Computing Resonant Inelastic X-ray Scattering spectra using the Density Matrix Renormalization Group Method

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Computing Resonant Inelastic X-Ray Scattering Spectra Using The Density Matrix Renormalization Group Method

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Poster PT-28

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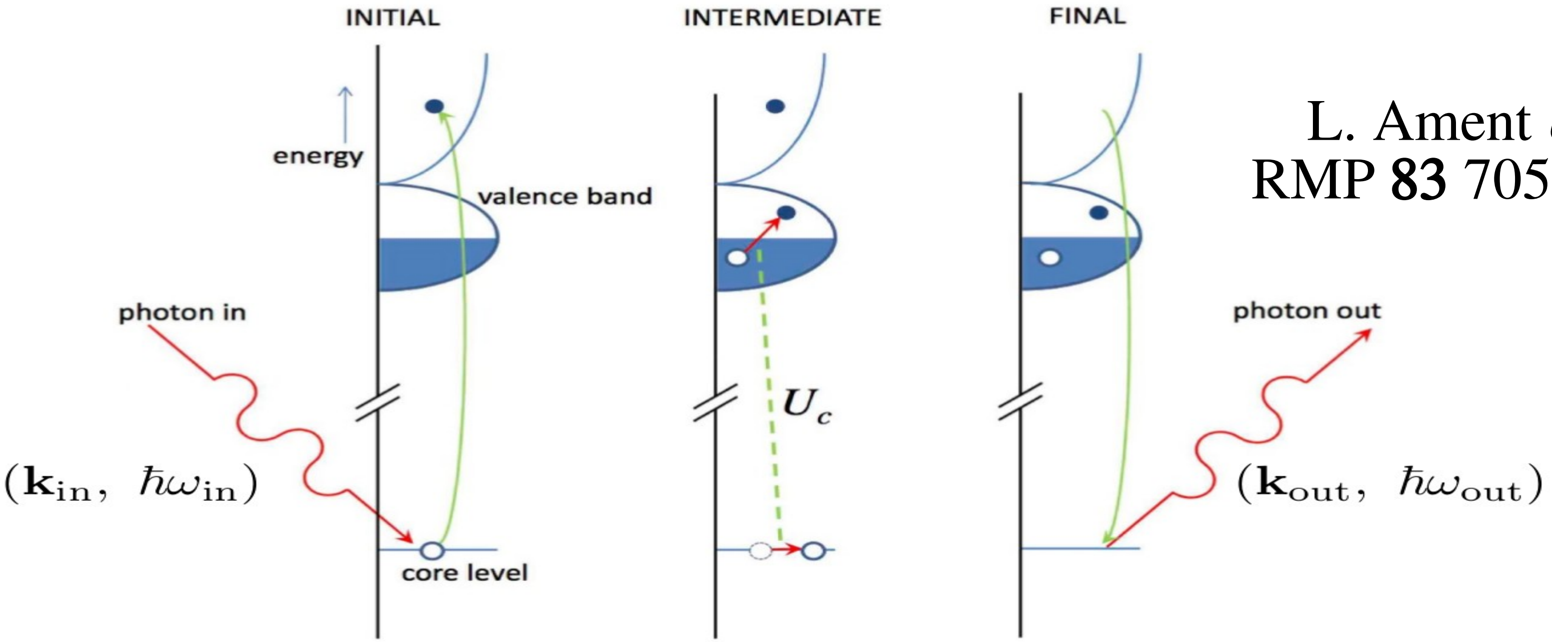
Outline

- Computing RIXS spectra for strongly correlated materials
  -Why is it challenging? Need for Exact Numerical methods

- Beyond Exact Diagonalization methods
  -dynamical spectra with DMRG: can we compute RIXS spectra?

- Krylov “trick”: Computing **full** RIXS spectra using DMRG
  -Application to quasi-1D cuprates

How to compute RIXS spectra?



L. Ament *et al.*
RMP **83** 705 (2011)

Transition Rate in "resonance" (Kramers-Heisenberg)

$$W_{g\to f} = \sum_f \left| \sum_n \frac{\langle f|\hat{H}'|n\rangle\langle n|\hat{H}'|g\rangle}{E_g - E_n} \right|^2 \delta(E_f - E_g)$$

$\hat{H}' =$ light-matter interaction

How we actually compute RIXS spectra

$$I_{\mathrm{RIXS}}(\mathbf{q}, \hbar\Omega) \propto \sum_f \left| \sum_{j,n} \frac{\langle f|\hat{D}_j^\dagger(\mathbf{k}_{\mathrm{out}})|n\rangle\langle n|\hat{D}_j(\mathbf{k}_{\mathrm{in}})|g\rangle}{\hbar\omega_{\mathrm{in}} - E_n^{\mathrm{ch}} + E_g + i\hbar\Gamma} \right|^2 \delta(\hbar\Omega - E_f + E_g)$$

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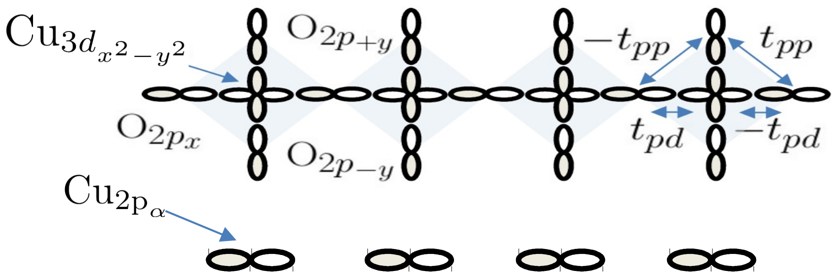
Multiband Hubbard model for valence band and core electrons



$$\hat{H}|\psi\rangle = E|\psi\rangle$$

$|g\rangle \rightarrow$ ground state $\rightarrow \hat{H}, E_g$

$|f\rangle \rightarrow$ final state $\rightarrow \hat{H}, E_f$

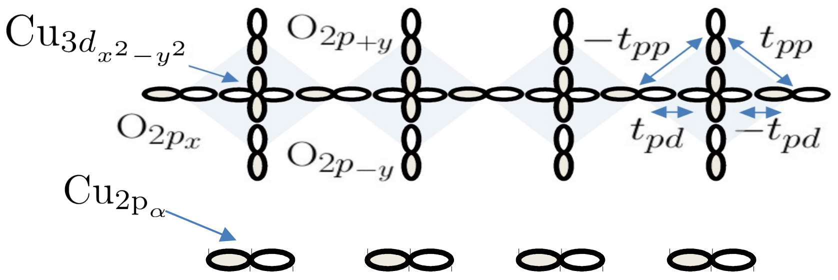
$$\hat{H}^{\mathrm{ch}}|n\rangle = E_n^{\mathrm{ch}}|n\rangle$$

$|n\rangle \rightarrow$ intermediate states $\rightarrow \hat{H}^{\mathrm{ch}}, E_n^{\mathrm{ch}}$

How we actually compute RIXS spectra

$$I_{\mathrm{RIXS}}(\mathbf{q},\hbar\Omega) \propto \sum_f \left| \sum_{j,n} \frac{\langle f|\hat{D}_j^\dagger(\mathbf{k}_{\mathrm{out}})|n\rangle\langle n|\hat{D}_j(\mathbf{k}_{\mathrm{in}})|g\rangle}{\hbar\omega_{\mathrm{in}} - E_n^{\mathrm{ch}} + E_g + i\hbar\Gamma} \right|^2 \delta(\hbar\Omega - E_f + E_g)$$

Multiband Hubbard model for valence band and core electrons



$$\hat{H}|\psi\rangle = E|\psi\rangle$$

$|g\rangle \rightarrow$ ground state $\rightarrow \hat{H}, E_g$

$|f\rangle \rightarrow$ final state $\rightarrow \hat{H}, E_f$

$$\hat{H}^{\mathrm{ch}}|n\rangle = E_n^{\mathrm{ch}}|n\rangle$$

$|n\rangle \rightarrow$ intermediate states $\rightarrow \hat{H}^{\mathrm{ch}}, E_n^{\mathrm{ch}}$

We need to solve two difficult many-body problems!

Standard solution: “brute force” Exact Diagonalization

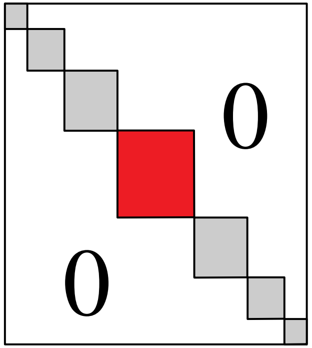
Schrödinger's Equation:

$$\hat{H}|\psi\rangle = E|\psi\rangle$$

Example

16 orbitals, Hubbard model

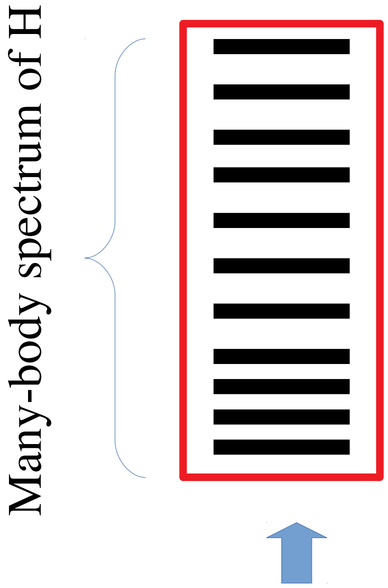
Size Hilbert space= 4^{16}~ 4.3x10^9



After symmetries:

Half-filling (8↑ and 8↓), translation

Size Hilbert Space ~ 1.0x10^7



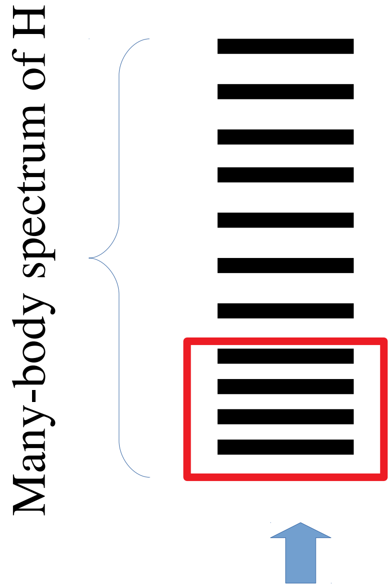
Powerful, but severely limited in system size!

We get the ALL the eigenstates

Standard iterative solution: Lanczos Exact Diagonalization

Schrödinger's Equation:

$$\hat{H}|\psi\rangle = E|\psi\rangle$$



We get the **low-energy** eigenstates

Project $\hat{H}$ onto the **Krylov** space iteratively

$$|\alpha_{n+1}\rangle = \hat{H}|\alpha_n\rangle - a_n|\alpha_n\rangle - b_n^2|\alpha_{n-1}\rangle$$

$$\hat{T} = \langle\alpha_n|\hat{H}|\alpha_m\rangle = \begin{pmatrix} a_1 & b_2 & & & & 0 \\ b_2 & a_2 & b_3 & & & \\ & b_3 & a_3 & \ddots & & \\ & & \ddots & \ddots & b_{k-1} & \\ & & & b_{k-1} & a_{k-1} & b_k \\ 0 & & & & b_k & a_k \end{pmatrix} \longrightarrow \text{Tridiagonal form!}$$

k<<Hilbert space size!

$$\hat{H} \simeq \hat{V}\hat{T}\hat{V}^\dagger = [\hat{V}\hat{W}]\hat{D}[\hat{W}^\dagger\hat{V}^\dagger] \longrightarrow \text{Diagonal form!}$$

Computation of RIXS spectra: Exact Diagonalization method

ED methods can obtain **excellent** results for small clusters

Long (not complete!) list of papers using ED methods
References for the 1D cuprates only

Okada, et al. Phys. Rev. B **63**, 045103 (2001).
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Tohyama, et al. Phys. Rev. B **92**, 014515 (2015).
Johnston et al. Nat. Commun. **7**, 10563 (2016).
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Schlappa, et al. Nat. Comm .**9**, 5394 (2018) .
Kumar, et al. New J. Phys. **20**, 073019 (2018).
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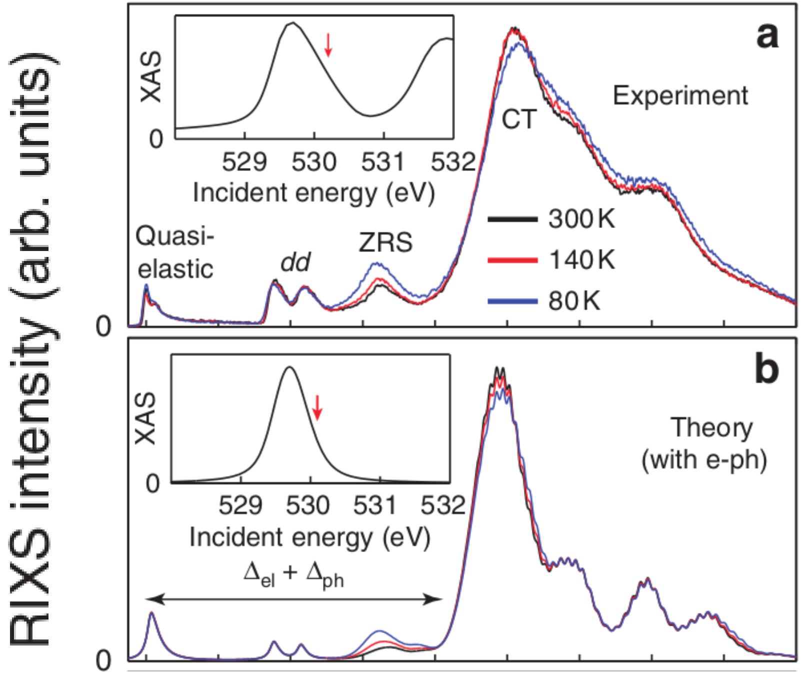
Li_2CuO_2@ O K-edge



S. Johnston et al.
Nat. Comm. **7**, 10563 (2016)
Cu_3O_8 cluster
(~25 orbitals + phonons)

Computation of RIXS spectra: Exact Diagonalization method

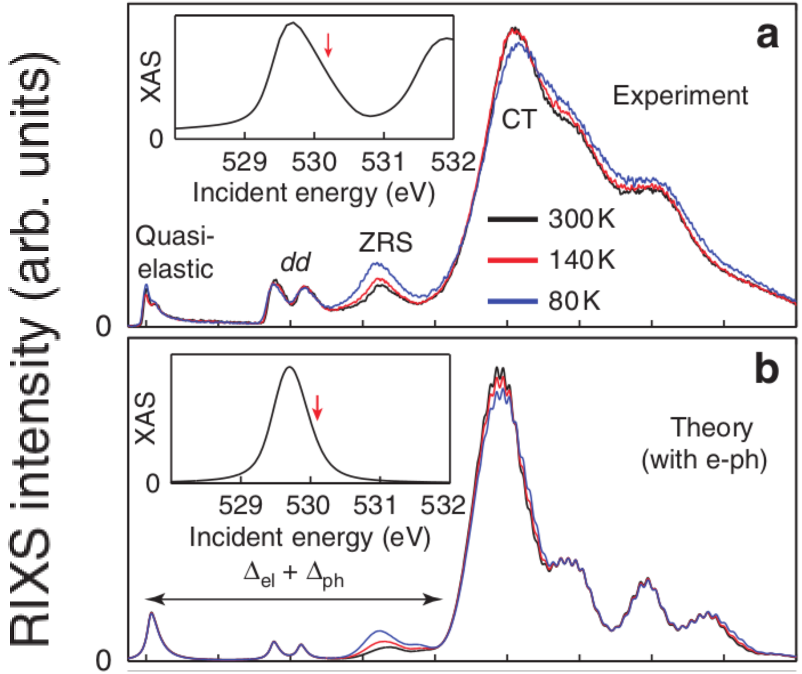
ED methods can obtain **excellent** results for small clusters

BUT

Limited resolution in momentum space!

CAN WE OVERCOME THIS PROBLEM?

Li_2CuO_2 @ O K-edge



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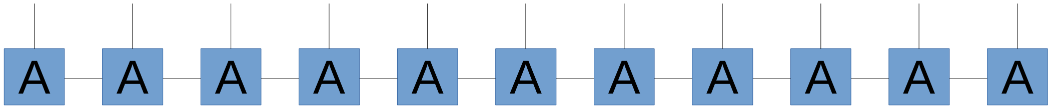
Beyond Exact Diagonalization methods: DMRG

S.R.White PRL **69** 2863 (1992)

- Variational method to "search" the **ground state** of arbitrary Many-Body Hamiltonians

- Matrix Product State (MPS) Anzatz for the wave-function for a lattice of L sites

$$|\psi\rangle = \sum_{f_1,\dots,f_L} A^{f_1} A^{f_2} \dots A^{f_{L-1}} A^{f_L} |f_1, \dots, f_L\rangle$$



- ADVANTAGE OVER ED METHODS: solution for **hundreds** of lattice orbitals!

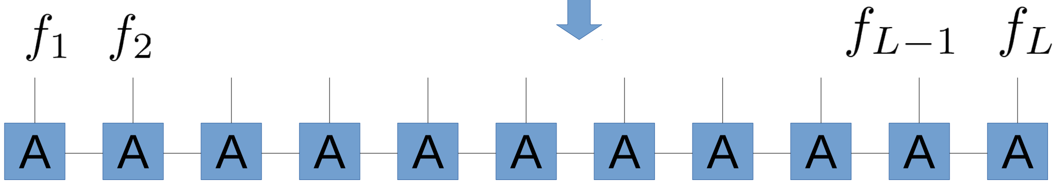
Review: U. Schollwöck, Annals of Physics. **326** 96–192 (2011)

Quantum many body wave-functions as Matrix Product States

How? Singular Value Decomposition!

$$|\psi\rangle = \sum_{f_1,\ldots,f_L} c^{f_1,\ldots,f_L} |f_1, \ldots, f_L\rangle \Rightarrow |\psi\rangle = \sum_{f_1,\ldots,f_L} A^{f_1} A^{f_2} \ldots A^{f_{L-1}} A^{f_L} |f_1, \ldots, f_L\rangle$$

Graphical representation of MPS:

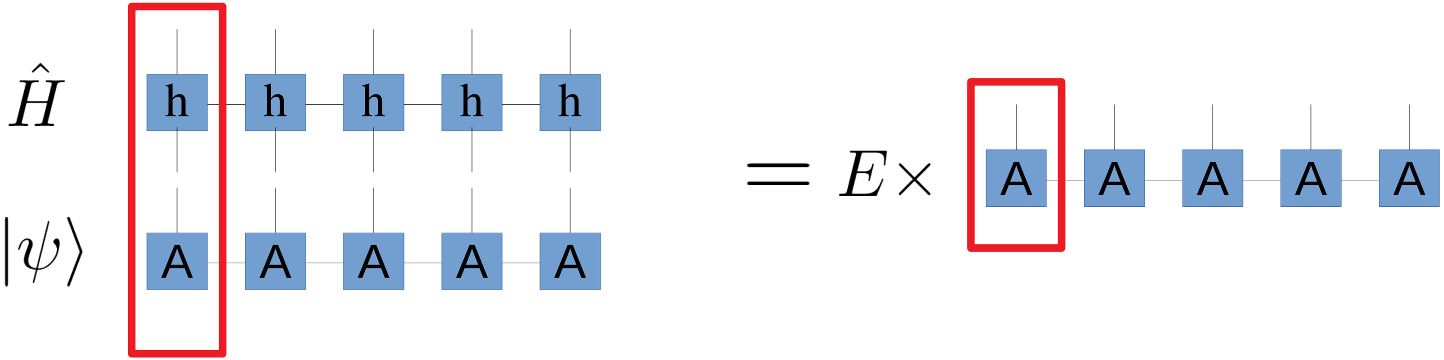


What’s the advantage of all this?
No approximations yet, still 4^L coefficients!

Review: U. Schollwöck, Annals of Physics. **326** 96–192 (2011)

Variational Optimization of an MPS: DMRG

$$\hat{H}|\psi\rangle = E|\psi\rangle$$



Tensors “A” can be optimized **one by one** “sweeping” back and forth through the lattice!

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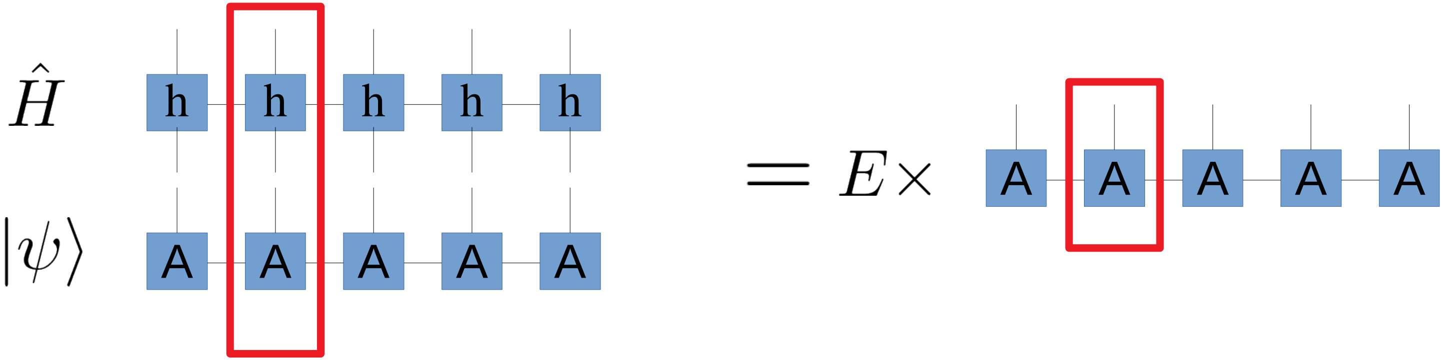

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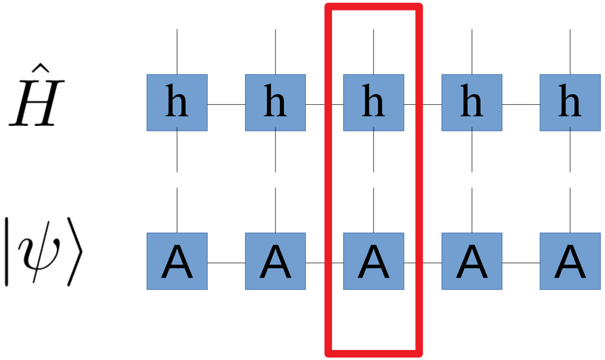
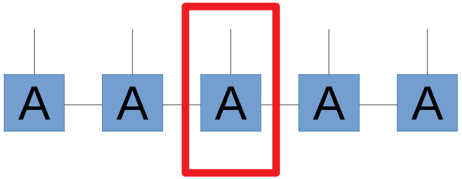


$= E\times$



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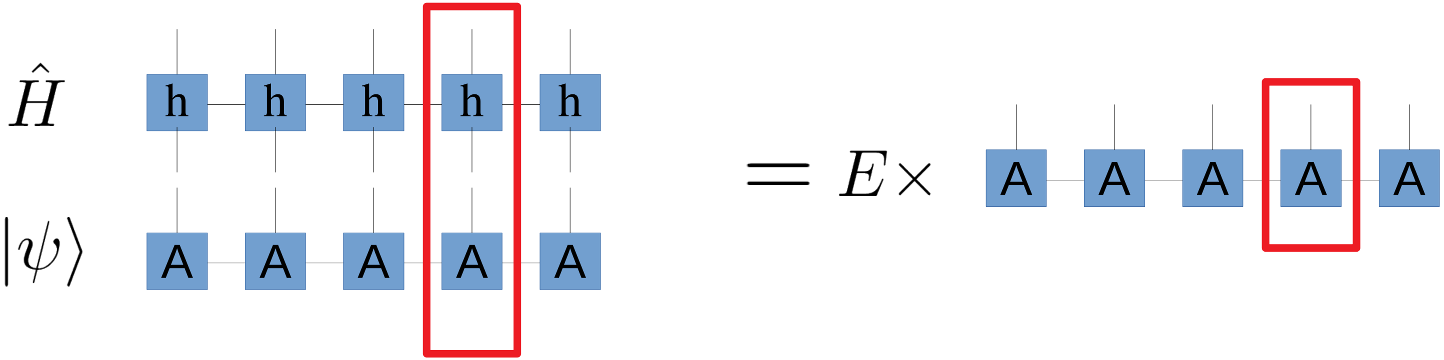

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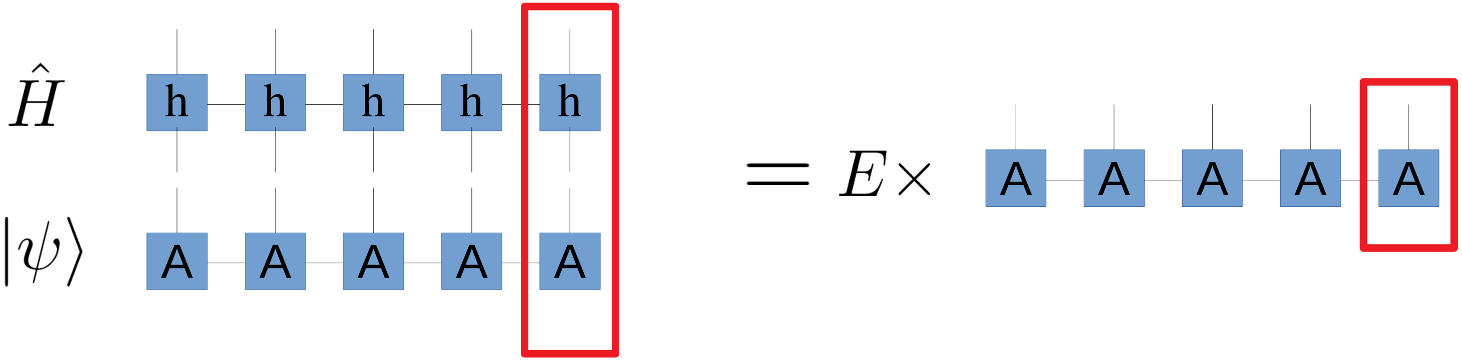

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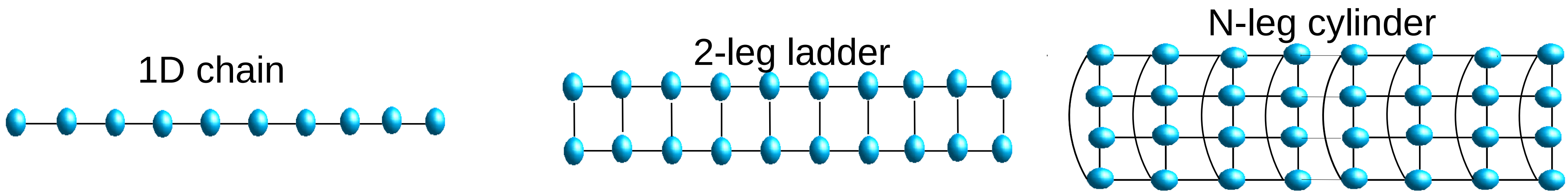
What can we do with DMRG?

- **Ground state** solution for "any" Hamiltonian on a 1D Lattice (or quasi 1D)
- **Time Evolution**: White & Feiguin, Phys. Rev. Lett. **93** 076401 (2004)
- **Finite Temperature**: Feiguin & White Phys. Rev. B 72, 220401(R) (2005)
- **Dynamical Response functions** (except RIXS until now! see next slides)

Limitations

- **Becomes inefficient for two dimensional systems (Entanglement area law)**



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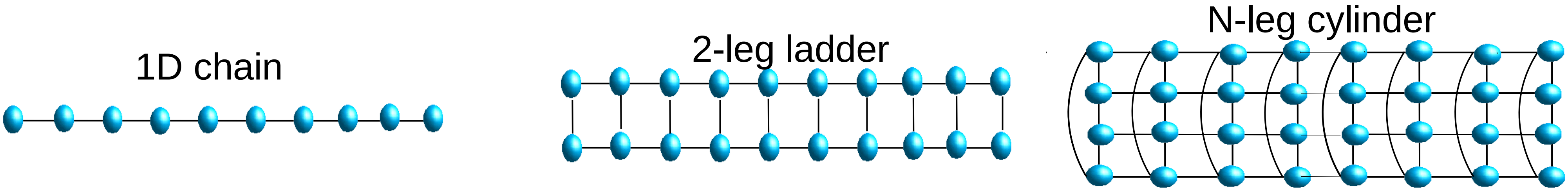

Review: Standard dynamical response functions with DMRG

EXAMPLE

Dynamical Spin Structure Factor

$$S(\mathbf{q},\omega) = \sum_{l,j=1}^{L} e^{i\mathbf{q}\cdot(\mathbf{R}_l - \mathbf{R}_j)} \langle g|\hat{S}_l^{-} \frac{1}{\omega - \hat{H} + E_g + i\eta} \hat{S}_j^{+}|g\rangle$$

Review: Standard dynamical response functions with DMRG

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Correction-vector

$$|x(\omega + i\eta)\rangle = \frac{1}{\omega - \hat{H} + E_g + i\eta}\hat{S}_j^{+}|g\rangle$$

Review: Standard dynamical response functions with DMRG

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Correction-vector

$$|x(\omega+i\eta)\rangle = \frac{1}{\omega-\hat{H}+E_g+i\eta}\hat{S}_j^+|g\rangle$$

DIFFICULT TO CALCULATE
NEED THE ENTIRE SPECTRUM OF H!

$$|x(\omega+i\eta)\rangle = \sum_n \frac{\langle n|\hat{S}_j^+|g\rangle}{\omega-E_n+E_g+i\eta}|n\rangle$$

$$1 = \sum_n |n\rangle\langle n|$$

It seems we need to go back to ED for this calculation!

Krylov "trick" to compute DMRG correction-vectors

EXAMPLE

Dynamical Spin Structure Factor

$$S(\mathbf{q},\omega) = \sum_{l,j=1}^{L} e^{i\mathbf{q}\cdot(\mathbf{R}_l - \mathbf{R}_j)} \langle g|\hat{S}_l^- \boxed{\frac{1}{\omega - \hat{H} + E_g + i\eta}\hat{S}_j^+|g\rangle}$$

Correction-vector

$$\boxed{|x(\omega + i\eta)\rangle = \frac{1}{\omega - \hat{H} + E_g + i\eta}\hat{S}_j^+|g\rangle}$$

$$\hat{V}^\dagger \hat{W}^\dagger \frac{1}{\omega + E_g - \hat{D} + i\eta} \hat{W}\hat{V}\hat{S}_j^\dagger|g\rangle$$

Current DMRG basis→ Tridiagonalization (Krylov)→ Diagonalization→ back to original basis!

$$\hat{H} \simeq \hat{V}\hat{T}\hat{V}^\dagger \qquad \hat{T} = \hat{W}\hat{D}\hat{W}^\dagger$$

For more details, see A. Nocera et al., Phys. Rev. E **94**, 053308 (2016)

How do we compute the RIXS spectra with DMRG?

$$\boxed{I_{\mathrm{RIXS}}(\mathbf{q}, \hbar\Omega) \propto \sum_f \left| \sum_{j,n} \frac{\langle f|\hat{D}_j^\dagger(\mathbf{k}_{\mathrm{out}})|n\rangle\langle n|\hat{D}_j(\mathbf{k}_{\mathrm{in}})|g\rangle}{\hbar\omega_{\mathrm{in}} - E_n^{\mathrm{ch}} + E_g + i\hbar\Gamma} \right|^2 \delta(\hbar\Omega - E_f + E_g)}$$

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$$\sum_n \frac{|n\rangle\langle n|}{\hbar\omega_{\mathrm{in}} - E_n^{\mathrm{ch}} + E_g + i\hbar\Gamma} \to \frac{1}{\hbar\omega_{\mathrm{in}} - \hat{H}^{\mathrm{ch}} + E_g + i\hbar\Gamma}$$

- **Take care of intermediate states**

$$\sum_f |f\rangle\langle f|\delta(\hbar\Omega - E_f + E_g) \to -\lim_{\eta\to 0} \mathrm{Im}\left[\frac{1}{\hbar\Omega - \hat{H} + E_g + i\eta}\right]$$

- **Take care of the final states**

A. Nocera et al., Scientific Reports **8**, 11080 (2018)

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$$\sum_n \frac{|n\rangle\langle n|}{\hbar\omega_{\mathrm{in}} - E_n^{\mathrm{ch}} + E_g + i\hbar\Gamma} \to \frac{1}{\hbar\omega_{\mathrm{in}} - \hat{H}^{\mathrm{ch}} + E_g + i\hbar\Gamma}$$

Take care of intermediate states

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Take care of the final states

$$I_{\mathrm{RIXS}}(\mathbf{q}, \hbar\Omega) \propto -\mathrm{Im}\left[\sum_{j,l=1}^{L} e^{i\mathbf{q}\cdot(\mathbf{R}_j - \mathbf{R}_l)} \langle g|\hat{\mathcal{D}}_l^\dagger \frac{1}{\hbar\omega_{\mathrm{in}} - \hat{H}^{\mathrm{ch}} + E_g - i\hbar\Gamma} \hat{\mathcal{D}}_l \frac{1}{\hbar\Omega - \hat{H} + E_g + i\eta} \hat{\mathcal{D}}_j^\dagger \frac{1}{\hbar\omega_{\mathrm{in}} - \hat{H}^{\mathrm{ch}} + E_g + i\hbar\Gamma} \hat{\mathcal{D}}_j|g\rangle\right]$$

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$$\sum_n \frac{|n\rangle\langle n|}{\hbar\omega_{\mathrm{in}} - E_n^{\mathrm{ch}} + E_g + i\hbar\Gamma} \rightarrow \frac{1}{\hbar\omega_{\mathrm{in}} - \hat{H}^{\mathrm{ch}} + E_g + i\hbar\Gamma}$$

Take care of intermediate states

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What do we do now?

A. Nocera et al., Scientific Reports **8**, 11080 (2018)

How do we compute the RIXS spectra with DMRG?

$$I_{\mathrm{RIXS}}(\mathbf{q},\hbar\Omega) \propto -\mathrm{Im}\left[\sum_{j,l=1}^{L} e^{i\mathbf{q}\cdot(\mathbf{R}_j-\mathbf{R}_l)}\langle g|\hat{\mathcal{D}}_l^\dagger \frac{1}{\hbar\omega_{\mathrm{in}}-\hat{H}^{\mathrm{ch}}+E_g-i\hbar\Gamma}\hat{\mathcal{D}}_l \frac{1}{\hbar\Omega-\hat{H}+E_g+i\eta}\hat{\mathcal{D}}_j^\dagger \frac{1}{\hbar\omega_{\mathrm{in}}-\hat{H}^{\mathrm{ch}}+E_g+i\hbar\Gamma}\hat{\mathcal{D}}_j|g\rangle\right]$$

Let's define new correction vectors $|\alpha_j(\omega_{\mathrm{in}},\Gamma)\rangle \equiv \dfrac{1}{\hbar\omega_{\mathrm{in}}-\hat{H}^{\mathrm{ch}}+E_g+i\hbar\Gamma}\hat{\mathcal{D}}_j|g\rangle$

A. Nocera et al., Scientific Reports **8**, 11080 (2018)

How do we compute the RIXS spectra with DMRG?

$$I_{\mathrm{RIXS}}(\mathbf{q},\hbar\Omega) \propto -\mathrm{Im}\left[\sum_{j,l=1}^{L} e^{i\mathbf{q}\cdot(\mathbf{R}_j-\mathbf{R}_l)}\langle g|\hat{\mathcal{D}}_l^\dagger \frac{1}{\hbar\omega_{\mathrm{in}}-\hat{H}^{\mathrm{ch}}+E_g-i\hbar\Gamma}\hat{\mathcal{D}}_l \frac{1}{\hbar\Omega-\hat{H}+E_g+i\eta}\hat{\mathcal{D}}_j^\dagger \frac{1}{\hbar\omega_{\mathrm{in}}-\hat{H}^{\mathrm{ch}}+E_g+i\hbar\Gamma}\hat{\mathcal{D}}_j|g\rangle\right]$$

Let's define new correction vectors $|\alpha_j(\omega_{\mathrm{in}},\Gamma)\rangle \equiv \frac{1}{\hbar\omega_{\mathrm{in}}-\hat{H}^{\mathrm{ch}}+E_g+i\hbar\Gamma}\hat{\mathcal{D}}_j|g\rangle$

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A. Nocera et al., Scientific Reports **8**, 11080 (2018)

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A. Nocera et al., Scientific Reports **8**, 11080 (2018)

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We know what to compute now!

(1) Compute $|\alpha_j(\omega_{\mathrm{in}},\Gamma)\rangle = \frac{1}{\hbar\omega_{\mathrm{in}} - \hat{H}^{\mathrm{ch}} + E_g + i\hbar\Gamma} \hat{\mathcal{D}}_j |g\rangle$

(2) Compute $|x_j(\Omega)\rangle = \frac{1}{\hbar\Omega - \hat{H} + E_g + i\eta} \hat{\mathcal{D}}_j^\dagger |\alpha_j(\omega_{\mathrm{in}},\Gamma)\rangle$

A. Nocera et al., Scientific Reports **8**, 11080 (2018)

How do we compute the RIXS spectra with DMRG?

$$I_{\mathrm{RIXS}}(\mathbf{q},\hbar\Omega) \propto -\mathrm{Im}\left[\sum_{j,l=1}^{L} e^{i\mathbf{q}\cdot(\mathbf{R}_j-\mathbf{R}_l)}\langle g|\hat{\mathcal{D}}_l^{\dagger}\frac{1}{\hbar\omega_{\mathrm{in}}-\hat{H}^{\mathrm{ch}}+E_g-i\hbar\Gamma}\hat{\mathcal{D}}_l\frac{1}{\hbar\Omega-\hat{H}+E_g+i\eta}\hat{\mathcal{D}}_j^{\dagger}\frac{1}{\hbar\omega_{\mathrm{in}}-\hat{H}^{\mathrm{ch}}+E_g+i\hbar\Gamma}\hat{\mathcal{D}}_j|g\rangle\right]$$

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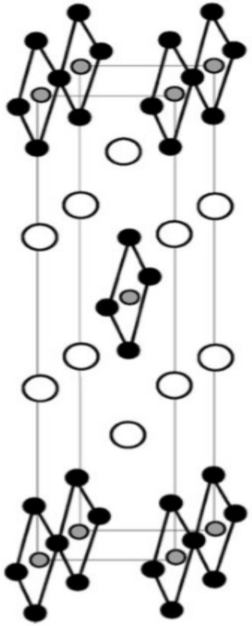
(2) Compute $|x_j(\Omega)\rangle = \frac{1}{\hbar\Omega-\hat{H}+E_g+i\eta}\hat{\mathcal{D}}_j^{\dagger}|\alpha_j(\omega_{\mathrm{in}},\Gamma)\rangle$

We can do it with the Krylov approach!

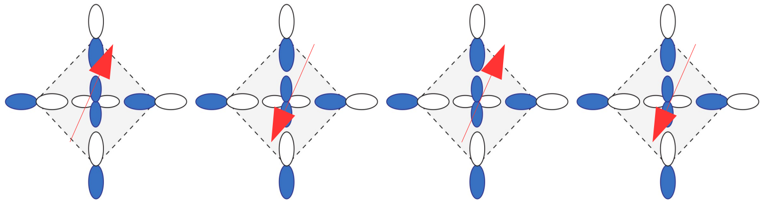
A. Nocera et al., Scientific Reports **8**, 11080 (2018)

Testing the RIXS-DMRG method: quasi-1D Cuprates

Sr_2CuO_3 (corner-shared)



- Same CuO_4 building blocks as 2D cuprates
- Large Hubbard U~8eV localizes the Cu spin (d^9 valence state)

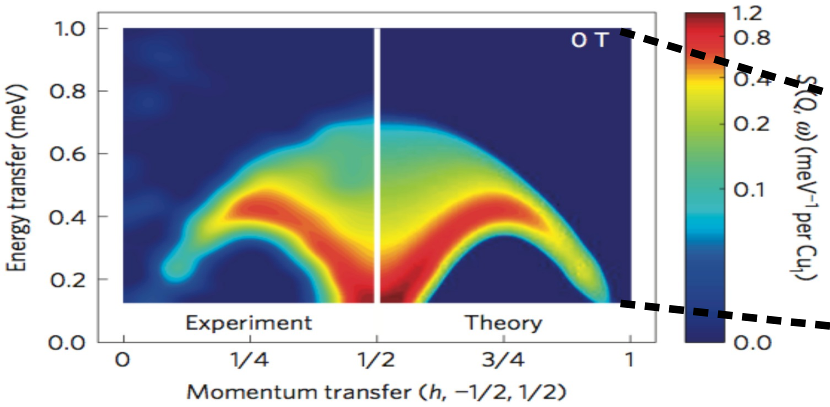


- For T>5K **interchain coupling is "zero"**

Neudert et al., PRB 62, 10752 (2000); Minzuno et al., PRB 57, 5326 (1998).

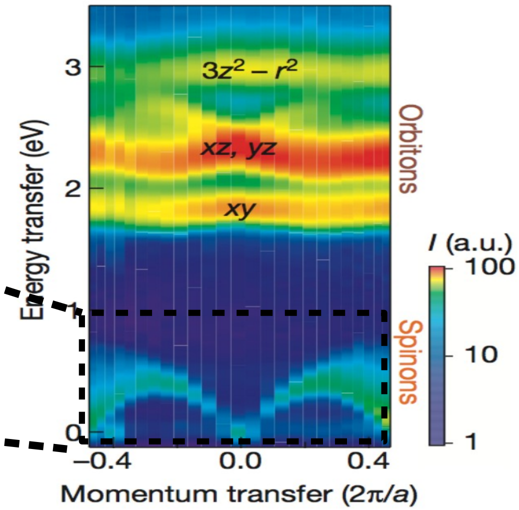
Low energy magnetic excitations in quasi-1D cuprates: Inelastic Neutron Scattering & RIXS

Magnetic excitations from INS



Mourigal et al., Nat. Phys. **9**, 435(2013)

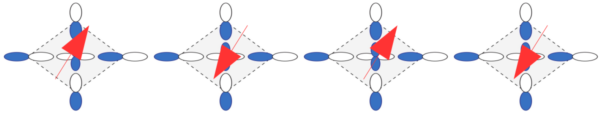
RIXS @ Cu L-edge

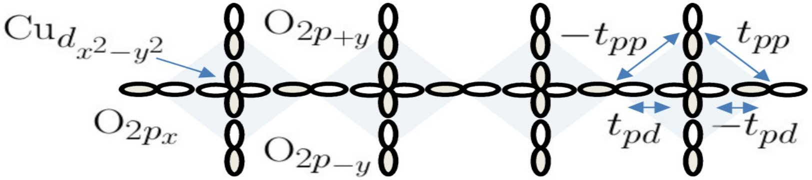


J. Schlappa et al. Nature **485**, 82 (2012)

Effective Models for 1D cuprate chains

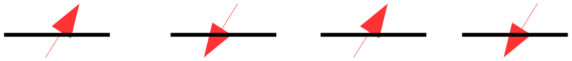
Multi-orbital *pd* model





$$H = \epsilon_d \sum_{i,\sigma} n^d_{i,\sigma} + \sum_{j,\sigma} \epsilon_{p,\gamma} n^p_{j,\gamma,\sigma} + \sum_{\substack{\langle i,j\rangle \\ \gamma,\sigma}} t^{ij}_{pd} (d^\dagger_{i,\sigma} p_{j,\gamma,\sigma} + \mathrm{h.c.})$$
$$+ \sum_{\substack{\langle j,j'\rangle \\ \gamma,\gamma',\sigma}} t^{jj'}_{pp} p^\dagger_{j,\gamma,\sigma} p_{j',\gamma'\sigma} + U_d \sum_i n^d_{i,\uparrow} n^d_{i,\downarrow}$$
$$+ U_p \sum_{i,\gamma} n^p_{j,\gamma,\uparrow} n^p_{j,\gamma,\downarrow} + U_{pd} \sum_{\substack{\langle i,j\rangle \\ \sigma,\sigma'}} n^d_{i,\sigma} n^p_{j,\gamma,\sigma'}.$$

1-orbital *t-J* model



$$H = -t \sum_{i,\sigma} (\tilde{d}^\dagger_{i,\sigma} \tilde{d}_{i+1,\sigma} + h.c.) + J \sum_i \mathbf{S}_i \cdot \mathbf{S}_{i+1}$$

Parameters for the pd-model

$\epsilon_d = 0$ $\quad \epsilon_{p,x} = 3$ $\quad \epsilon_{p,y} = 3.5$

$|t_{(p,x)d}| = 1.5$ $\quad |t_{(p,y)d}| = 1.8$ $\quad |t_{pp}| = 0.75$

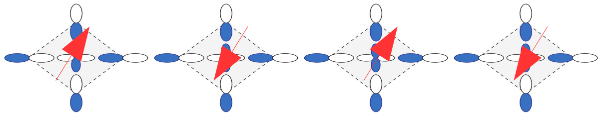
$U_d = 8$ $\quad U_{pd} = 1$ $\quad U_p = 4$

K. Wohlfeld et al., PRB 88, 195138 (2013)

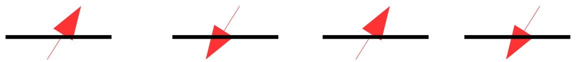
A. Nocera et al., Scientific Reports **8**, 11080 (2018)

Benchmarking our DMRG method against ED

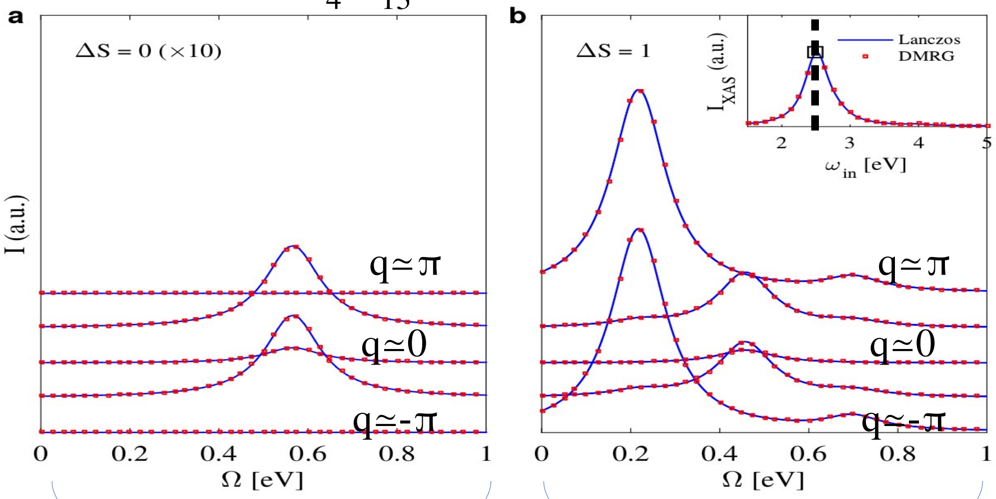
Multi-orbital *pd* model



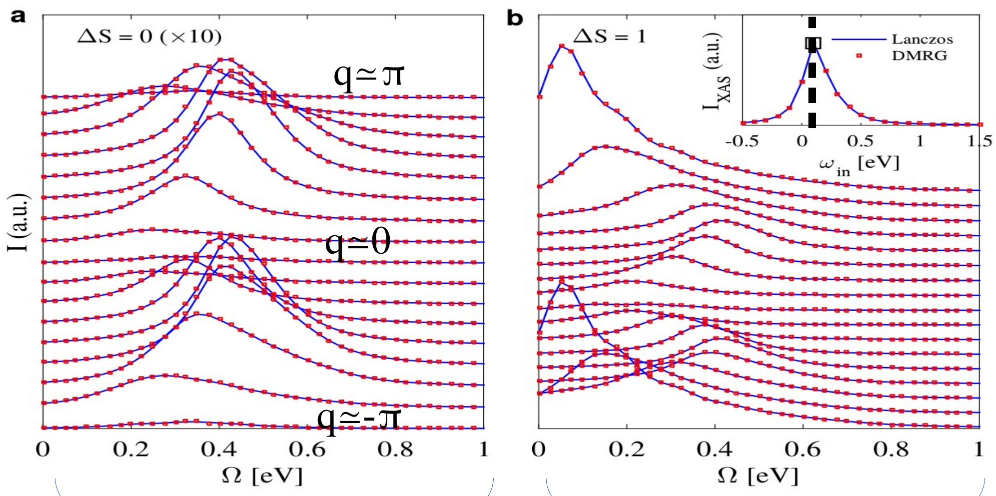
1-orbital *t-J* model



Cu_4O_{13} cluster 4 holes



L=16 sites cluster 16 holes

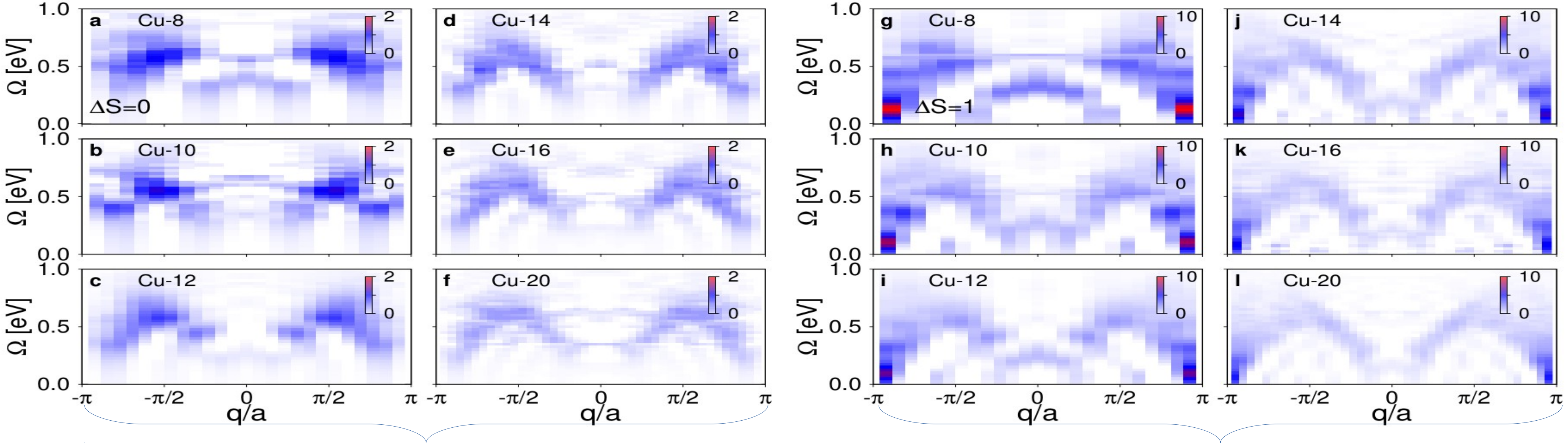


No spin-flip | Spin-flip | No spin-flip | Spin-flip

A. Nocera et al., Scientific Reports **8**, 11080 (2018)

Beyond ED: large sizes with DMRG!

Multi-orbital *pd* model



No Spin-flip

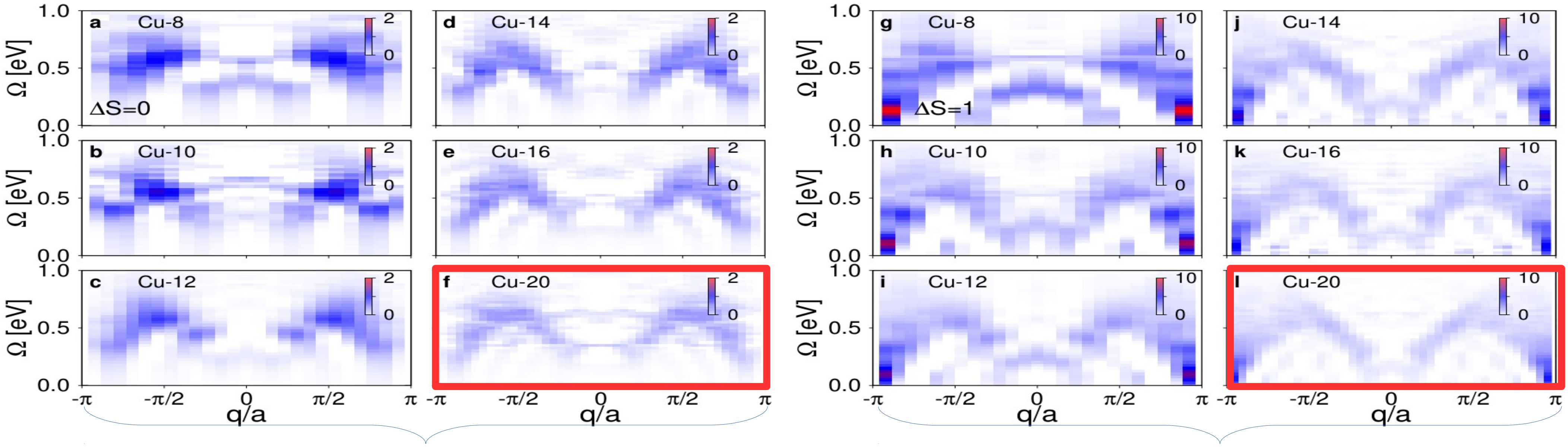
Spin-flip

A. Nocera et al., Scientific Reports **8**, 11080 (2018)

Beyond ED: large sizes with DMRG!

Multi-orbital *pd* model

DMRG can go up (and beyond) to $Cu_{20}O_{61}$ cluster (~80 orb) with 20 holes!



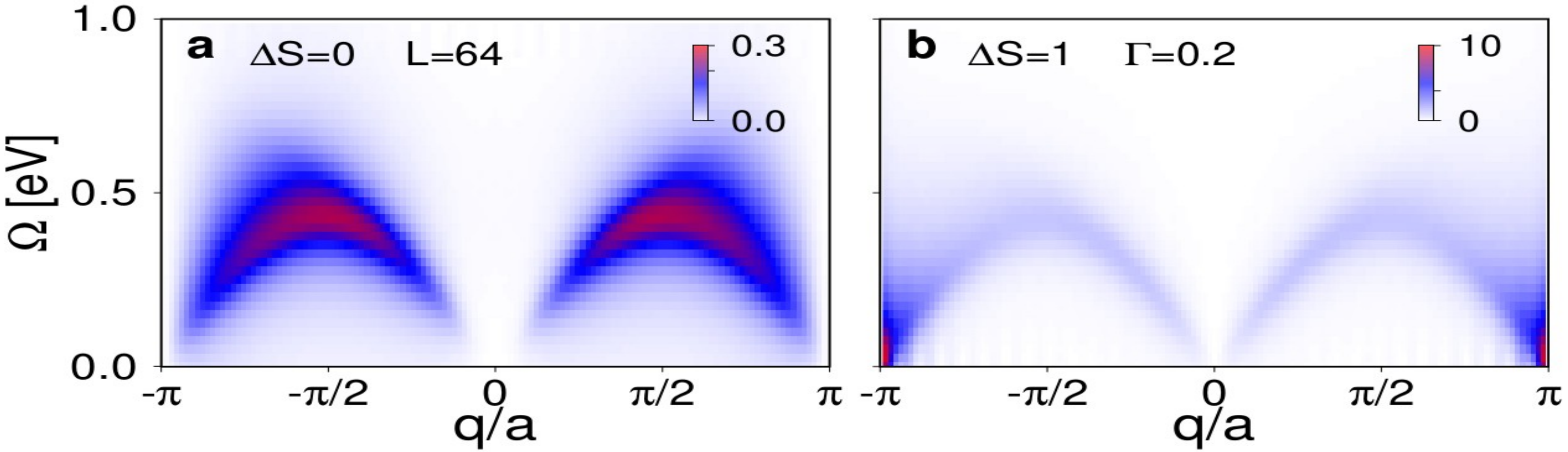
No Spin-flip

Spin-flip

A. Nocera et al., Scientific Reports **8**, 11080 (2018)

Beyond ED: large sizes with DMRG!

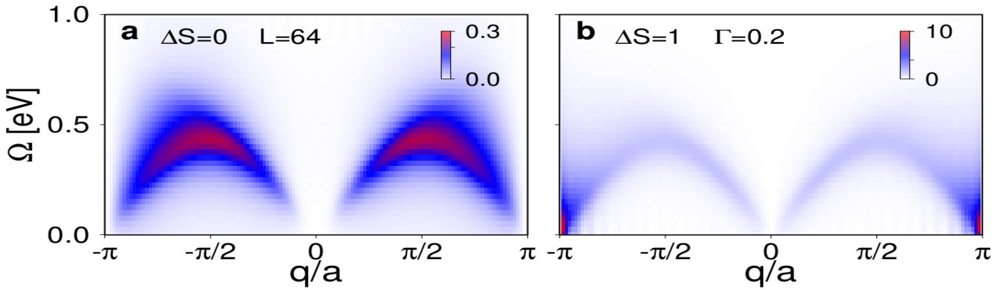
1-orbital *t-J* model with L=64 sites (ED can only do ~20 sites)



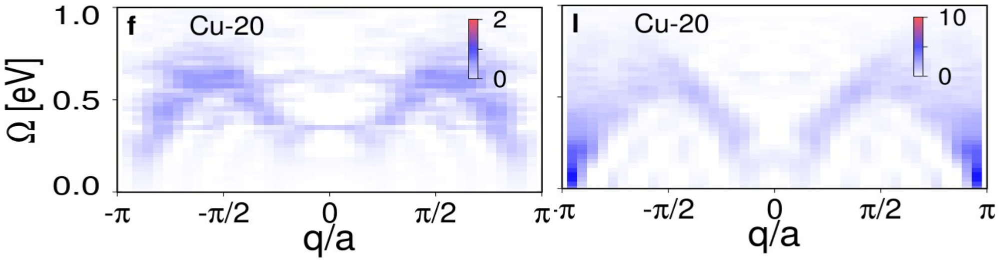
A. Nocera et al., Scientific Reports **8**, 11080 (2018)

Comparison between multi-orbital *pd* and *t-J* models

1-orbital *t-J* model

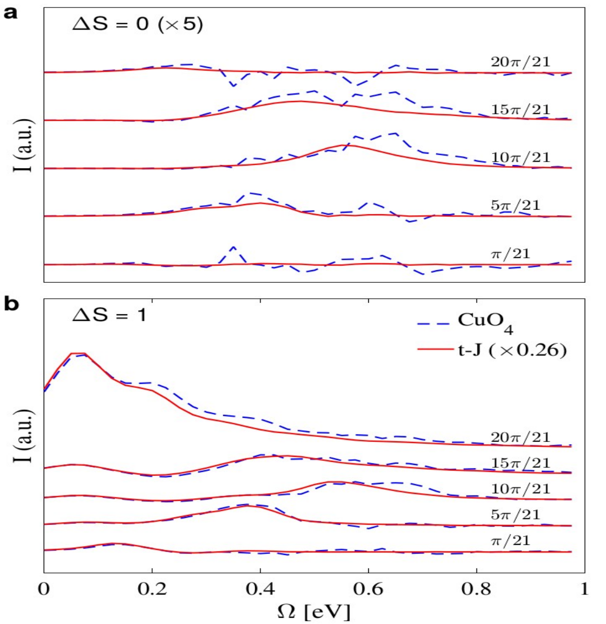


Multi-orbital *pd* model



$t = 0.5$ eV, $J = 0.325$ eV, $V_C = 4$ eV, $\Gamma = 0.2$ eV,

L=20 for both models



Scaling factor=0.26

Covalency effects are important!

A. Nocera et al., Scientific Reports **8**, 11080 (2018)

Summary & Current work

- Proposed a DMRG based algorithm for computing full RIXS spectra for strongly correlated materials

- Improvement of the RIXS-DMRG algorithm

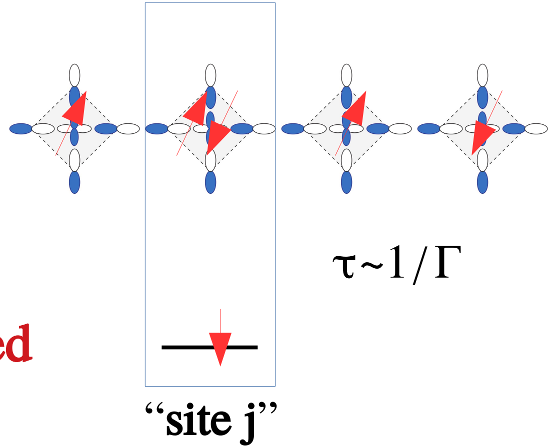
- Application of the method to different RIXS “edges”, geometries, and Hamiltonians

- Future: Non-equilibrium RIXS-DMRG?

Thank you for your attention!

How do we treat the core-hole in the intermediate state?

We consider the core-holes (or electrons) completely localized!

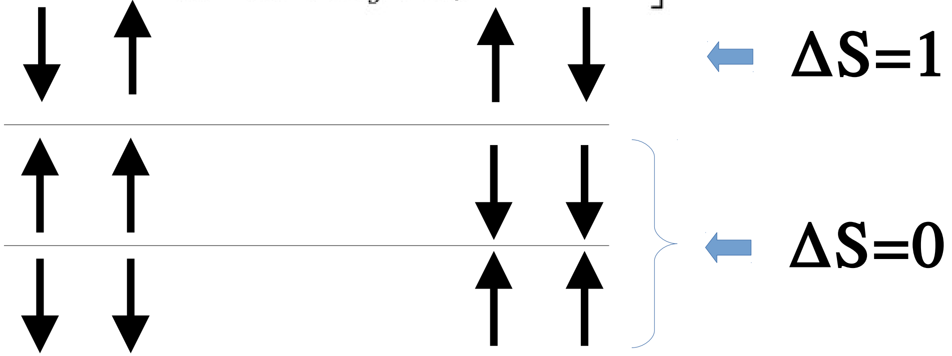


$$\hat{H}_{\mathrm{ch}} = \hat{H} + \hat{H}^C \qquad H^C = V_C \sum_{j,\sigma,\sigma'} \hat{n}^d_{j,\sigma}(1 - \hat{n}^{p_\alpha}_{j,\sigma'})$$

Also, we flip the core-hole spin "by hand" so Total Spin is conserved

NOTE: We can compute spin-conserving and non-spin cons. channels!

$$I_{\mathrm{RIXS}}(\mathbf{q},\Omega) \propto -\mathrm{Im}\left[\sum_{i,j=1}^{L} \sum_{\substack{\gamma,\gamma' \\ \sigma,\sigma'}} e^{i\mathbf{q}\cdot(\mathbf{R}_i - \mathbf{R}_j)} \langle \alpha_{i,\gamma}|\hat{D}_{i,\gamma'} \frac{1}{\Omega - \hat{H} + E_g + i\eta} \hat{D}^\dagger_{j,\sigma'}|\alpha_{j,\sigma}\rangle\right]$$

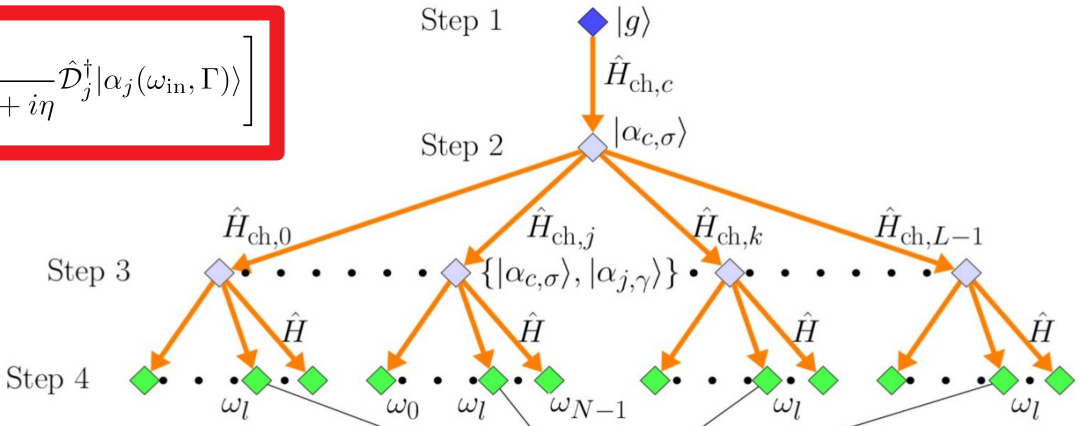


Three separate calculations

A. Nocera et al., Scientific Reports **8**, 11080 (2018)

Parallel Algorithm for computing RIXS spectra with DMRG

$$I_{\mathrm{RIXS}}(\mathbf{q},\hbar\Omega) \propto -\mathrm{Im}\left[\sum_{j,l=1}^{L} e^{i\mathbf{q}\cdot(\mathbf{R}_j-\mathbf{R}_l)}\langle\alpha_l(\omega_{\mathrm{in}},\Gamma)|\hat{\mathcal{D}}_l\frac{1}{\hbar\Omega-\hat{H}+E_g+i\eta}\hat{\mathcal{D}}_j^{\dagger}|\alpha_j(\omega_{\mathrm{in}},\Gamma)\rangle\right]$$



$$I(\mathbf{q},\omega_l) \propto -\mathrm{Im}[\sum_{\substack{j,\gamma,\gamma',\\ \sigma,\sigma'}} e^{i\mathbf{q}\cdot(\mathbf{R}_j-\mathbf{R}_c)}\langle\alpha_{j,\gamma}|\mathcal{D}_{j,\gamma'}|x_{c,\sigma',\sigma}\rangle] \quad ; \quad |x_{c,\sigma',\sigma}\rangle = \frac{1}{\omega_l-\hat{H}+E_g+i\eta}\mathcal{D}^{\dagger}_{c,\sigma'}|\alpha_{c,\sigma}\rangle$$

Computational Cost ~ (number of sites) × (number of “omegas”) simulations!

HOWEVER, these can be performed independently in parallel!
NEED SUPERCOMPUTERS IN CHALLENGING PROBLEMS

NOTE: IT CAN BE APPLIED FOR ANY PROBLEM WHERE DMRG CAN BE USED

A. Nocera et al., Scientific Reports **8**, 11080 (2018)

Krylov method for computing DMRG correction-vectors

$$\boxed{|x(\omega+i\eta)\rangle = \frac{1}{\omega - \hat{H} + E_g + i\eta}\hat{O}|g\rangle}$$

(1) Project $\hat{H}$ onto the "**Krylov**" space generated by $|v\rangle = \hat{O}|g\rangle$

$$|\alpha_{n+1}\rangle = \hat{H}|\alpha_n\rangle - a_n|\alpha_n\rangle - b_n^2|\alpha_{n-1}\rangle \longrightarrow \langle\alpha_n|\hat{H}|\alpha_m\rangle = \begin{pmatrix} a_1 & b_2 & & & & 0 \\ b_2 & a_2 & b_3 & & & \\ & b_3 & a_3 & \ddots & & \\ & & \ddots & \ddots & b_{k-1} & \\ & & & b_{k-1} & a_{k-1} & b_k \\ 0 & & & & b_k & a_k \end{pmatrix}$$

k<<Hilbert space size!

(2) Diagonalize into the "**Krylov**" space

$$\longrightarrow \langle\tilde{n}|\hat{H}|\tilde{m}\rangle = \begin{pmatrix} \epsilon_1 & & & & & 0 \\ & \epsilon_2 & & & & \\ & & \epsilon_3 & & & \\ & & & \ddots & & \\ & & & & \epsilon_{k-1} & \\ 0 & & & & & \epsilon_k \end{pmatrix}$$

Stop the procedure for some $\tilde{n}_{\max} < k$ until convergence on $\{\epsilon_{\tilde{n}}\}$ for $\tilde{n} = 1, \tilde{n}_{\max}$

(3) Compute the correction vector as $|x(\omega+i\eta)\rangle \simeq \sum_{\tilde{n}=1}^{\tilde{n}_{\max}} \frac{\langle\tilde{n}|\hat{S}_j^+|g\rangle}{\omega - \epsilon_{\tilde{n}} + E_g + i\eta}|\tilde{n}\rangle$