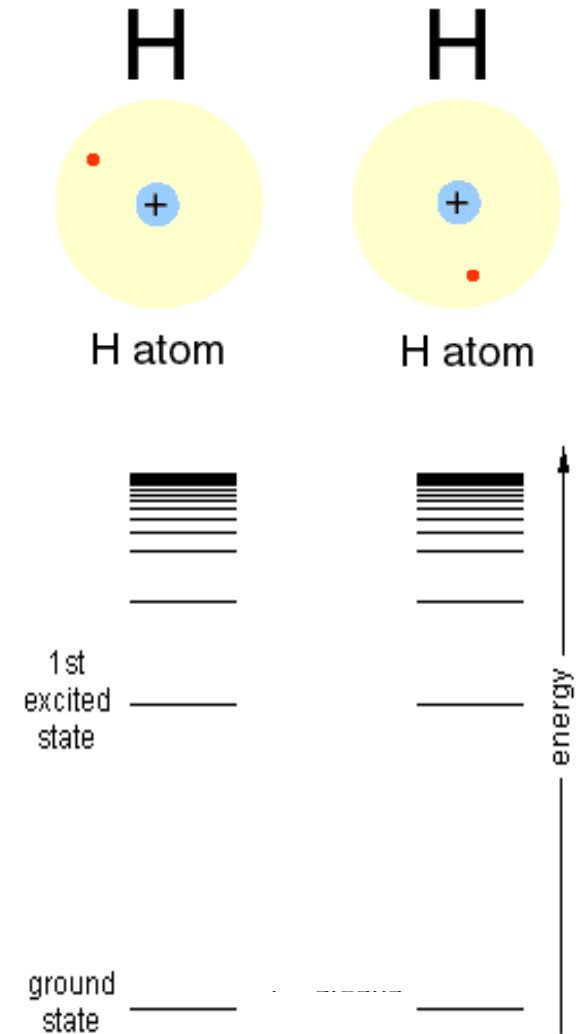
# Numerical Obstacle: Exponential Wall

- ✓ Fixing orbital number  $N$   
the total degree of freedom  
increases exponentially with the  
number of electrons
- ✓ Orbital number  $N$  is infinity

e.g., for  $H_2$  molecule:

If  $N$  atomic orbitals are used for each  
atom, then the total number of many-  
body bases is

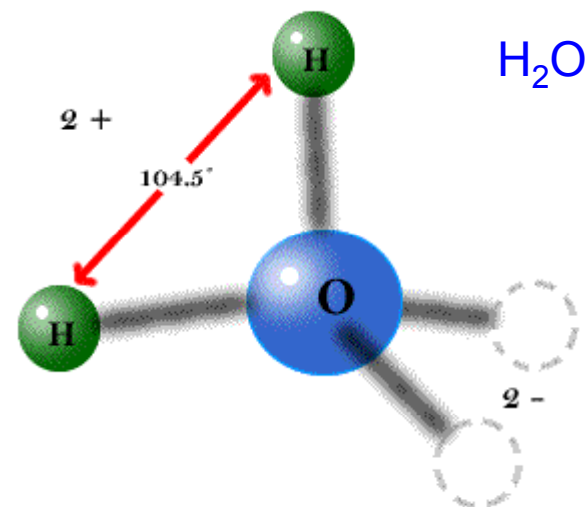
$$N^2 (2N^2 - 1)$$



# How many degrees of freedom for H<sub>2</sub>O ?

If  $N$  molecule orbitals are used,  
the total number of basis states  
is

$$C_N^5 C_N^5 \sim N^{10}$$



***The total degree of freedom increases exponentially  
with the number of electrons!***

# Weak Coupling Approach

Convert a many-body problem into a one-body problem

- ✓ **Hartree-Fock self-consistent mean field theory**
- ✓ **Density Functional Theory**
  - Most successful numerical method for treating weak coupling systems
  - Based on LDA or other approximations, less accurate



# Strong Coupling Approach

Use a finite set of many-body basis states to treat a correlated system

- ✓ **Configuration Interactions (CI)**
  - Conceptually simple, but can only deal a small number of orbitals
- ✓ **Coupled Cluster Expansion (CC)**
  - Perturbative
- ✓ **Quantum Monte Carlo**
  - Not variational, suffer from the “minus-sign” problem
- ✓ **Density matrix renormalization group**
  - Variational, accurate and highly controllable

# Correlation and Excitations

	orbitals	HF	single- S	double- D	triple-excitations T
<p>✓ <b>CI or CC:</b> Include single (S), double (D), triple (T) excitations</p>	—	—	—	—	—
	—	—	—	—	—
	—	—	—	—	—
	—	—	—	—	—
	—	—	—	—	—
	—	—	—	—	—
	—	—	—	—	—
	—	—	—	—	—
	—	—	—	—	↑
	—	—	—	↑	↑
<p>✓ <b>DMRG:</b> Can handle arbitrary many excitations</p>	—	↓	↓	↓	↓
	↑↓	↑	↑	↑	↑
	↑↓	↑↓	↓	↓	↓
	↑↓	↑↓	↑↓	↑↓	↓
	↑↓	↑↓	↑↓	↑↓	↑↓
	↑↓	↑↓	↑↓	↑↓	↑↓
	↑↓	↑↓	↑↓	↑↓	↑↓
	↑↓	↑↓	↑↓	↑↓	↑↓
	↑↓	↑↓	↑↓	↑↓	↑↓
	↑↓	↑↓	↑↓	↑↓	↑↓
Real many-body wave-function	$\Phi_0$	$\Phi_i^a$	$\Phi_{ij}^{ab}$	$\Phi_{ijk}^{abc}$	
		$\hat{T}_1$	$\hat{T}_2$	$\hat{T}_3$	
		$\hat{C}_1$	$\hat{C}_2$	$\hat{C}_3$	

# Configuration Interaction and Coupled Cluster Expansion

Full CI

$$|\Phi\rangle = c_0 |\Phi_0\rangle + c_S |S\rangle + c_D |D\rangle + c_T |T\rangle + c_Q |Q\rangle + \dots$$

CISD

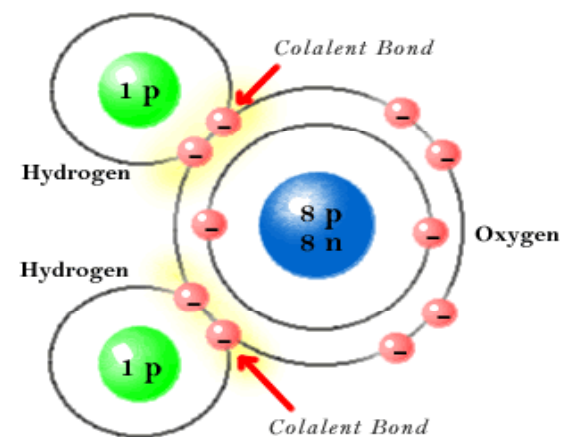
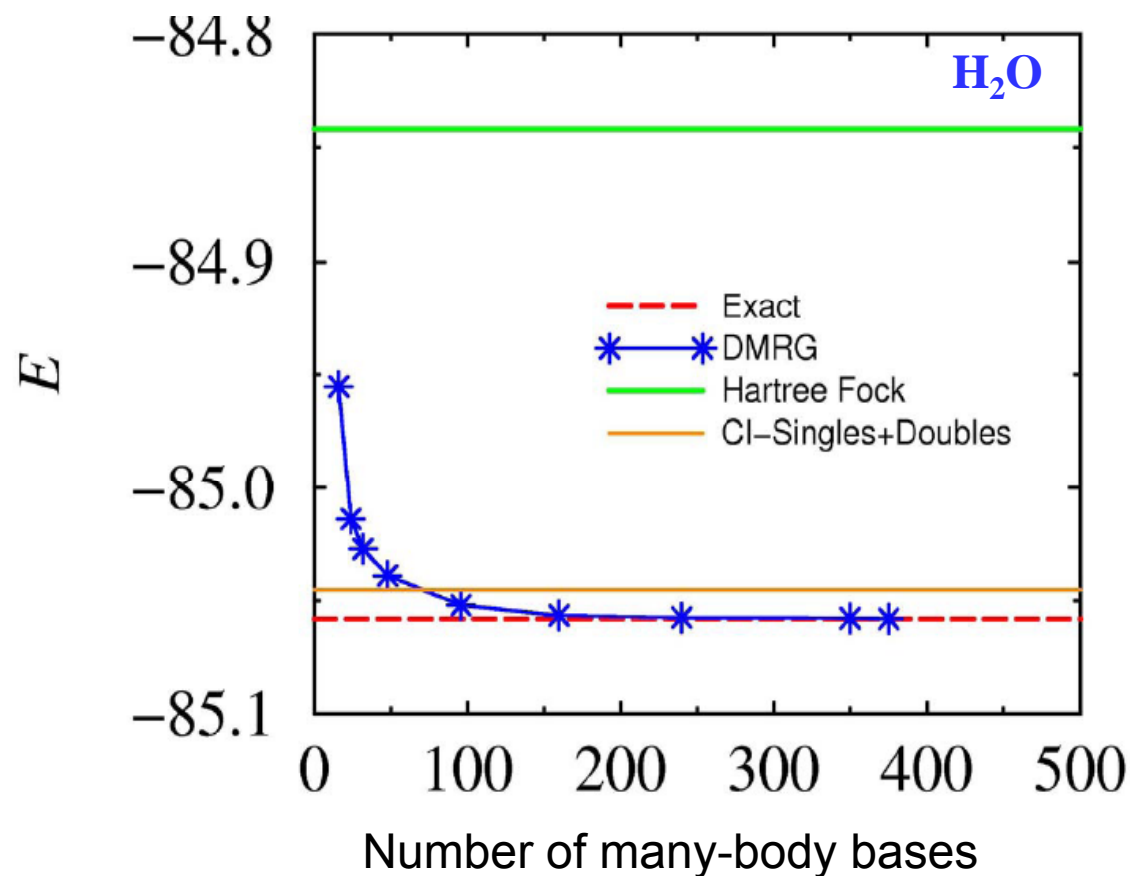
$$|\Phi\rangle = c_0 |\Phi_0\rangle + c_S |S\rangle + c_D |D\rangle + \cancel{c_T |T\rangle} + \cancel{c_Q |Q\rangle} + \dots$$

Coupled-cluster expansion

$$\Psi_{cc} = \exp(T) |\Phi_0\rangle = \left(1 + T + \frac{1}{2}T^2 + \dots\right) |\Phi_0\rangle$$

$$T = T_1 + T_2 + \dots$$

# Accuracy of different methods



**Use 25 H<sub>2</sub>O  
molecular orbitals**

**DMRG: 1s<sup>2</sup> of  
oxygen is frozen**

S. R. White and R. Martin, J. Chem. Phys. 110, 4127 (1999)

Main difficulty:

treating accurately the correlation, needing large basis

## Feature Article

# Electron Correlation: The Many-Body Problem at the Heart of Chemistry

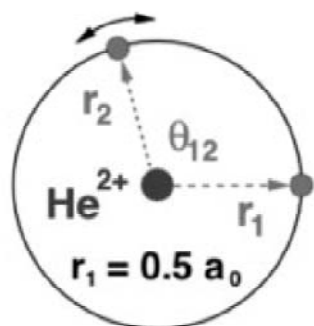
**DAVID P. TEW,<sup>1</sup> WIM KLOPPER,<sup>1</sup> TRYGVE HELGAKER<sup>2</sup>**

*<sup>1</sup>Lehrstuhl für Theoretische Chemie, Institut für Physikalische Chemie, Universität Karlsruhe (TH),  
D-76128 Karlsruhe, Germany*

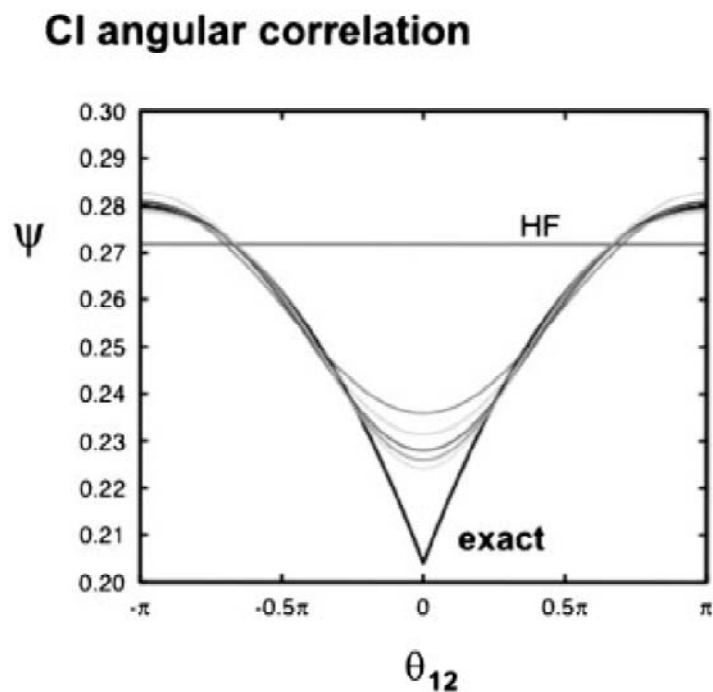
*<sup>2</sup>Department of Chemistry, University of Oslo, P.O.Box 1033 Blindern, N-0315 Oslo, Norway*

J. Comput. Chem. 28, 1307 (2007)

A toy model for Helium atom:



CI	$n_{\max}$
—	3
—	4
—	5
—	6
—	7



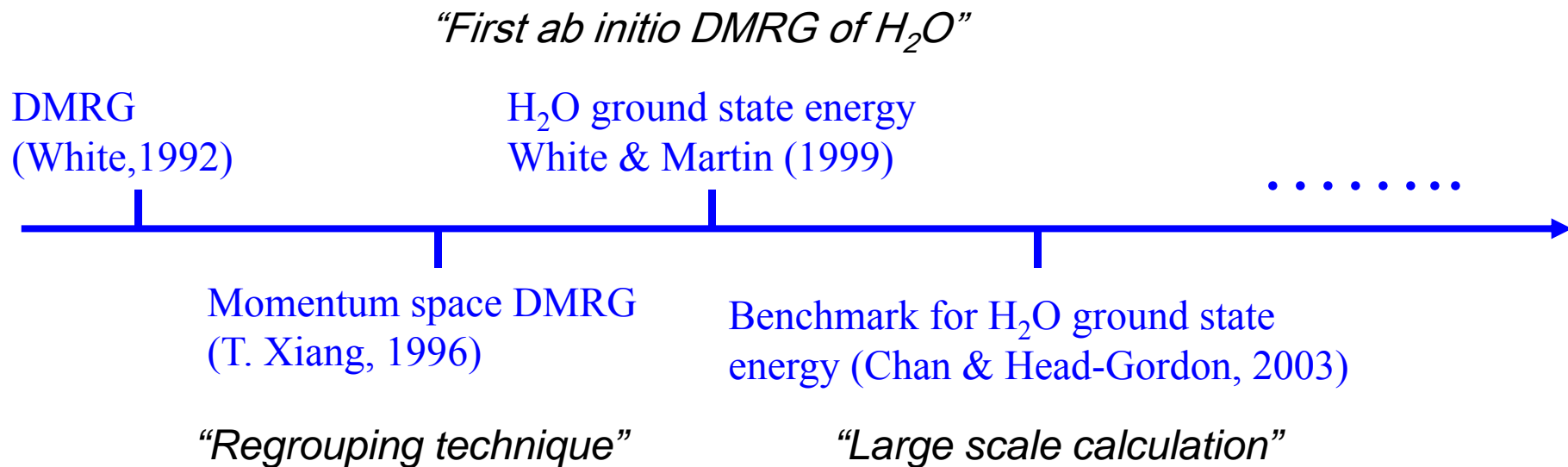
The electron-electron cusp:

A problem can not be precisely treated with the products of smooth orbital functions

J. Comput. Chem. 28, 1307 (2007)

# **Quantum chemistry density matrix renormalization group**

# Brief history of the *ab initio* quantum chemistry DMRG





# How to apply DMRG to Quantum Chemistry

To represent the Hamiltonian using a set of HF molecular orbitals

$$H = \sum_{ij\sigma}^N t_{ij} c_{i\sigma}^+ c_{j\sigma} + \frac{1}{2} \sum_{ijrt, \sigma\sigma'}^N V_{ijrt} c_{i\sigma}^+ c_{j\sigma'}^+ c_{t\sigma} c_{r\sigma}$$

$$t_{ij} = \int dr \varphi_i^*(r) \left[ -\frac{1}{2} \Delta - \sum_{k=1} \frac{Z_k e^2}{|r - r_k|} \right] \varphi_j(r)$$

$$V_{ijrt} = \iint dr dr' \varphi_i^*(r) \varphi_j^*(r') \left[ \frac{e^2}{|r_i - r_j|} \right] \varphi_t(r') \varphi_r(r)$$

Molecular orbital: HF single particle basis

But,

$$\hat{H} = \sum_{ij}^N t_{ij} c_i^+ c_j + \frac{1}{2} \sum_{ijkl}^N V_{ijkl} c_i^+ c_j c_k^+ c_l$$

$N^4$  terms!

There are too many interaction terms:  $O(N^4)$   
compute time and storage cost:  $> O(N^5)$

Only a small number of molecular orbitals can be used

**Impractical !**

# Regrouping Operators:

T Xiang, Phys Rev B 53,R10445 (96)

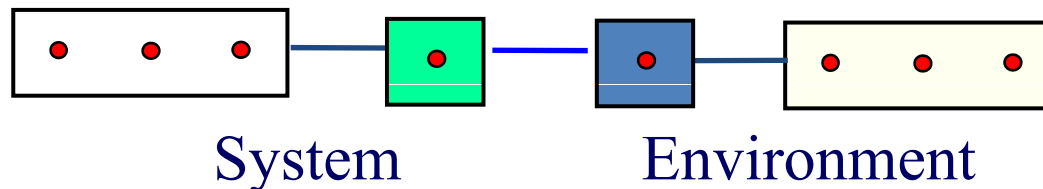
$$\hat{H} = \sum_{ij}^N t_{ij} c_i^+ c_j + \frac{1}{2} \sum_{ijkl}^N V_{ijkl} c_i^+ c_j c_k^+ c_l$$

$$O = \sum_{ijkl \in \text{Sys}} V_{ijkl} c_i^+ c_j c_k^+ c_l \quad \longrightarrow \quad O(N^4) \rightarrow O(1)$$

$$O_i = \sum_{jrt \in \text{Sys}} V_{ijrt} c_j^+ c_t c_r \quad \longrightarrow \quad O(N^4) \rightarrow O(N)$$

$$O_{ij} = \sum_{rt \in \text{Sys}} V_{ijrt} c_t c_r \quad \longrightarrow \quad O(N^4) \rightarrow O(N^2)$$

$O(N^4)$  terms  $\rightarrow$   $O(N^2)$  terms



But,

$$\hat{H} = \sum_{ij}^N t_{ij} c_i^+ c_j + \frac{1}{2} \sum_{ijkl}^N V_{ijkl} c_i^+ c_j c_k^+ c_l$$

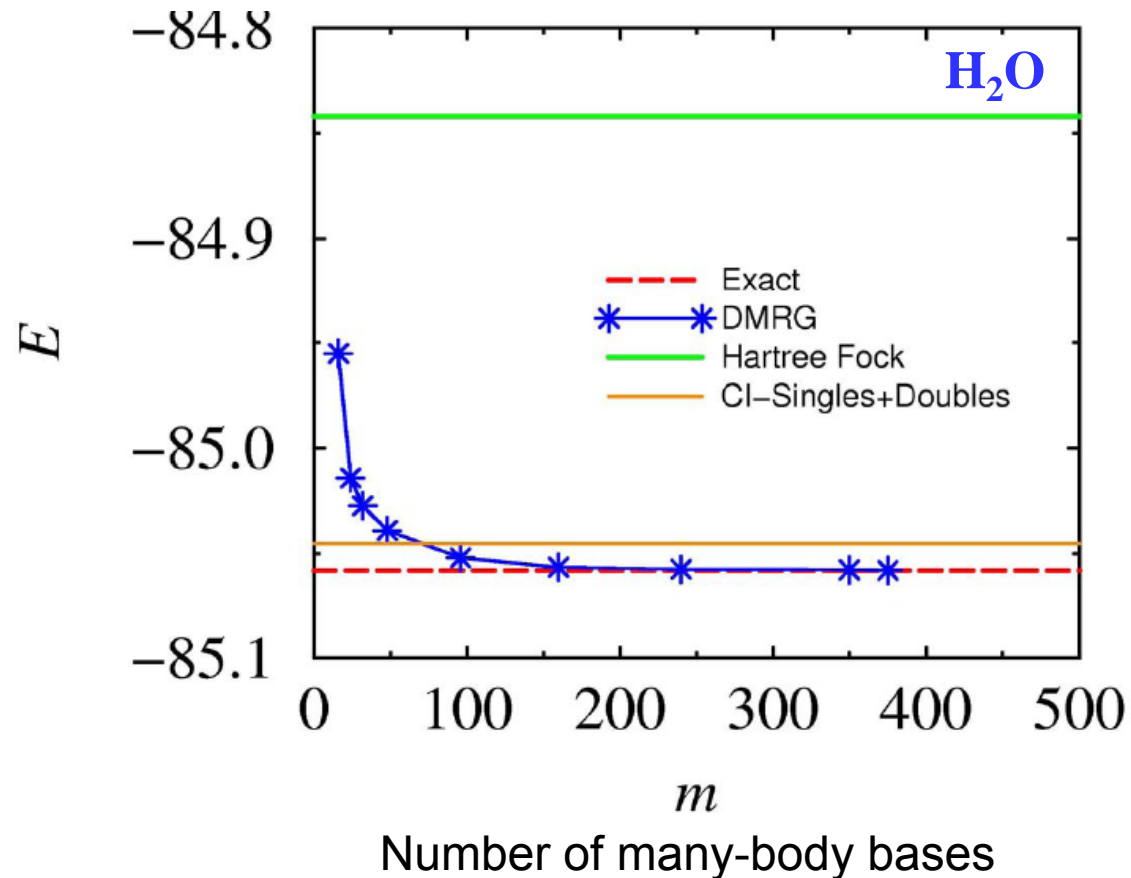
~~$N^4$  terms!~~

There are too many interaction terms:  $O(N^4)$   
compute time and storage cost:  $> O(N^5)$

Only a small number of molecular orbitals can be used

**Practical !**

# First *ab initio* DMRG Calculation of Molecules



Use 25  $\text{H}_2\text{O}$   
molecular orbitals

DMRG:  $1s^2$  of  
oxygen is frozen

S. R. White and R. Martin, J. Chem. Phys. 110, 4127 (1999)

## Exact solution (within a triple-zeta, double polarization basis set) of the electronic Schrodinger equation for water

Garnet Kin-Lic Chan

*Department of Chemistry, University of Cambridge, United Kingdom CB2 1EW*

Martin Head-Gordon

*Department of Chemistry, University of California, Berkeley, California 94070*

41 orbitals

Orbital #: $N$	Unrestricted Hartree-Fock	Coupled Cluster (SDT)	DMRG <sup>1</sup>	Quantum Monte Carlo <sup>2</sup>
41	-85.254555	-85.511075	-85.511649 (M=6000)	-85.5132(6)

Benchmark

- ✓ This was the largest “exact” calculation done (Dimension of Hilbert space:  $5.6 \times 10^{11}$ )

<sup>2</sup> Shiwei Zhang et al J. Chem. Phys. 124, 224101 (2006)

	UHF	CCSD(T)	FCI/DMRG	QMC
<b>STO-6G</b>				
H	-0.471 039	-0.471 039	-0.471 039	-0.471 039
O	-74.516 816	-74.516 816	-74.516 816	-74.516 816
H <sub>2</sub> O	-75.676 506	-75.727 931	-75.727 991	-75.728 5(1)
BE	0.217 612	0.269 037	0.269 097	0.269 5(1)
<b>cc-pVDZ</b>				
H	-0.499 278	-0.499 278	-0.499 278	-0.499 278
O	-74.792 166	-74.911 552	-74.911 744	-74.909 6(1)
H <sub>2</sub> O	-76.024 039	-76.241 201	-76.241 860	-76.242 4(2)
BE	0.233 317	0.331 093	0.331 560	0.334 3(2)
<b>DZ ANO 41 HF orbitals</b>				
H	-0.499 944	-0.499 944	-0.499 944	-0.499 944
O	-74.816 273	-74.961 956	-74.962 350	-74.959 6(1)
H <sub>2</sub> O	-76.057 621	-76.314 141	-76.314 715	-76.316 3(6)
BE	0.241 460	0.352 959	0.352 477	0.356 8(7)
<b>TZ ANO 92 HF orbitals</b>				
H	-0.499 973	-0.499 973		-0.499 973
O	-74.818 648	-75.000 129	?	-74.997 0(4)
H <sub>2</sub> O	-76.060 589	-76.367 528		-76.370(1)
BE	0.241 995	0.367 453		0.373(1)

W.A. Al-Saidi, Shiwei Zhang,  
and H. Krakauer, J. Chem.  
Phys. 124, 224101 (2006)

Chan and Head-  
Gordon, 2003

Benchmark is missing!

The number of single-electron basis states that can be handled by the DMRG is larger than in Full CI, but still limited

Orbital #: $N$	Unrestricted Hartree-Fock	Coupled Cluster (SDT)	DMRG <sup>1</sup>	Quantum Monte Carlo <sup>2</sup>
41	-85.254555	-85.511075	-85.511649 (M=6000)	-85.5132(6)
92	-85.257519	-85.564462	?	-85.567(1)

How to solve this problem?

Optimize the single-electron basis state by DMRG



# Optimize the single-electron basis state by DMRG

Step 1:

- Start from a large basis set (say 200 orbitals), partition the basis set into three subsets, core orbitals, active orbitals and virtual orbitals
- Freeze the core and virtual orbitals
- Perform the DMRG calculation in the active space

Core orbitals  
doubly occupied

Active space

Virtual orbitals  
unoccupied

$$H = \sum_{ij} t_{ij} a_i^+ a_j + \sum_{ijkl} v_{ijkl} a_i^+ a_j^+ a_l a_k$$



$$H_{active} = E_{frozen} + \sum_{ij \in active} t_{ij}^{active} a_i^+ a_j + \sum_{ijkl \in active} v_{ijkl} a_i^+ a_j^+ a_l a_k$$

DMRG treatment

# Optimize the single-electron basis state by DMRG

Step 2:

- Optimize the single-electron basis states by diagonalizing the single-particle density matrix deduced from the DMRG wave function in the active space

Core orbitals  
doubly occupied

Active space

Virtual orbitals  
unoccupied

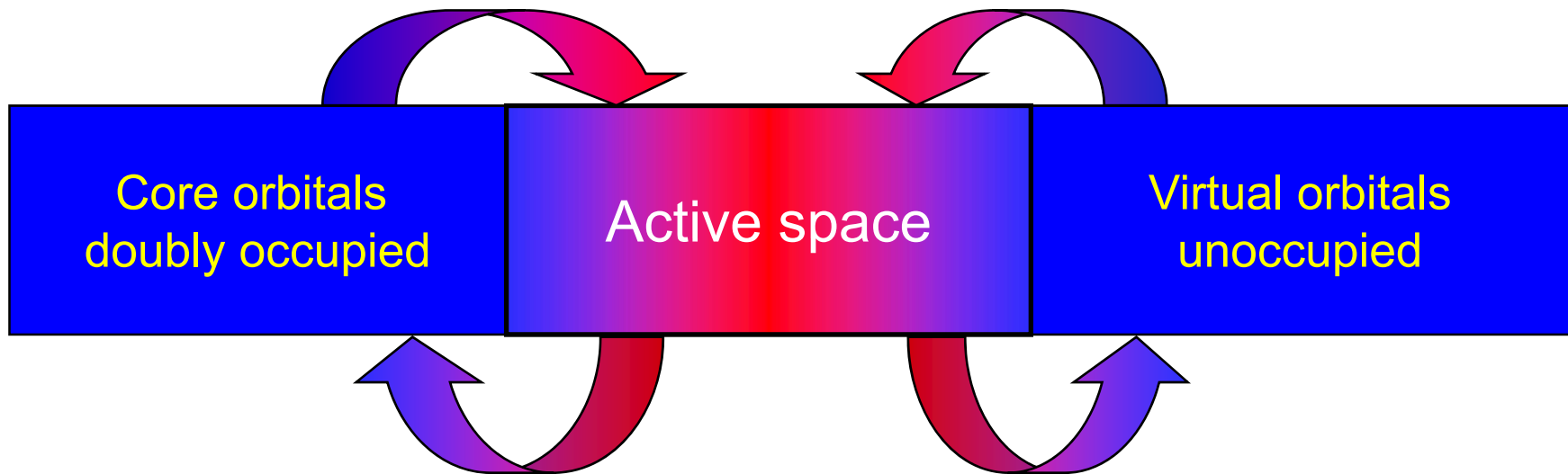
$$\rho_{ij} = \langle \psi | c_i^\dagger c_j | \psi \rangle$$

# Optimize the single-electron basis state by DMRG

Step 3:

- Swap a few orbitals between the active and inactive spaces, so that some inactive orbitals can be activated and optimized

Activate a few 'frozen' previously orbitals in the inactive spaces.

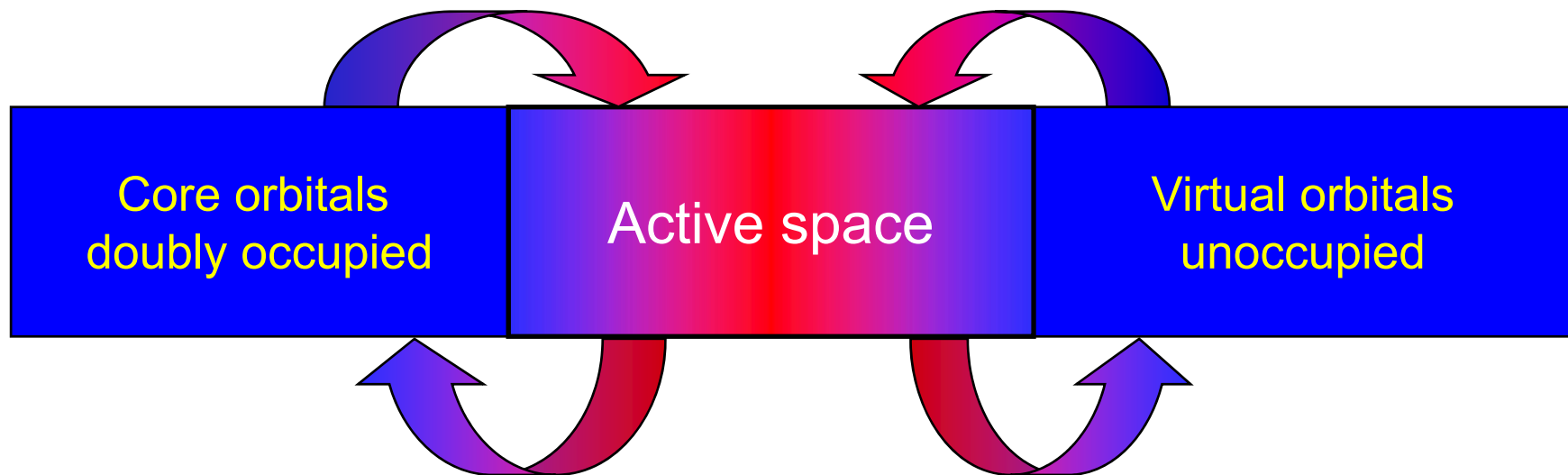


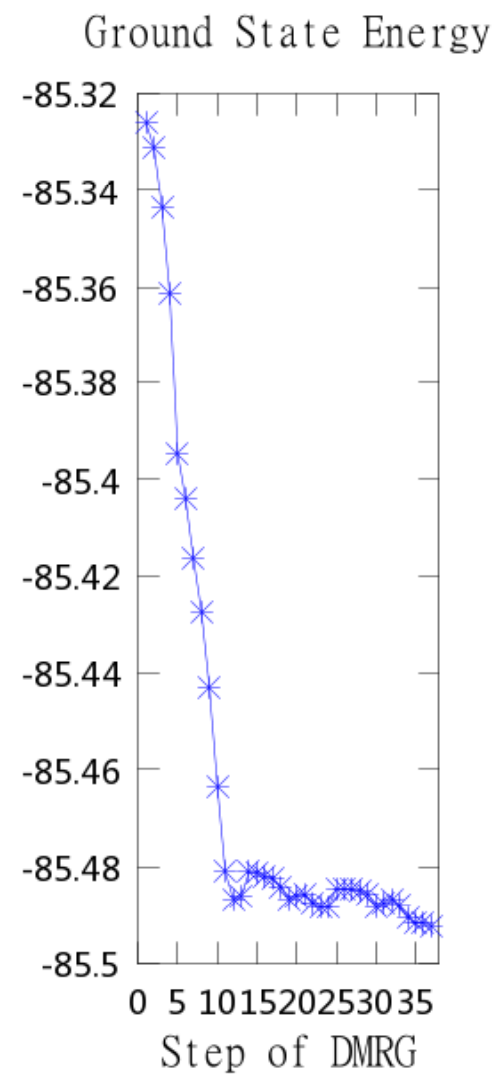
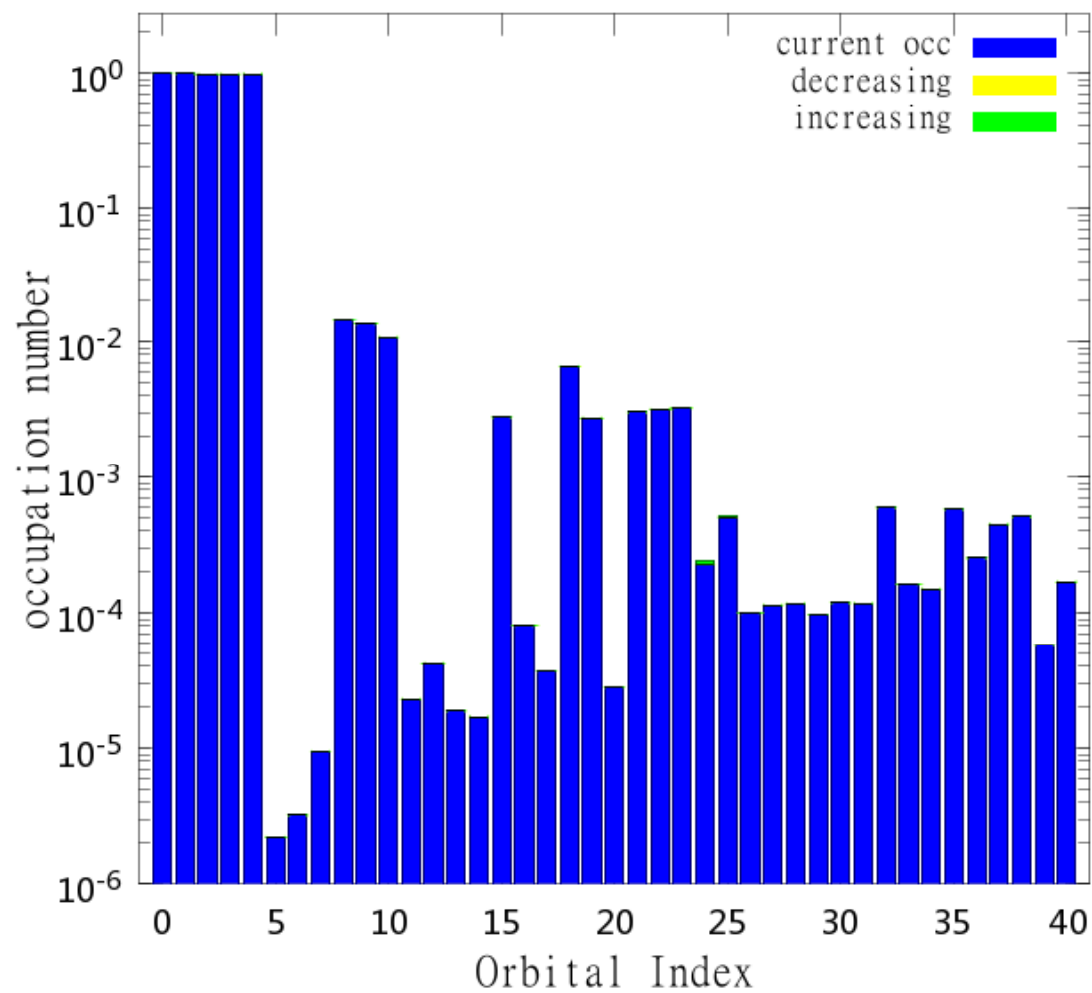
Meanwhile, freeze the less important a few orbitals in the active space.

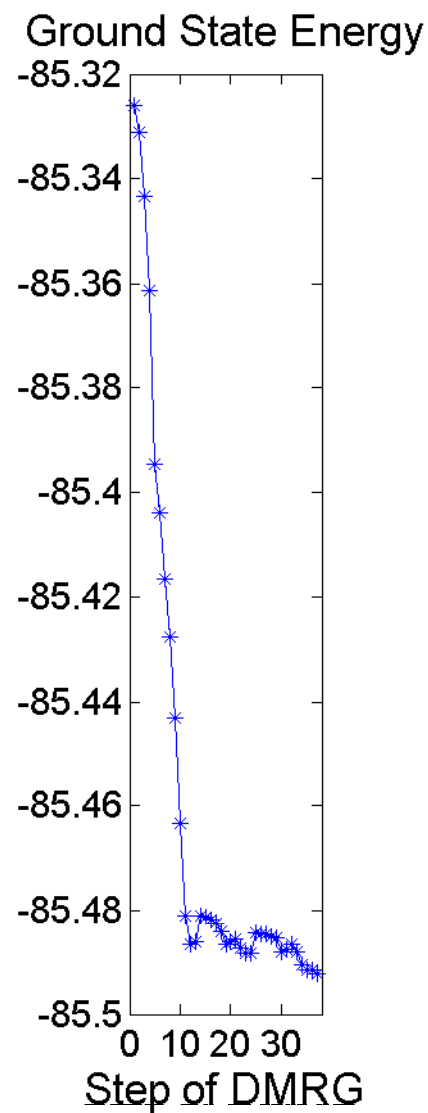
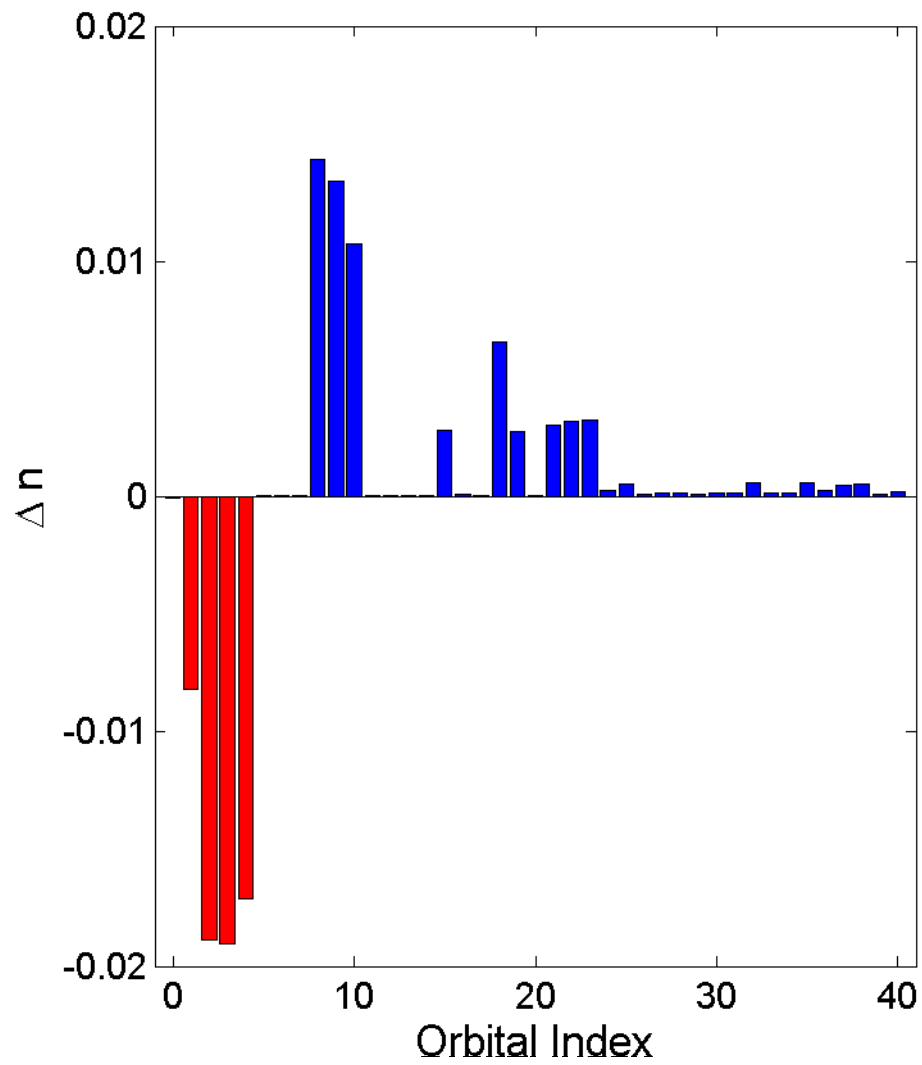
# Optimize the single-electron basis state by DMRG

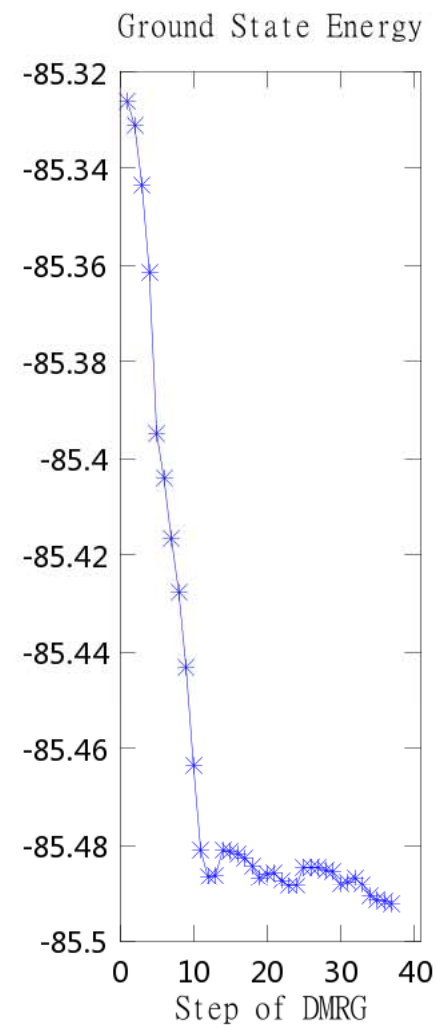
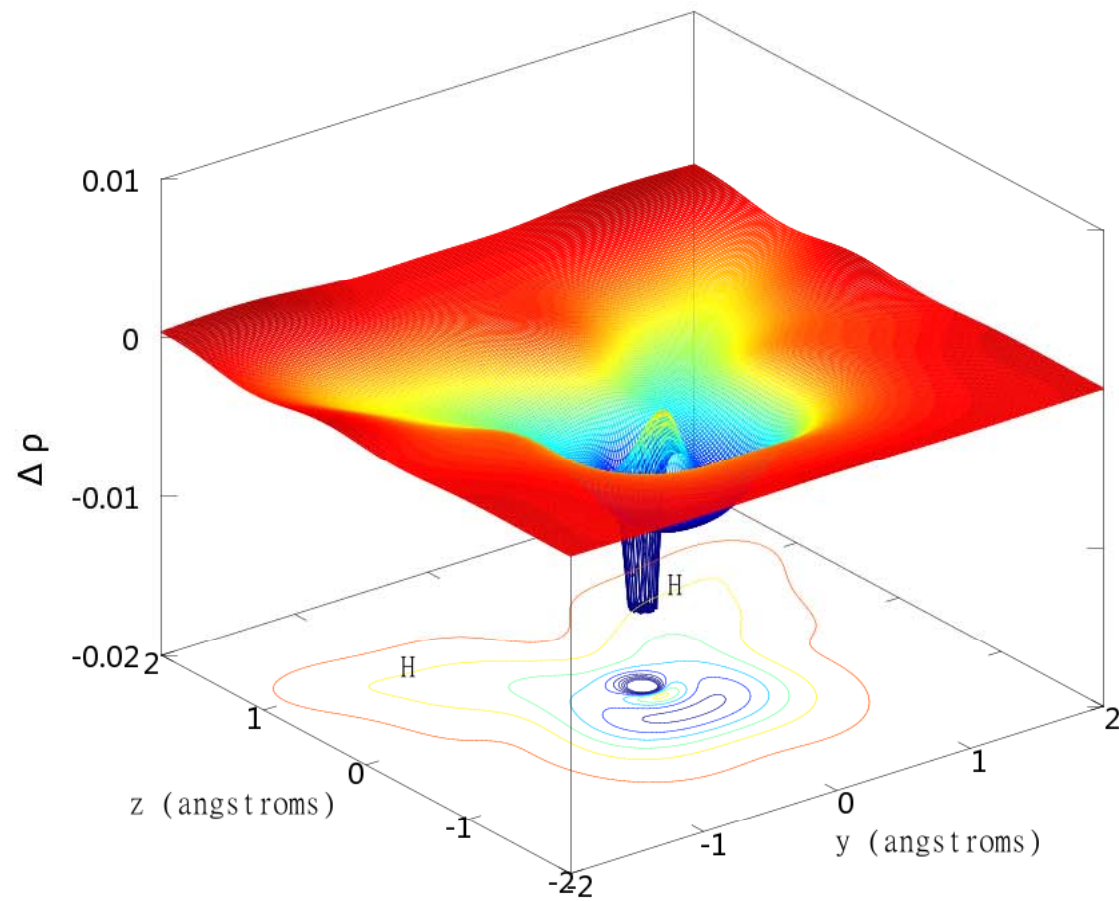
Step 4:

- Repeat the above steps so that all orbitals are activated and optimized, this complete a full cycle of optimization
- This cycle can be repeated more than once



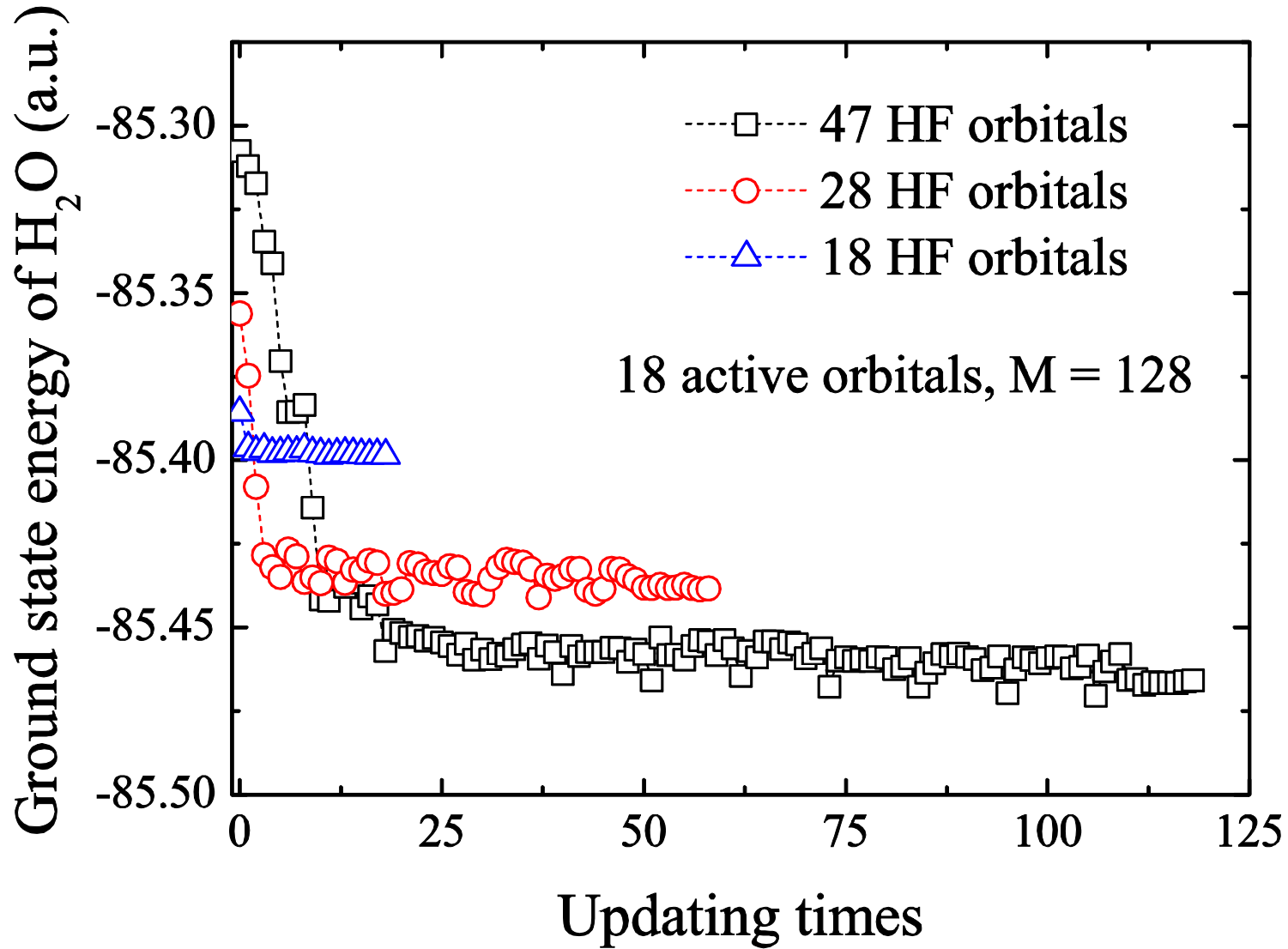




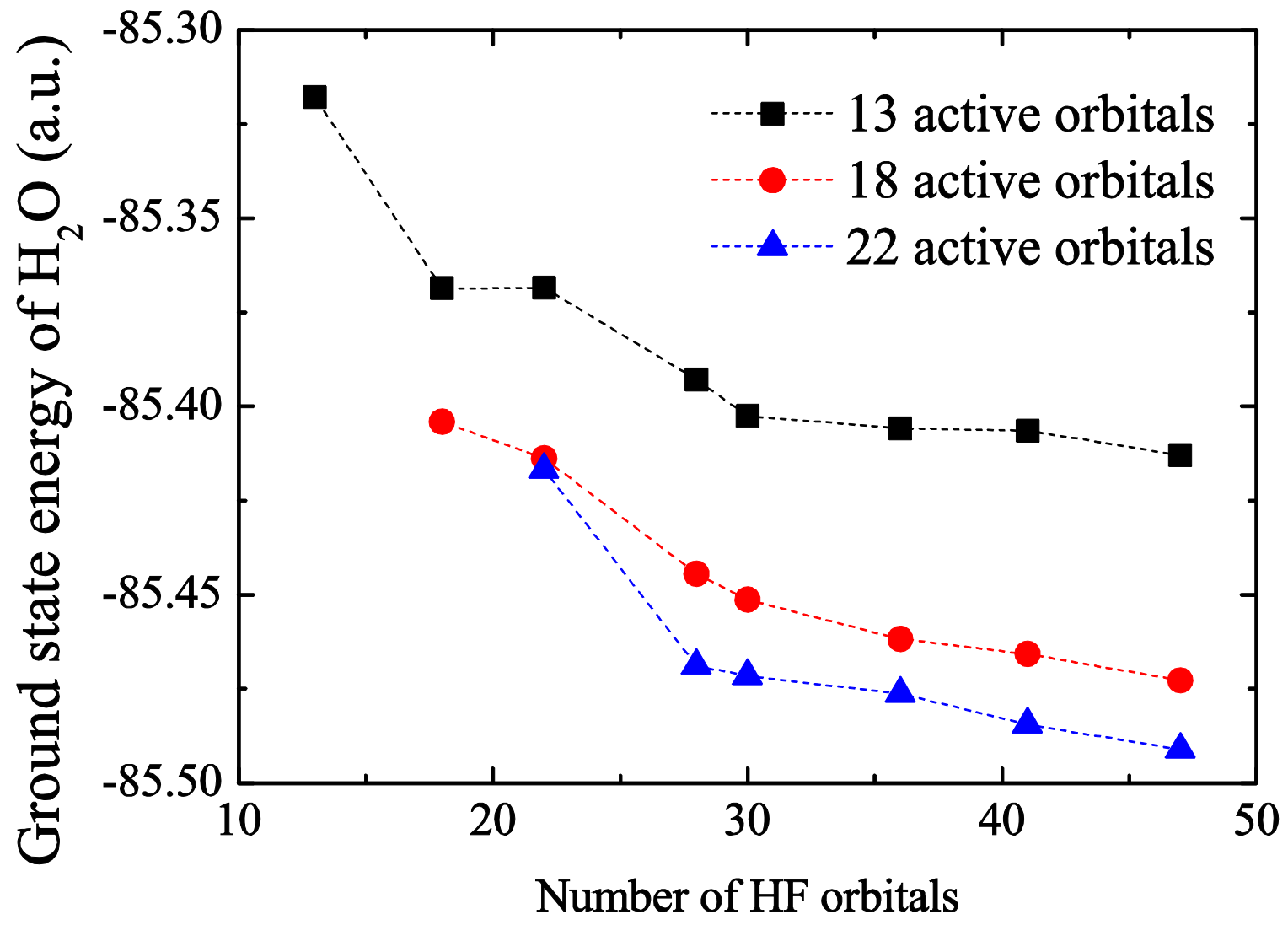




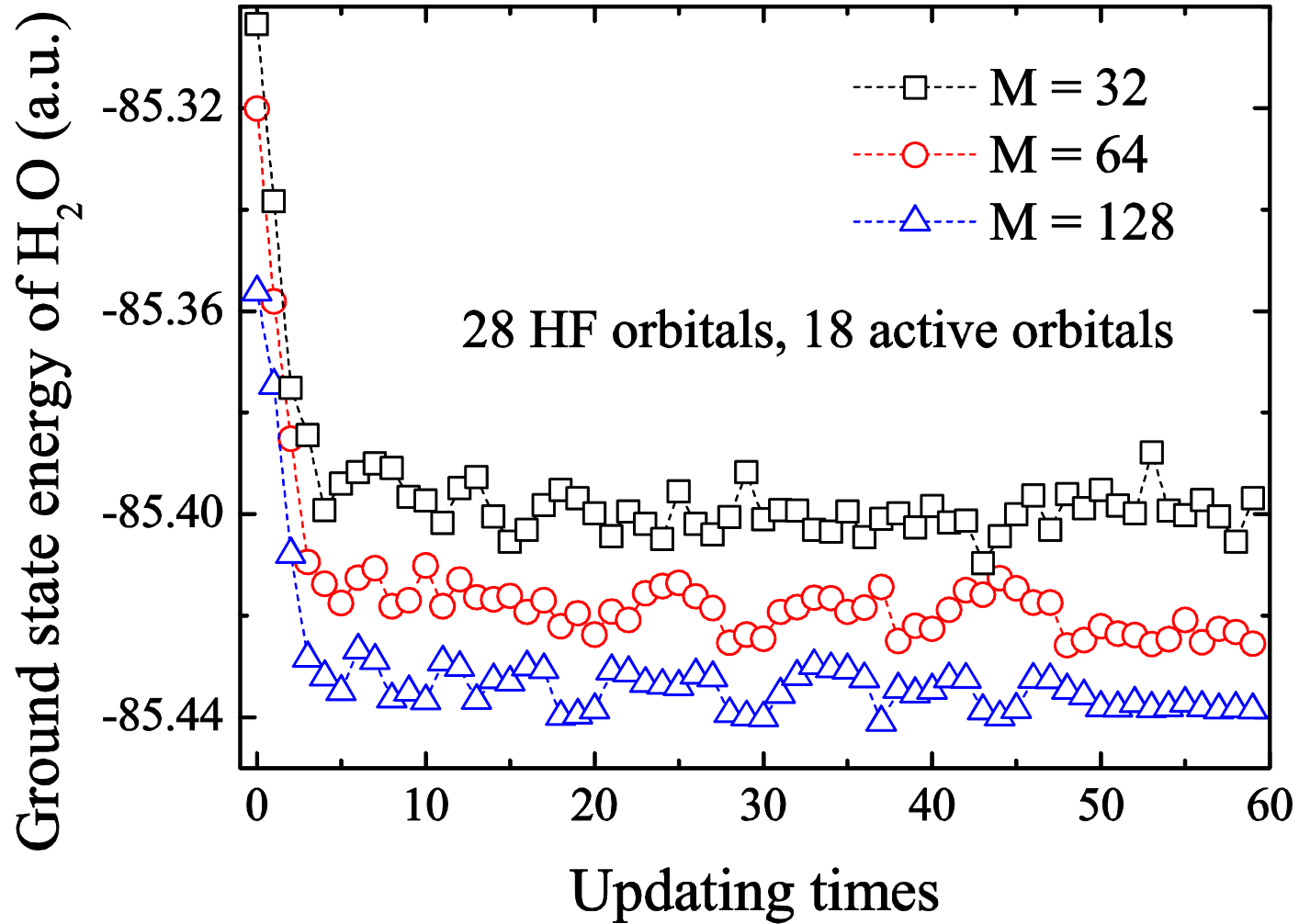
Some details

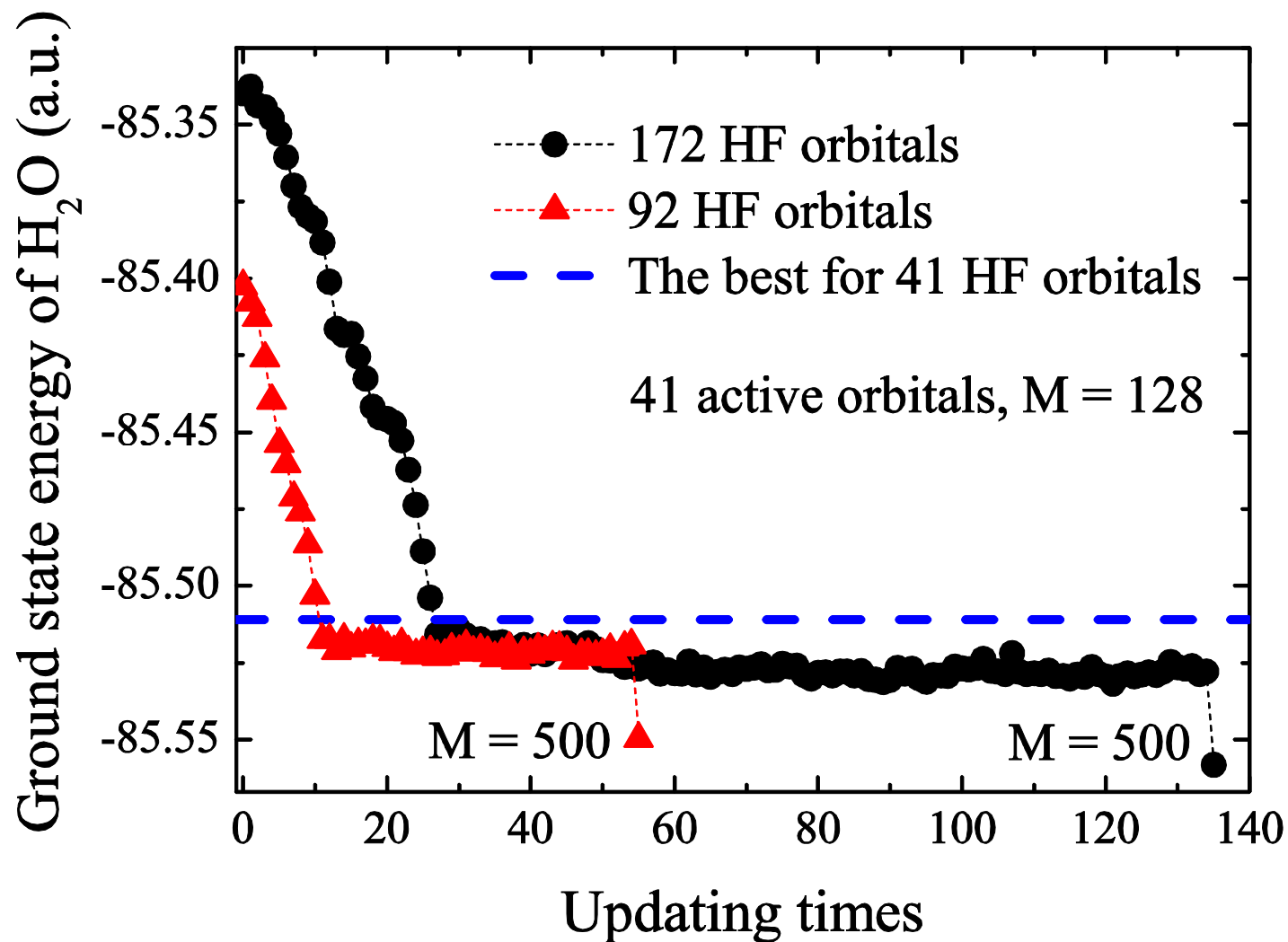


Each point is a full cycle of DMRG iteration



M = 128





After the single-particle basis is optimized, the result is significantly improved with much cheaper cost.

H.-G. Luo, M.-P. Qin, T. Xiang, PRB **81**, 235129 (2010); arXiv:1002.1287

# Ground state energy of water molecule

$$E_{\text{gs}} = -85.575(\text{a.u.})$$

( $N_{\text{tot}} = 172$  orbitals,  $N = 61$  orbitals,  $M = 128$ ,  $M_{\text{max}} = 512$ )

Orbital #: $N$	Unrestricted Hartree-Fock	Coupled Cluster (SDT)	DMRG <sup>1</sup>	Quantum Monte Carlo <sup>2</sup>	Exp
41	-85.254555	-85.511075	-85.511649 ( $M=6000$ )	-85.5132(6)	-85.677
92	-85.257519	-85.564462		-85.567(1)	?

- The result agrees with the CCSDT and QMC results with  $N=92$
- DMRG has great potential to further improve the result

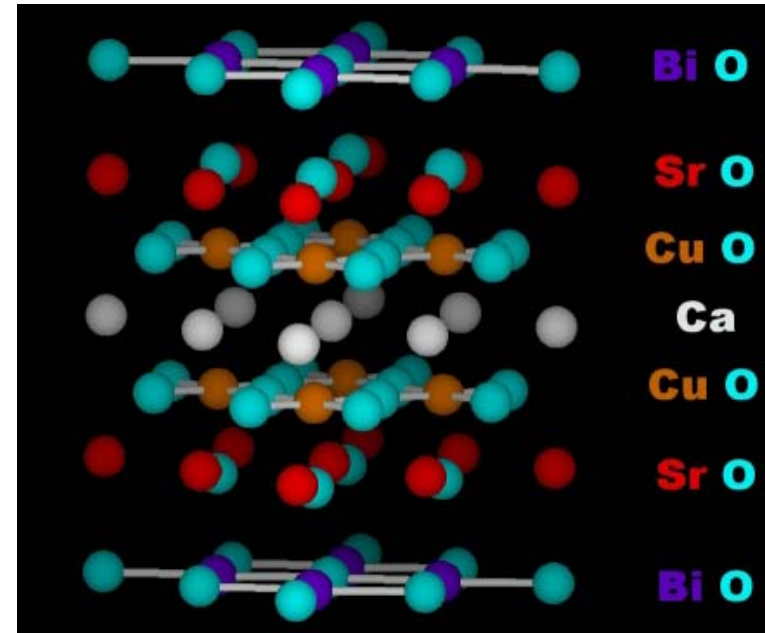
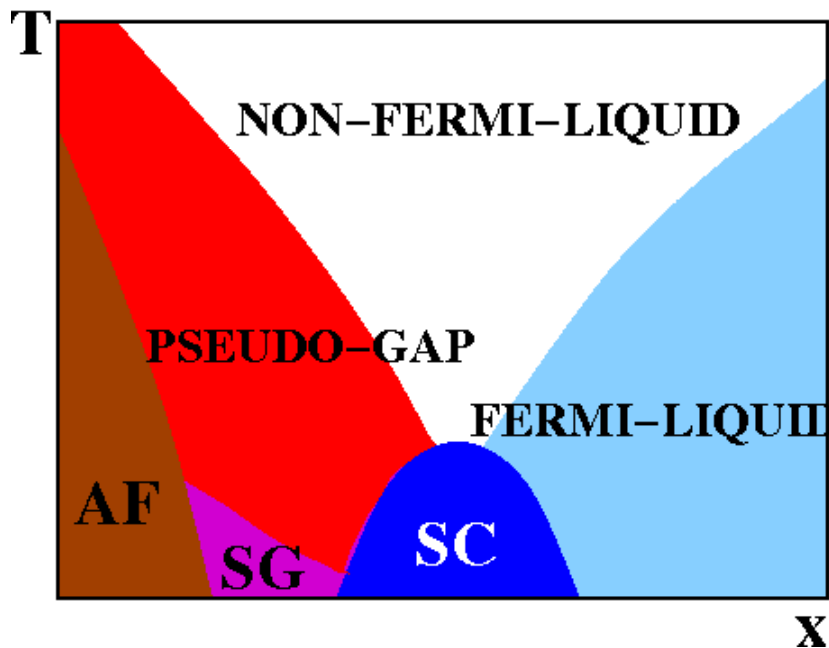
## Outline:

- 1) Density matrix renormalization group with long-range interaction
- 2) The t-J-V-W model realized in polar molecule systems
- 3) Some preliminary results

# Why t-J model ?

An important model for strongly correlated electron systems, especially for the theoretical description of high-temperature superconductors (HTS).

$$H = -t \sum_{\langle i,j \rangle, \sigma} (c_i^+ c_j + h.c.) + J \sum_{\langle i,j \rangle} \left( S_i^+ S_j^- + S_i^z S_j^z - \frac{1}{4} n_i n_j \right)$$



2-d plane of HTS

Schematic phase diagram of high-T<sub>c</sub> superconductors



# Why polar molecule ?

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A polar molecular in a dc electric field is describe by the rigid-rotor Hamiltonian:

$$H_0 = B\hat{N}^2 - d_0 E$$

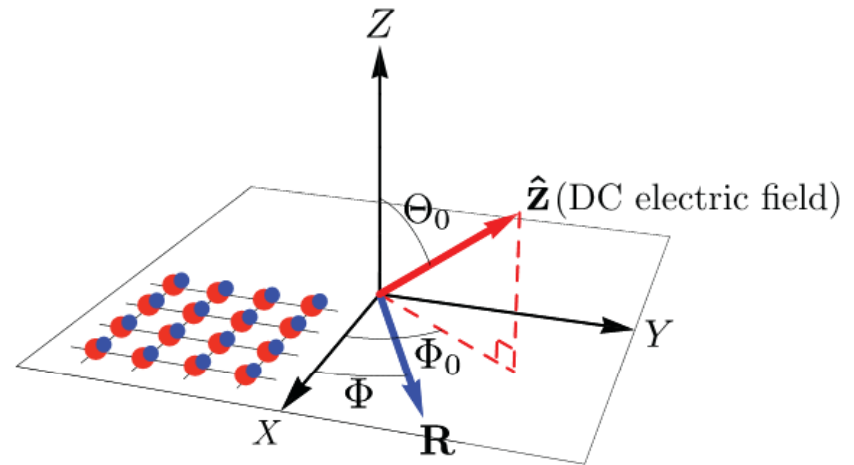
with the rotational constant  $B$  and the angular momentum operator  $N$ .

At  $E=0$ ,  $H_0$  has eigenstates  $|N, M\rangle$ :

$$\hat{N}^2 |N, M\rangle = N(N+1) |N, M\rangle$$

$$N_z |N, M\rangle = M |N, M\rangle$$

while  $M = -N, \dots, 0, \dots, N$ .



A.V.Gorshkov *et al.* Phys. Rev. A  
84, 033619 (2011)

*Two important features:*

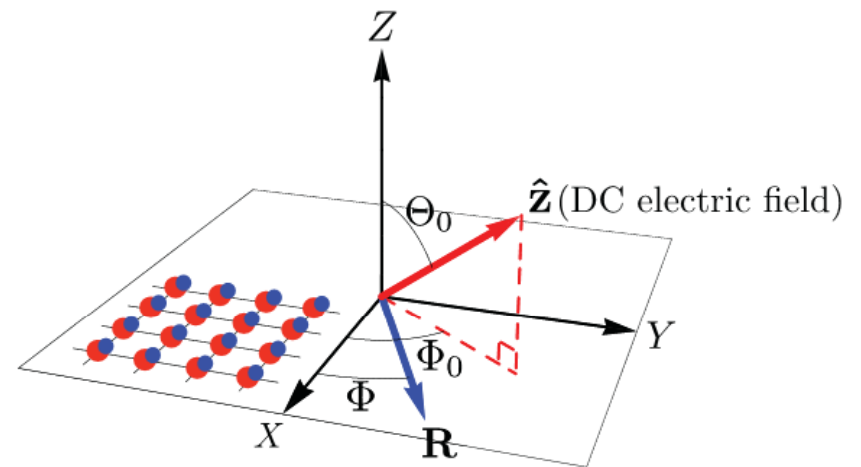
- 1) *Permanent dipole moment  $\rightarrow$  manipulated with external fields*
- 2) *Rich internal level structure of diatomic polar molecules  $\rightarrow$  Microwave field control*

$$\begin{aligned}
E_{dd} &= \frac{1}{4\pi\epsilon_0|\mathbf{R}_i - \mathbf{R}_j|^3} [\boldsymbol{\mu}_i \cdot \boldsymbol{\mu}_j - 3(\boldsymbol{\mu}_i \cdot \hat{\mathbf{z}})(\boldsymbol{\mu}_j \cdot \hat{\mathbf{z}})] \\
&= V_{dd}(\mathbf{R}_i - \mathbf{R}_j)(\mu_0 n_{im_0} + \mu_1 n_{im_1})(\mu_0 n_{jm_0} + \mu_1 n_{jm_1}) \\
&= V_{dd}(\mathbf{R}_i - \mathbf{R}_j)[J_z S_i^z S_j^z + V n_i n_j + W(n_i S_j^z + n_j S_i^z)]
\end{aligned}$$

where  $J_z = (\mu_0 - \mu_1)^2$ ,  $V = (\mu_0 + \mu_1)^2/4$ , and  $W = (\mu_0^2 - \mu_1^2)/2$ . The  $V$  term describes density-density interactions, and

$$\boldsymbol{\mu}_i = (\mu_0 n_{im_0} + \mu_1 n_{im_1}) \hat{\mathbf{z}}$$

$$\boldsymbol{\mu}_j = (\mu_0 n_{jm_0} + \mu_1 n_{jm_1}) \hat{\mathbf{z}}$$



A.V.Gorshkov *et al.* Phys. Rev. A  
84, 033619 (2011)

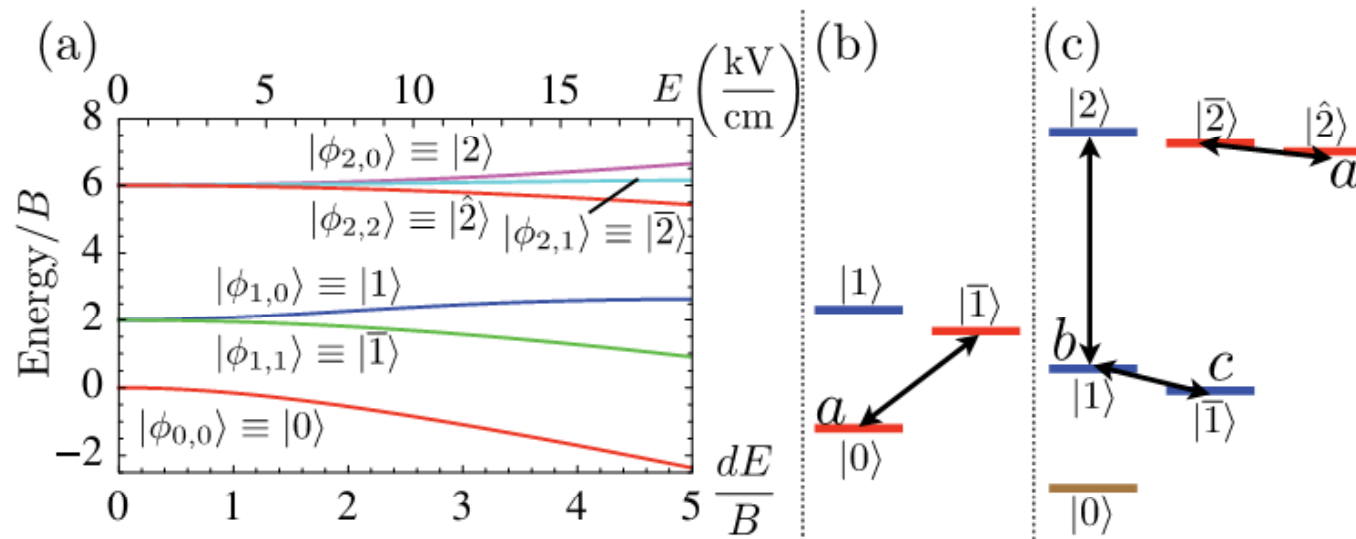
Combining the microwave fields and the external dc electric fields, plus the tunneling effect, the magnitude and its sign can be tuned independently

$$\begin{aligned}
 H = & - \sum_{\langle i,j \rangle m \sigma} t_m [c_{im\sigma}^\dagger c_{jm\sigma} + \text{H.c.}] + \frac{1}{2} \sum_{i \neq j} V_{\text{dd}}(\mathbf{R}_i - \mathbf{R}_j) \\
 & \times \left[ J_z S_i^z S_j^z + \frac{J_\perp}{2} (S_i^+ S_j^- + S_i^- S_j^+) + V n_i n_j \right. \\
 & \left. + W (n_i S_j^z + n_j S_i^z) \right].
 \end{aligned}$$

A. V. Gorshkov et al., PRL 107, 115305 (2011)

A. V. Gorshkov et al., PRA 84, 033619 (2011)

# a generation of t-J model in experiment



(a) Eigenenergies as a function of  $E$ ;  
Examples of level configurations  $\{|m_0\rangle, |m_1\rangle\}$

(b)  $\{\sqrt{a}|0\rangle + \sqrt{1-a}|\bar{1}\rangle, |1\rangle\}$

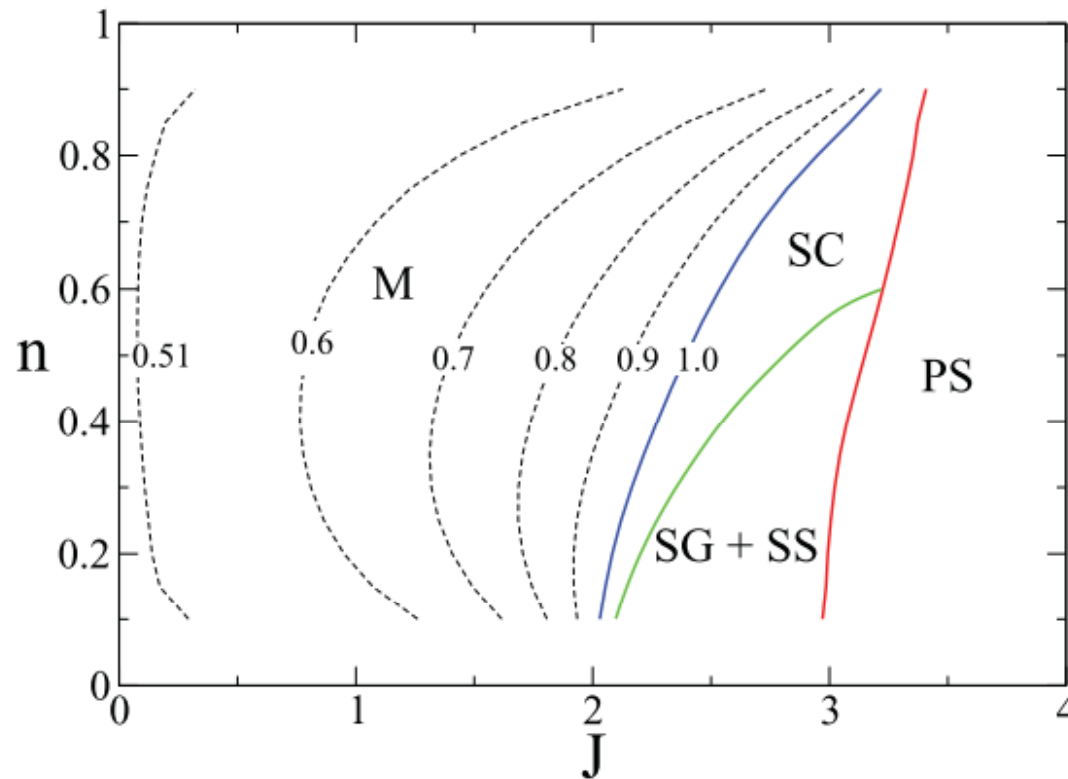
(c)  $\{\sqrt{a}|\hat{2}\rangle + \sqrt{1-a}|\bar{2}\rangle, \sqrt{b}|1\rangle + \sqrt{c}|\bar{1}\rangle + \sqrt{1-b-c}|2\rangle\}$

where the coefficient  $a, b, c$  in the dressed state  $|m_0\rangle, |m_1\rangle$  is controlled by the ratio between the Rabi frequency and the detuning of a  $\sigma^+$ -polarized microwave field acting on the  $|0\rangle \leftrightarrow |\bar{1}\rangle$  transition.

A.V.Gorshkov *et al.* Phys. Rev. Lett 107, 115301 (2011)

# Ground phase diagram of 1-d t-J model with nearest-neighbor interaction

The phases of 1-d t-J model display a correspondence to the ones present in HTS of 2-d case.



Metallic phase

gapless superconducting phase

singlet-superconducting phase with spin gap

phase separation

A. Moreno *et al.* Phys. Rev. B 83, 205113 (2011)

**Motivations of our work with long-range interaction:**

- 1) Enlarge the superconducting region;*
- 2) Challenge for density matrix renormalization group (DMRG).*

# phase diagram of 1-d t-J-V-W model

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We consider a simple case of the t-J-V-W model that  
 $J_z = J_\perp = V$  and  $W = 0$  :

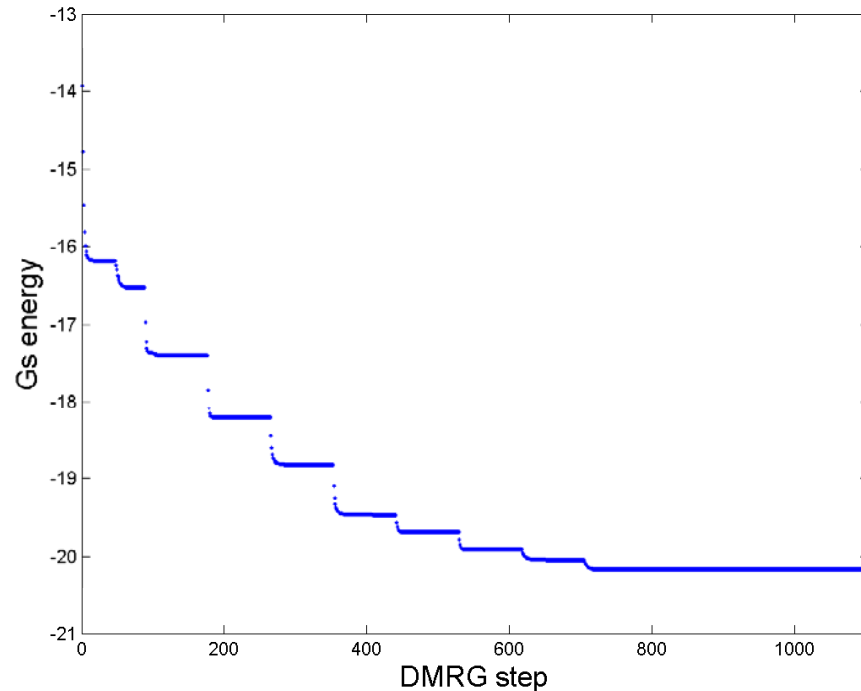
$$H = -t \sum_{\langle i,j \rangle, \sigma} (c_i^\dagger c_j + h.c.) + \sum_{i \neq j} |R_i - R_j|^{-3} J \left( S_i^+ S_j^- + S_i^z S_j^z - \frac{1}{4} n_i n_j \right)$$

We set  $t=1$ .

Calculations are taken in the parameter space of  $n=0.1 \sim 0.9$   
with  $\Delta n=0.1$  and  $J=0.25 \sim 4.0$  with  $\Delta n=0.25$ .

# DMRG techniques

## What is the reasonable number of sweeps?



Ground state energy as a function of dmrp steps is showed in left figure. Energy leap occurs between each two neighbor sweeps.

We define  $\langle E(i) \rangle$  as the average energy of sweep  $i$  and the energy error:

$$Error(i) = \frac{\langle E(i-1) \rangle - \langle E(i) \rangle}{\langle E(i) \rangle}$$

In our calculation, dmrp process complete when  $Error(i) < 10^{-8}$ , number of sweeps float from 11 to 67.



# phase diagram of 1-d t-J-V-W model

---

DMRG directly get the wavefunction and ground state energy of the system. All observables should be calculated by:

$$O = \langle \Psi | \hat{A} | \Psi \rangle$$

1-d system with weak interactions could be will described by the Luttinger-liquid theory, that its correlation functions can be explicitly evaluated.

Main structure factors and correlation functions:

		$X(k) = \frac{1}{L} \sum_{i,j=1}^L e^{ik(x_i-x_j)} X_{ij}$
density-density correlation:	$N_{ij} = \langle n_i n_j \rangle - \langle n_i \rangle \langle n_j \rangle$	$N(k)$
spin-spin correlation:	$S_{ij} = \langle S_i^z S_j^z \rangle$	$S(k)$
Green's function:	$G_{ij}^\sigma = \langle c_{i,\sigma}^+ c_{j,\sigma} \rangle$	$n(k)$ (momentum distribution)

# Metal and SC

Luttinger parameter  $K_\rho$  could be the order parameter to characterize the phase of metallic ( $K_\rho < 1$ ) and superconducting ( $K_\rho > 1$ ) in Luttinger liquid region.

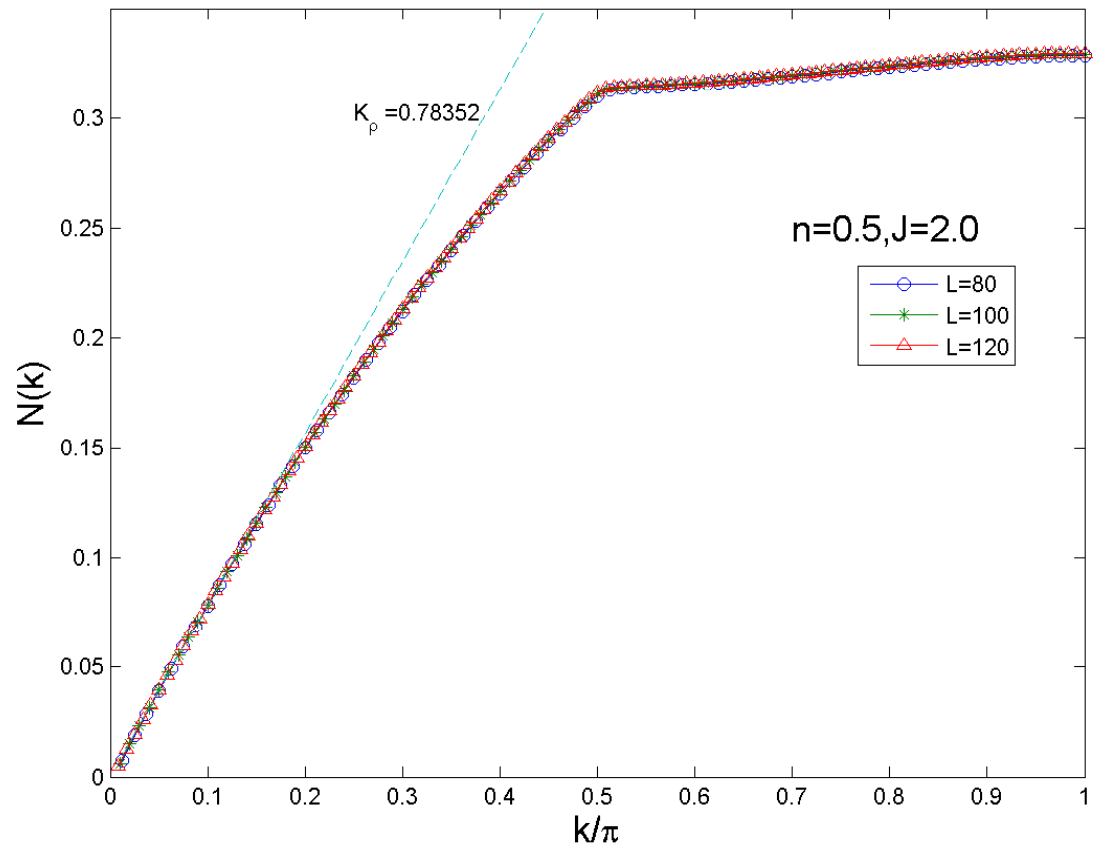
$K_\rho$  is determined by the relationship:

$$N(k) \rightarrow \frac{a|k|}{\pi} K_\rho \quad \text{when } k \rightarrow 0$$

where lattice constant  $a=1$ .

A clear linear behavior for small  $k$  can be observed in right figure.

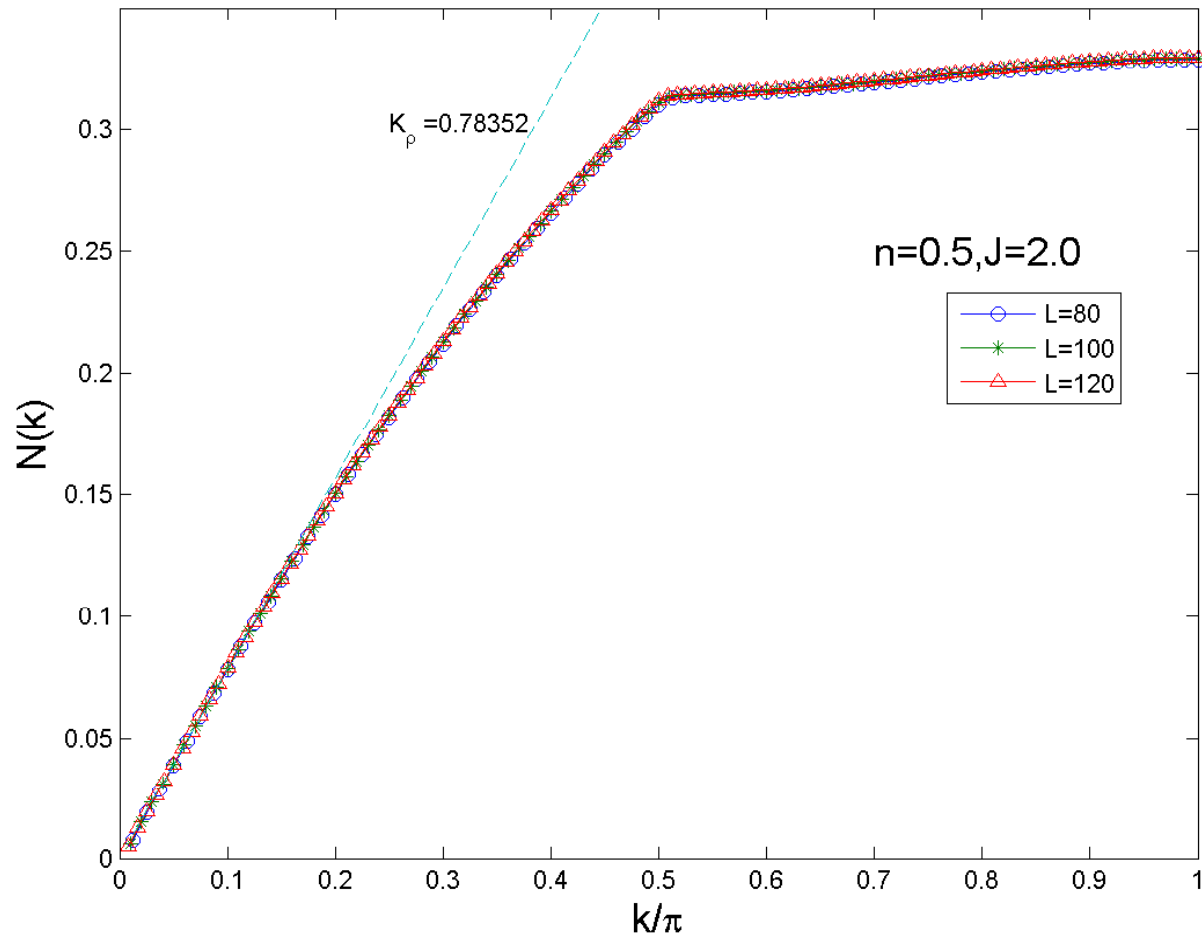
In our calculation, we choose:  
 $K_\rho = N(k=0.05\pi)$ .



Structure factor  $N(k)$  of density-density correlation

# Metal and SC

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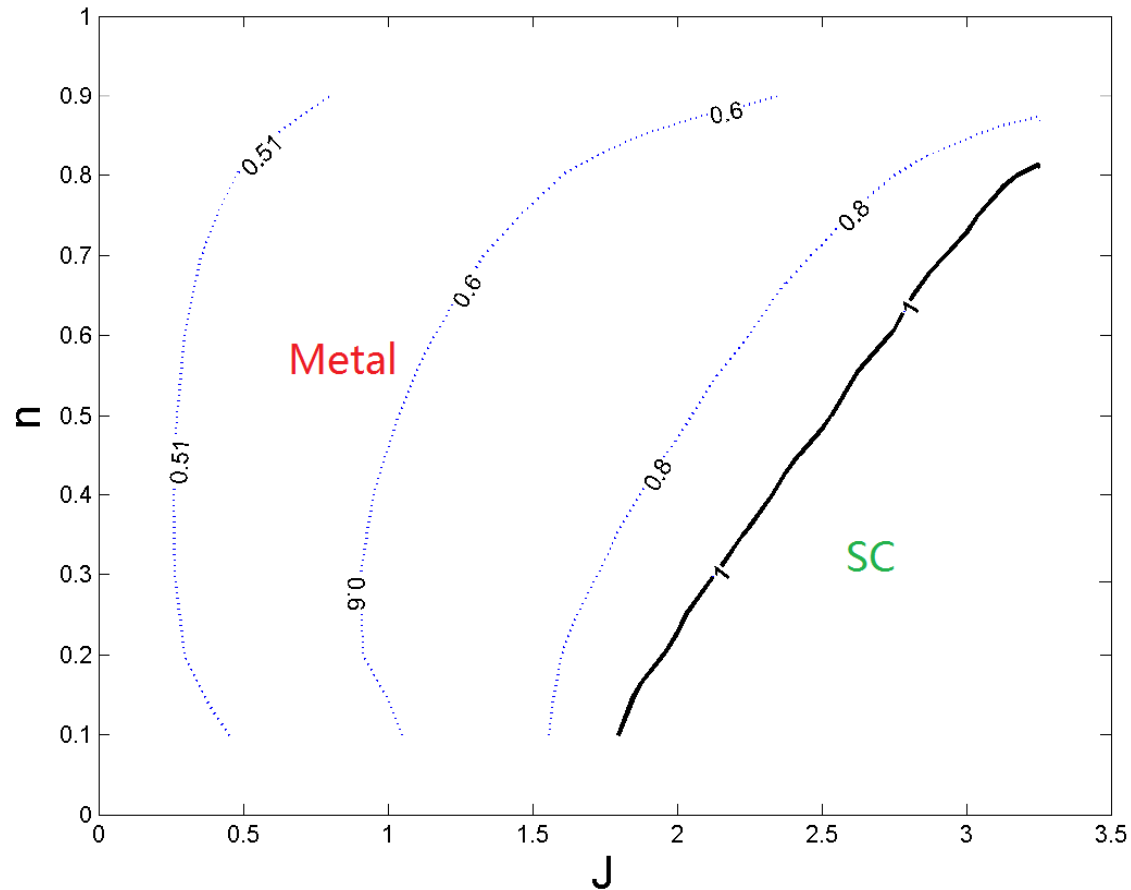
Structure factor  $N(k)$  of density-density correlation

$N(k)$  appears to be almost independent of the lattice size, as shown in this figure.

Results showed in the report we choose  $L=100$  as the approximation of thermal dynamics limit.

# Metal and SC

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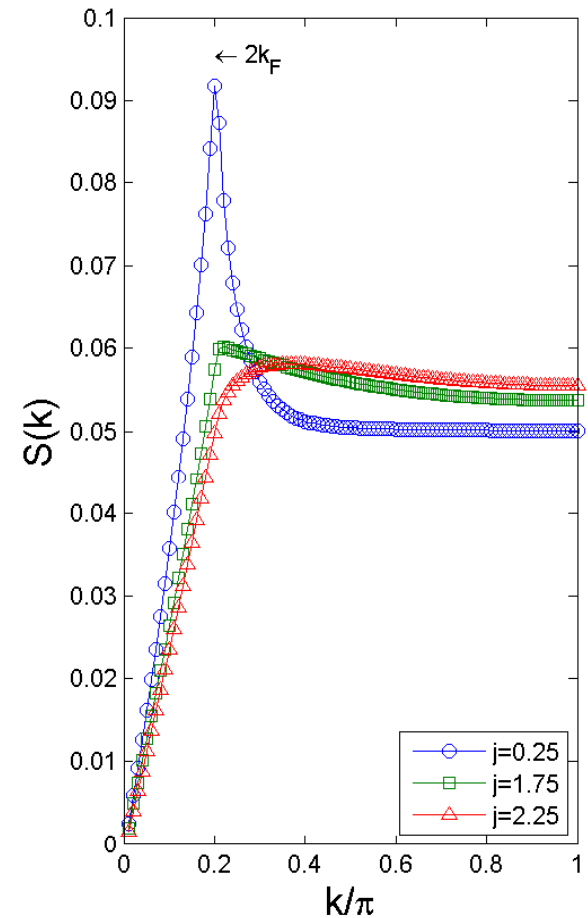
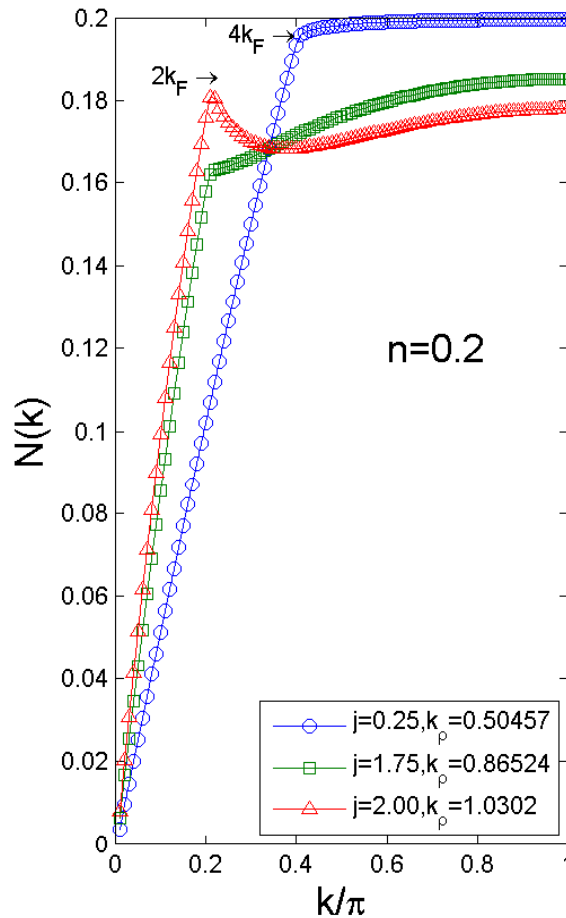
A contour plot of  $K_\rho$ .  
 $K_\rho=1$  as the phase boundary between metal and  
superconductor.

# Metal and SC

## Characteristic peaks of the structure factors

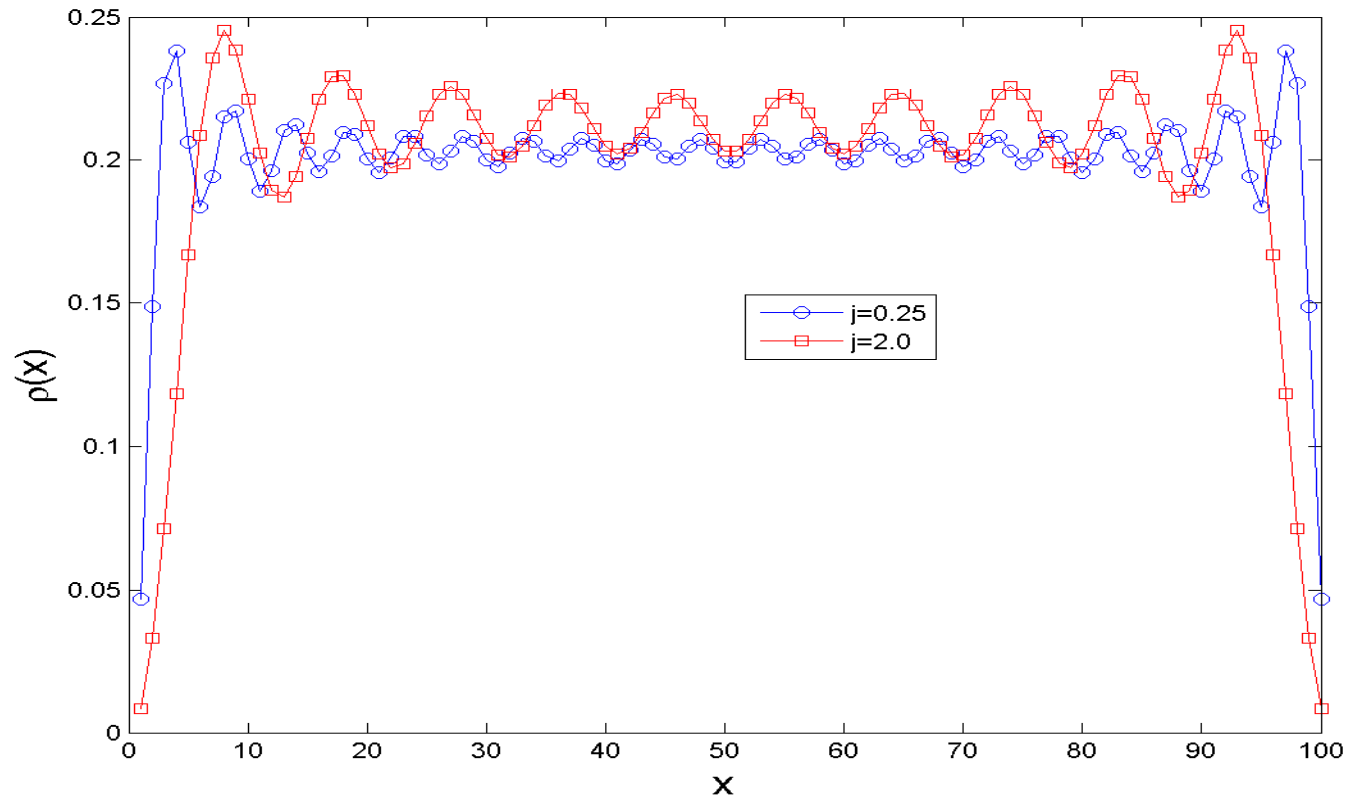
$N(k)$ :  
 $4k_F$  peak in metallic phase  
as  $J$  increases, this peak  
is suppressed and a  $2k_F$   
cusp form  
in superconducting phase

$S(k)$ :  
 $2k_F$  peak in metallic phase  
(antiferromagnetic order  
means no SC)



# Metal and SC

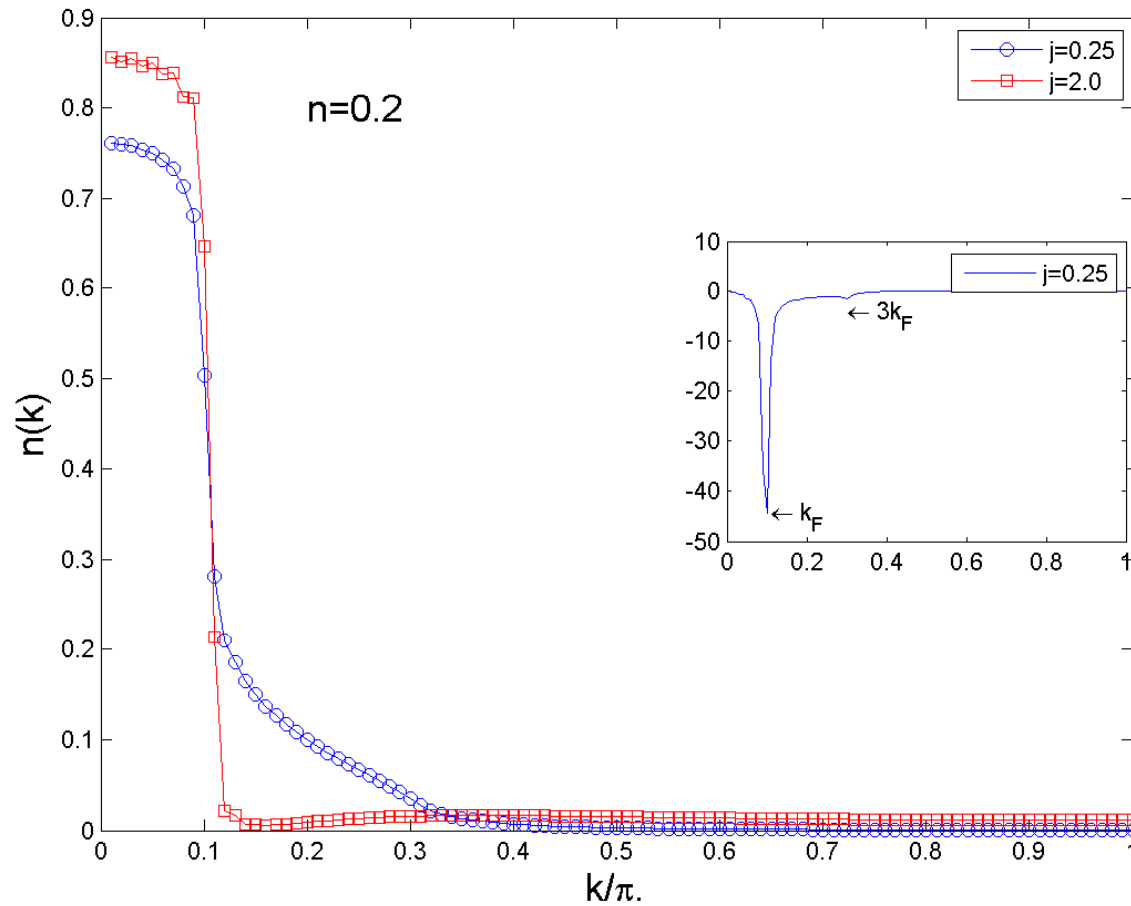
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Real space density.

Either metallic or superconducting phase shows charge density wave behavior (Friedel oscillations) in real space.

# Metal and SC



Momentum distribution in  $k$  space.

There is a clear fermi surface in both case, and a weak anomaly at  $3k_F$ .

Superconducting states more fermi?

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