OAK RIDGE National Laboratory

Quantum States of Matter in Strongly Correlated Electron Systems: Insight Through Advanced Computing

Thomas A. Maier

Oak Ridge National Laboratory

SciDAC PI Meeting, July 2019

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SciDAC Scientific Discovery through Advanced Computing

OLCF Oak Ridge Leadership Computing Facility

U.S. DEPARTMENT OF ENERGY INCITE LEADERSHIP COMPUTING

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Outline

1. CompFUSE — Introduction
2. Some highlights
3. Bond-stretching phonons in $Ba_{1-x}K_xBiO_3$
4. Two pairing domes in cuprates
5. Acknowledgments

CompFUSE Team

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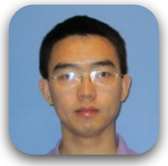
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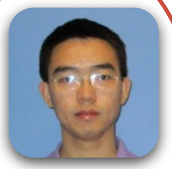
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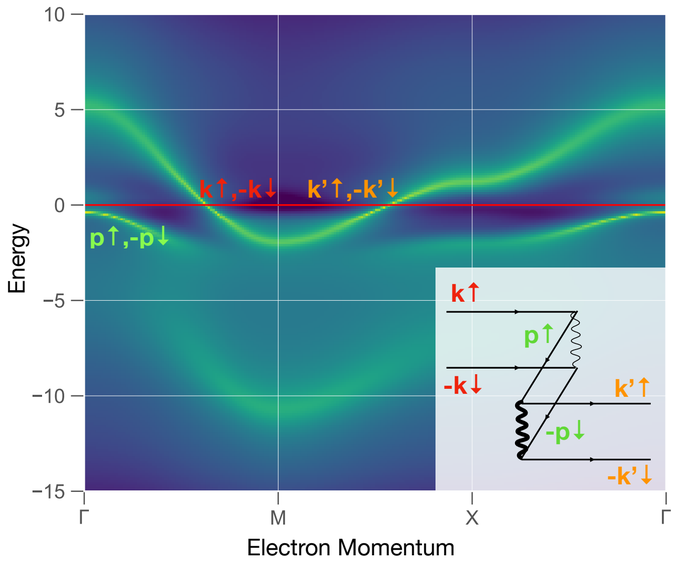
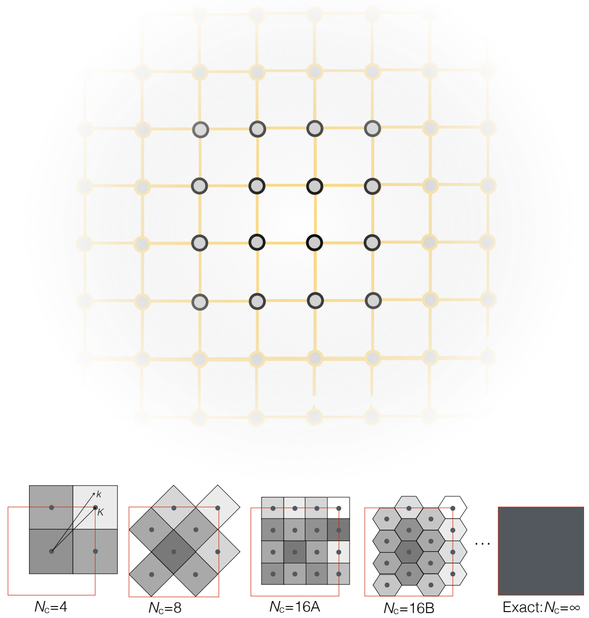
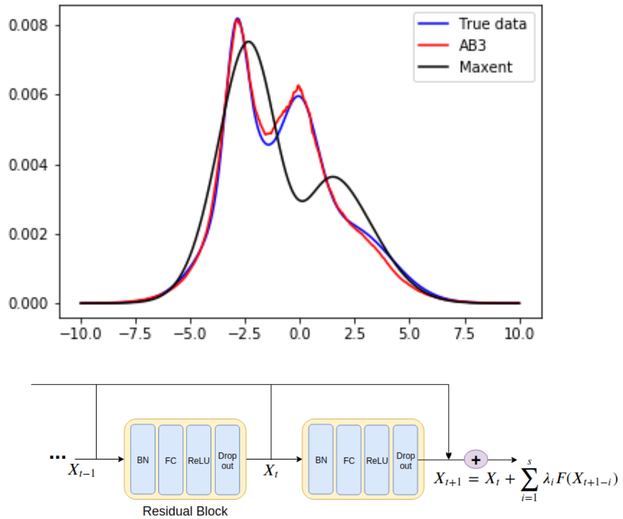
CompFUSE: Computational framework for unbiased studies of correlated electron systems



Advanced simulations of correlated quantum materials, including **unconventional superconductors** and **quantum spin liquids**.



Development of **accelerated algorithms based on DQMC, DCA and DMRG** and **efficient implementations** on leadership class supercomputers.



Development of **new approaches for analytic continuation** of QMC results to extract real time dynamics from imaginary time data.

Focus: Dynamics, excited states, finite temperatures, mechanisms

Dynamical and thermal magnetic properties of α-$RuCl_3$

Failure of available spin models for α-$RuCl_3$

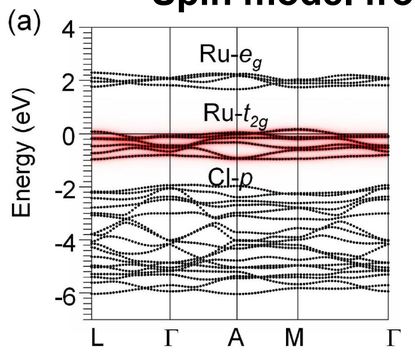
- ***Derived a spin model for $RuCl_3$*** and studied several available models from the literature
- Using exact diagonalization based methods, calculated the ***specific heat*** and the ***inelastic neutron scattering response***
- All ***existing models fail to completely explain the experiments***

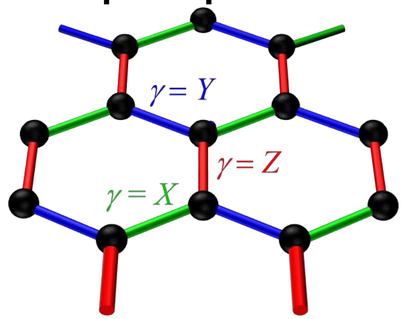
Research Details

- DFT calculation of $RuCl_3$, Wannier transformation to Ru-t_{2g} states
- Interaction parameters from constrained Random Phase Approximation
- Spin-only model from perturbation theory

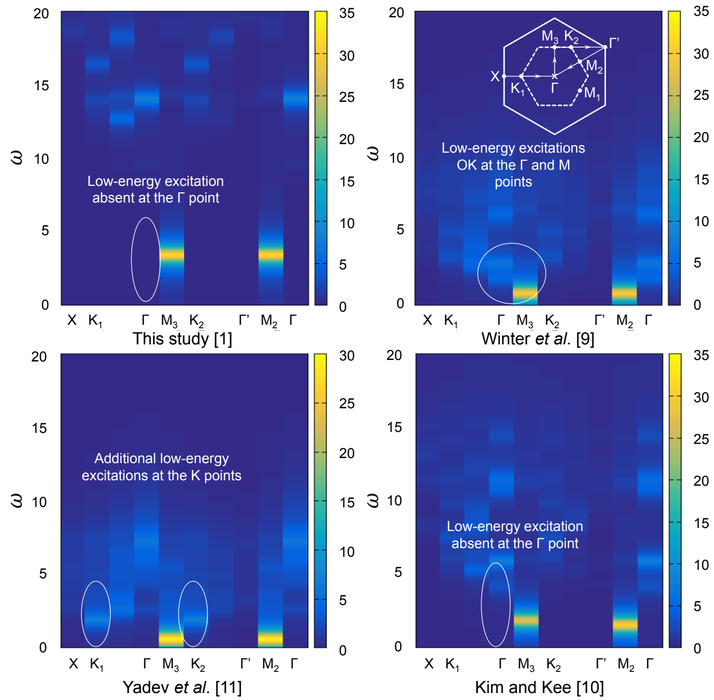
For details see poster by Satoshi Okamoto

Spin model from first principles

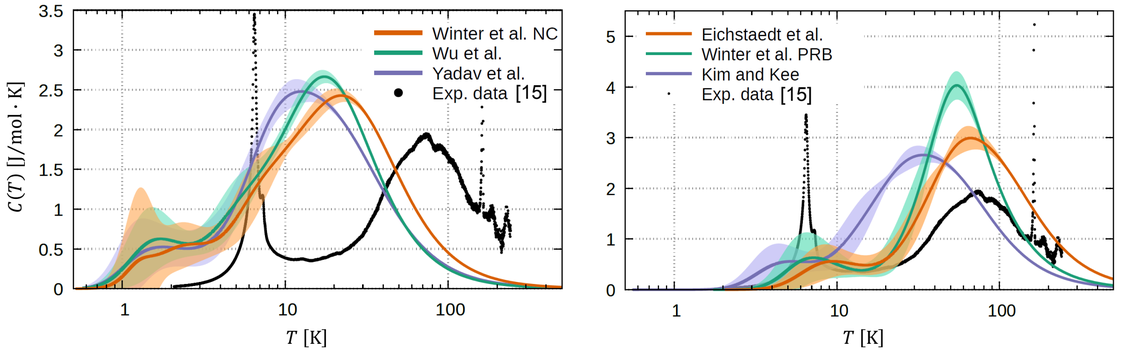




Inelastic neutron scattering



Magnetic specific heat



Eichstaedt et al., arXiv:1904.01523
Laurell & Okamoto, arXiv:1906.07579

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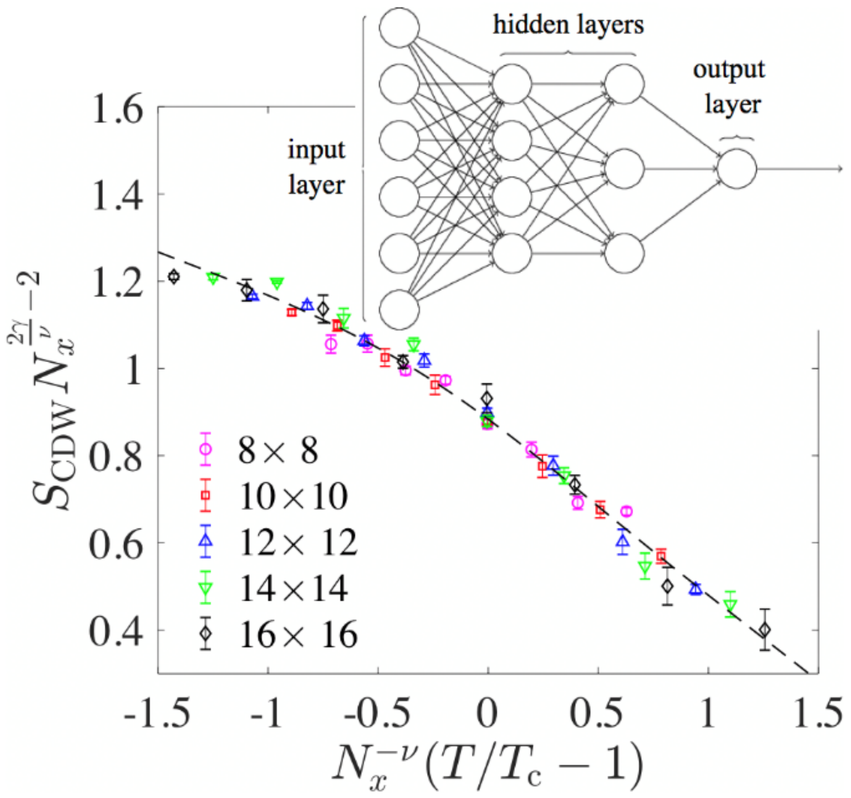
AI to accelerate determinantal lattice quantum Monte Carlo simulations



Thermodynamic scaling analysis of the ***charge density wave structure factor*** of the half-filled two-dimensional Holstein model in proximity to the metal-to-CDW insulator transition T_c

Neural networks to speed up DQMC

- ***Artificial neural networks (ANNs)*** can ***predict*** with near-perfect accuracy ***moves in quantum Monte Carlo (QMC)*** simulations of many-body Hamiltonians and obtain an order of magnitude reduction in the run time.
- Demonstration that machines can learn to perform efficient QMC simulations—without an underlying physics model and given only limited information about the configuration space—means the ***method can be easily generalized even to other challenging models, such as the Fermi-Hubbard model***.

Research Details

- ANNs were designed to predict the acceptance probabilities of local and global moves in determinantal QMC simulations of the Holstein model.
- This development in artificial intelligence (AI) granted access to large systems at low-temperatures, overcame long autocorrelation times for the model, and facilitated a thermodynamic scaling analysis.

For details see poster by Steve Johnston

Li et al., arXiv:1905.10430

Analytic continuation of noisy data using multistep neural network

New data-driven framework

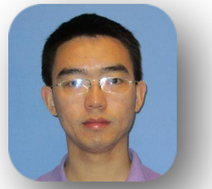
$$G(\tau) = \int K(\tau,\omega)A(\omega)d\omega\,;\;\; K(\tau,\omega) = \frac{e^{-\omega\tau}}{1+e^{-\beta\omega}}$$

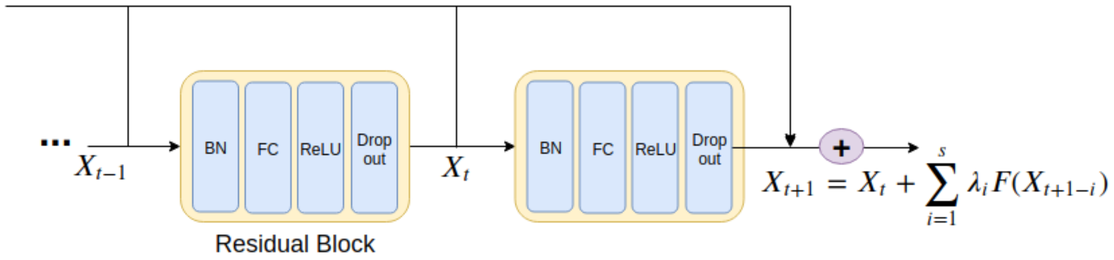
- Use ***novel linear multi-step residual neural network (ResNet)*** to learn the inverse of the kernel
- Generate training data set as a sum of R uncorrelated Gaussians
- 100k training data set, 1000 samples for validation and testing

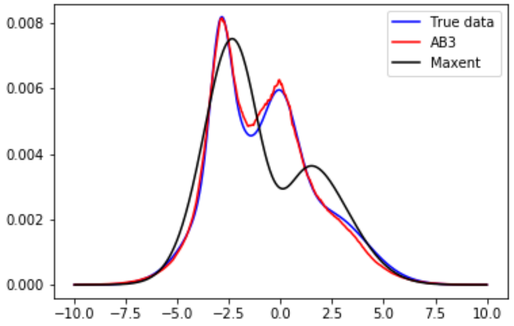
Compared to Maximum Entropy

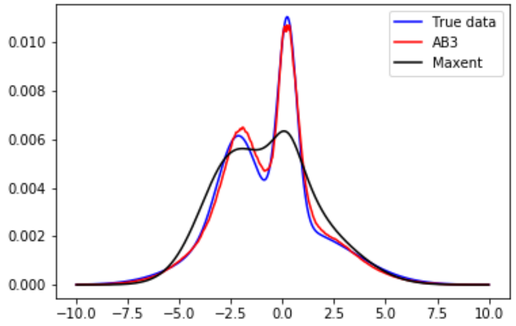
- 2-step ResNet gives similar results for high-quality data, and much better results at high noise levels.

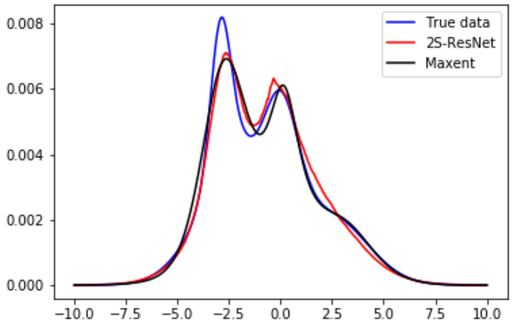
For details see poster by Xuping Xie

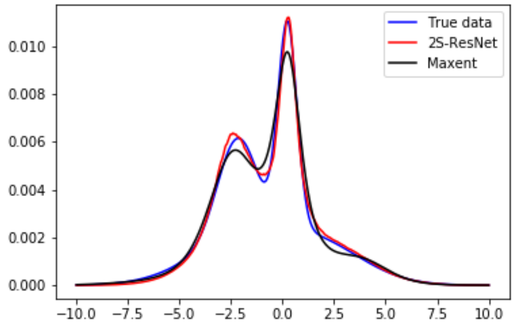












Xie et al., arXiv:1905.10430

Accelerating DCA++ on Summit

DCA++ code

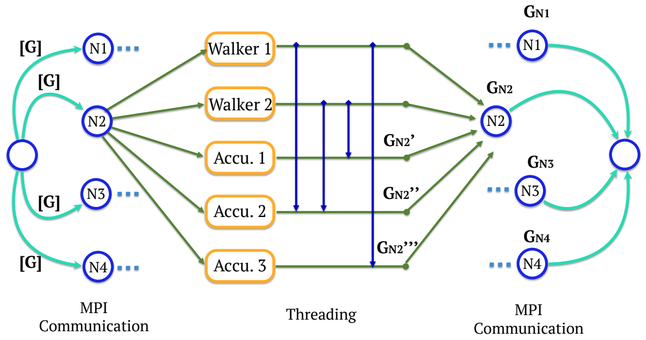
- Two kernels: (1) Coarse-graining of momentum space to map lattice problem onto effective cluster problem, (2) ***quantum Monte Carlo cluster (QMC) solver***
- ***Two-level parallelization (MPI internode + std::threads intranode)***

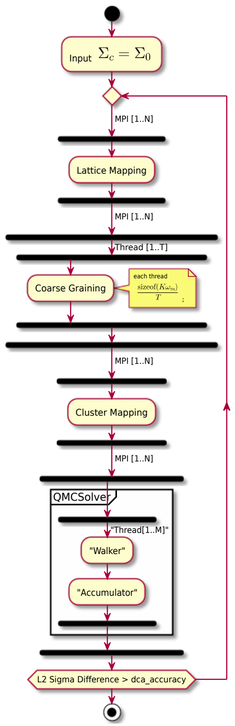
Several optimizations

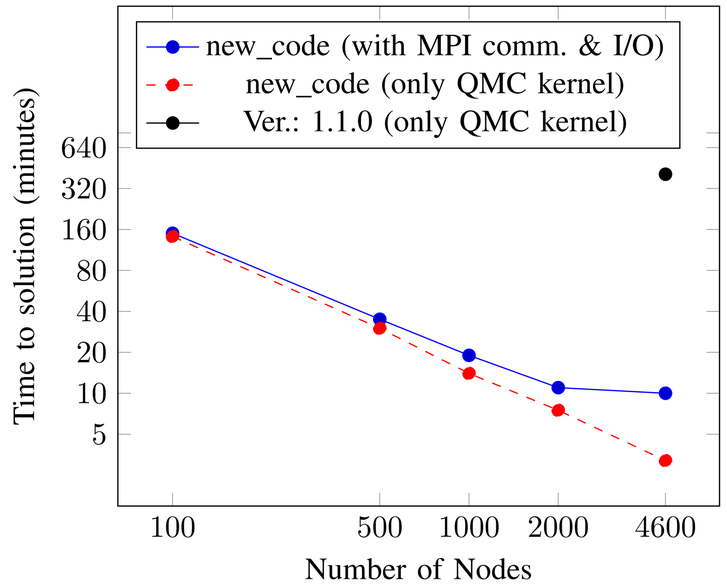
- ***Extensive profiling*** (in collaboration with RAPIDS institute)
- Coarse-graining: Improved multi-threading and intra-process scheduling
- **QMC solver:** (1) Improved asynchronous CPU-GPU communication
  (2) ***GPU support for measurement accumulation of observables***
  (3) Improved (direct) walker - accumulator communication
  (4) Dynamic intra-node workload distribution
  (5) Overlapping computation and communication
  (6) ***Mixed precision*** – Double precision (walker), single precision (two-particle vertex accumulation)

Speedup & Performance on Summit

- ~ 100 x speedup
- ~ 75 PFlops







Chatterjee et al., submitted to PACT '19

Accelerating DMRG++ for dynamics and finite temperatures

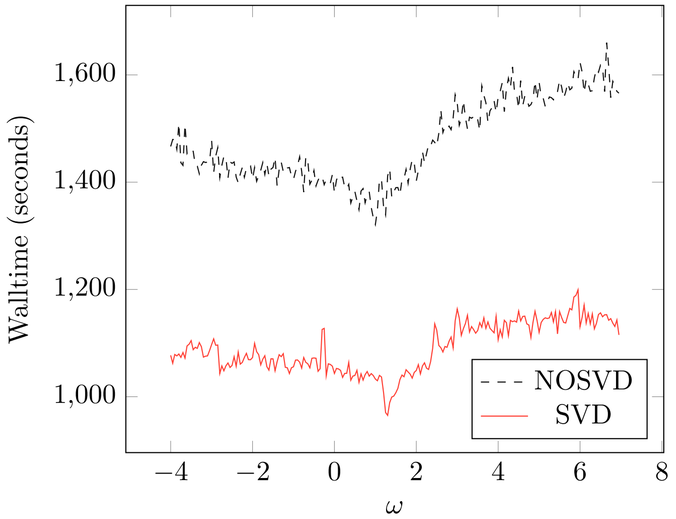
Targeting multiple states in DMRG

- ***Multiple states*** are needed in density matrix matrix for ***finite temperature calculations and dynamics***
- Replacing the density matrix by a ***singular value decomposition (SVD) increases performance*** significantly and enables multiple targeting

Optimized GPU support for Summit

- Use of ***multiple GPUs*** for larger problems (more states)
- Use of ***atomic updates in MAGMA*** library to take advantage of symmetry of Hamiltonian to reduce HBM storage on GPU
- Calculation of spectral functions is embarrassingly parallel and thus can take full advantage of parallelization on Summit, significantly increasing frequency resolution

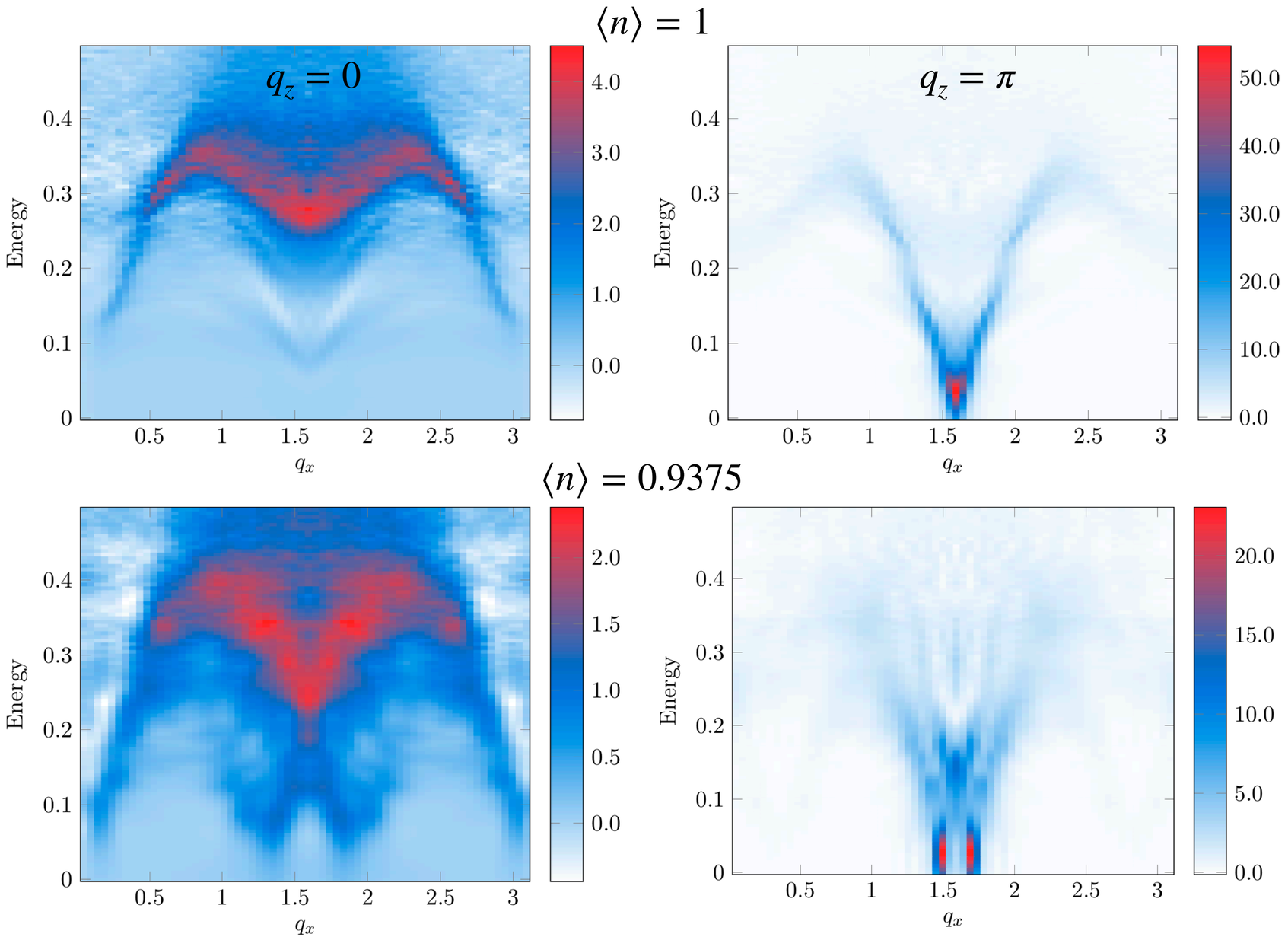
D'Azevedo et al., submitted to Phys. Rev. E (2018)



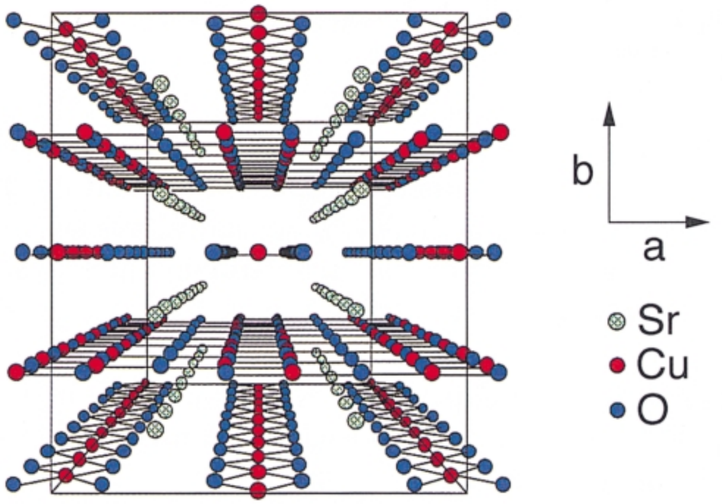
	GPU		CPU	
ω	fullRunSVD	fullRunDM	fullRunSVD	fullRunDM
GS	191	231	193	241
−2	1073	1406	1690	1969
0	1043	1380	1710	1947
2	1048	1402	1757	2014

Run-time in seconds on a Volta GPU on x86 Linux workstation. Note all SVD performed on CPU.

DMRG: S(Q,ω) for telephone ladder cuprates



$Sr_{14}Cu_{24}O_{41}$



From Matsuda et al., Riken Review '99

Parameters:
2-leg Hubbard ladder; 64 x 2;
intermediate to strong coupling

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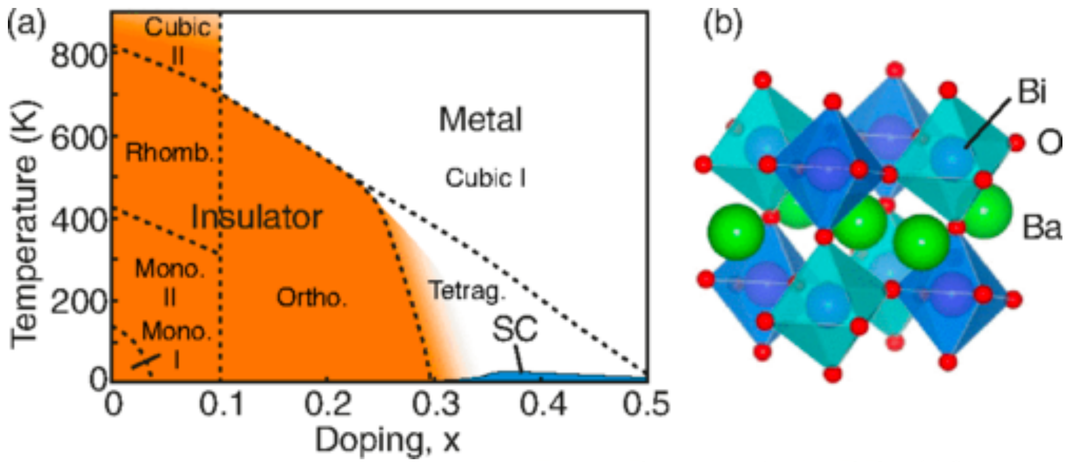
1. CompFUSE — Introduction
2. Some highlights
3. **Bond-stretching phonons in $Ba_{1-x}K_xBiO_3$**
4. Two pairing domes in cuprates
5. Acknowledgments

DQMC study of bond phonons in $Ba_{1-x}K_xBiO_3$

Li et al., arXiv:1901.07612

$Ba_{1-x}K_xBiO_3$

- Negative charge transfer regime
- Holes self-dope from cation to ligand oxygen atoms
- Hybridization between cation and oxygen atom leads to sizable electron-phonon coupling
- ***Charge-ordered insulator*** for x=0
- ***Superconductor*** (T_c ~ 30 K) at finite x



From Plumb et al., PRL '16

3-orbital Su-Schrieffer-Heger (SSH) model

$$H_0 = -t_{sp}\sum_{\langle \mathbf{r},\delta\rangle,\sigma}(P_\delta s^\dagger_{\mathbf{r},\sigma}p_{\mathbf{r},\delta,\sigma} + \mathrm{h.c.}) + t_{pp}\sum_{\langle \mathbf{r},\delta,\delta'\rangle,\sigma} P_{\delta,\delta'}p^\dagger_{\mathbf{r},\delta,\sigma}p_{\mathbf{r},\delta',\sigma} + \sum_{\mathbf{r},\sigma}\left[(\epsilon_s-\mu)\hat{n}^s_{\mathbf{r}\sigma} + (\epsilon_p-\mu)(\hat{n}^{p_x}_{\mathbf{r},\sigma}+\hat{n}^{p_y}_{\mathbf{r},\sigma})\right]$$

$$H_{\mathrm{lat}} = \sum_{\mathbf{r}}\left(\frac{\hat{P}^2_{\mathbf{r},x}}{2M} + K\hat{X}^2_{\mathbf{r},x} + \frac{\hat{P}^2_{\mathbf{r},y}}{2M} + K\hat{X}^2_{\mathbf{r},y}\right)$$

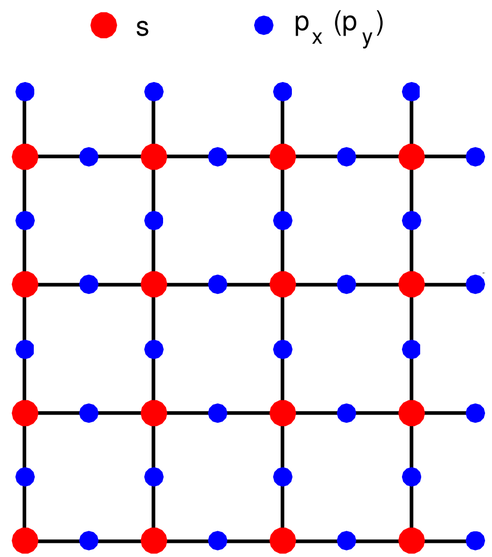
$$H_{\mathrm{e-ph}} = \alpha t_{sp}\sum_{\langle \mathbf{r},\delta\rangle,\sigma}\left(\hat{u}_{\mathbf{r},\delta}s^\dagger_{\mathbf{r},\sigma}p_{\mathbf{r},\delta,\sigma} + \mathrm{h.c.}\right)$$

Phonon frequency $\Omega = \sqrt{2}t_{sp}$

e-ph coupling strength $\alpha = 4a^{-1}$

Sign-problem free DQMC algorithm!

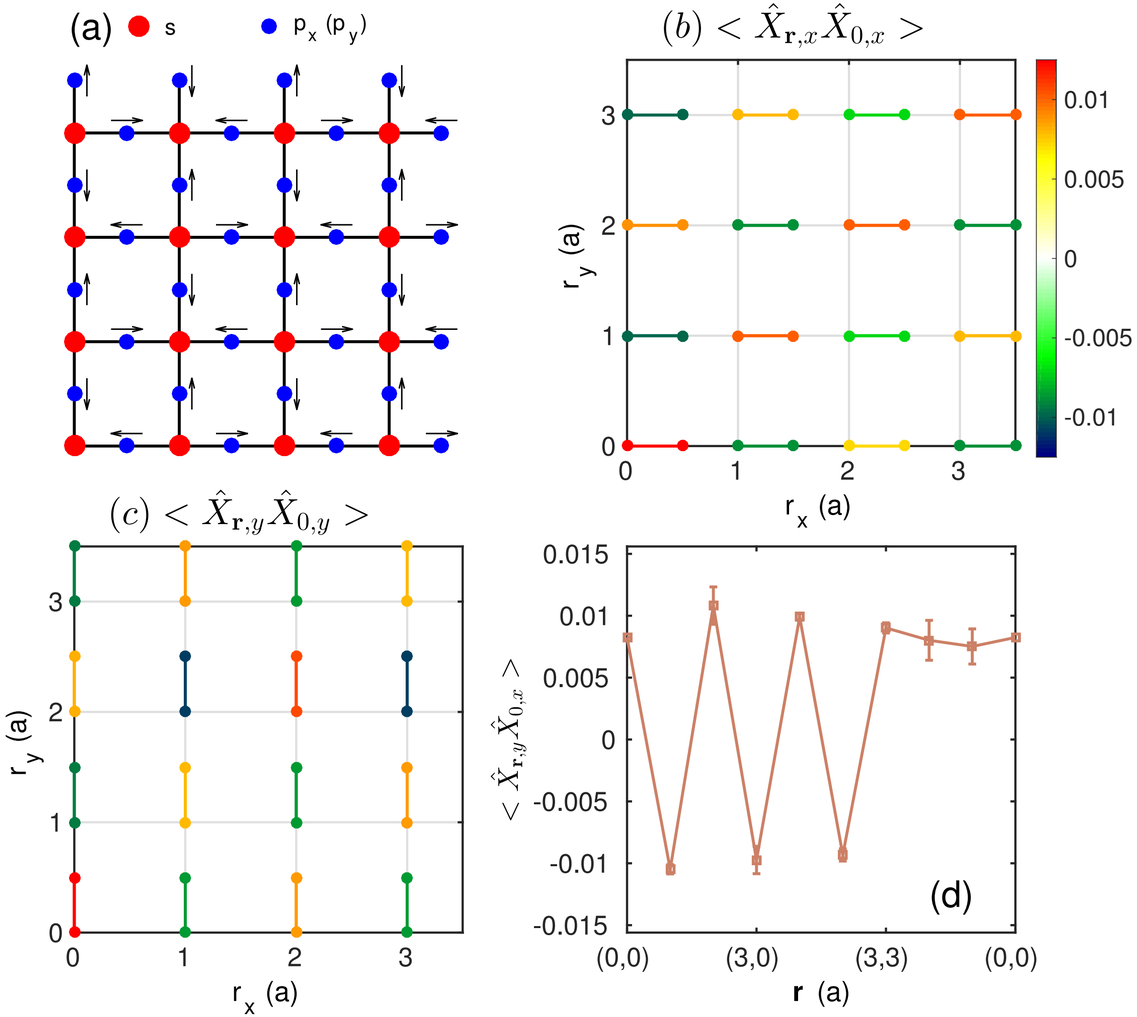
First non-perturbative treatment in > 1D!



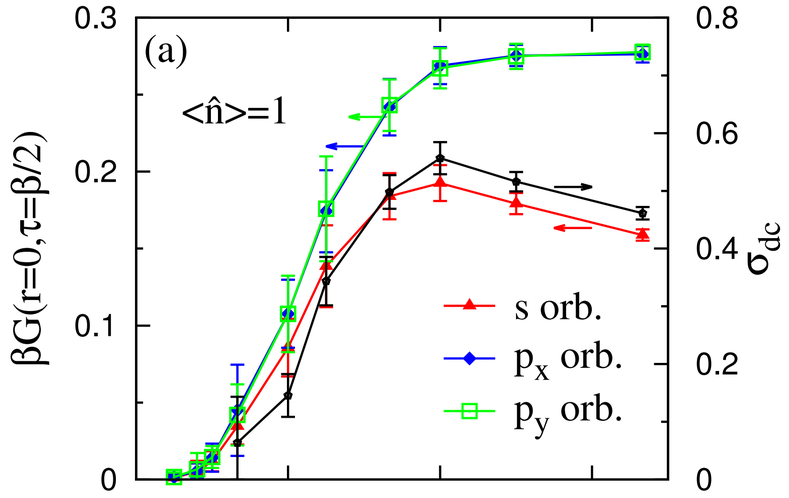
2D Lieb lattice

Bond disproportionated, charge ordered state at half-filling

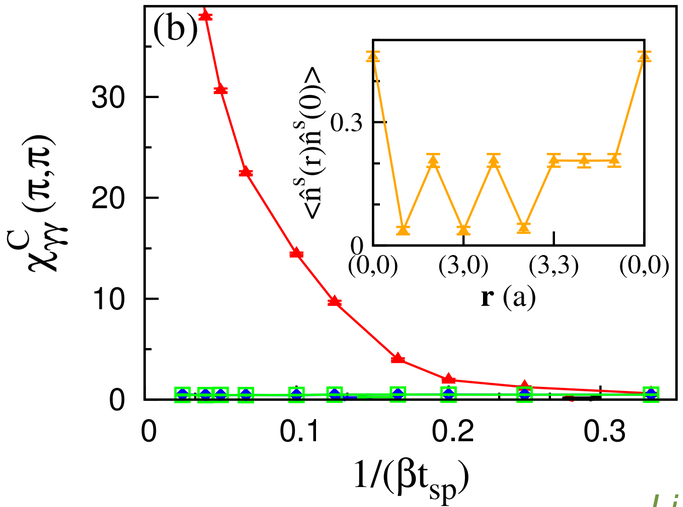
Displacement correlation functions



dc conductivity and spectral weight at E_F



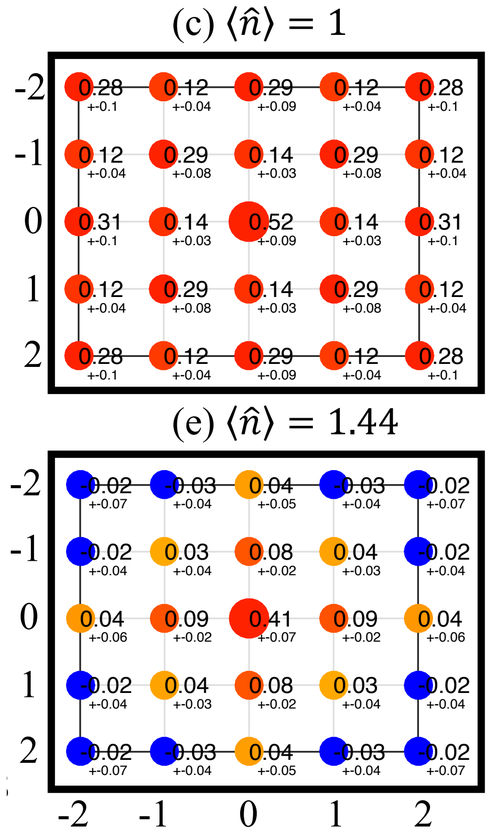
CDW susceptibility



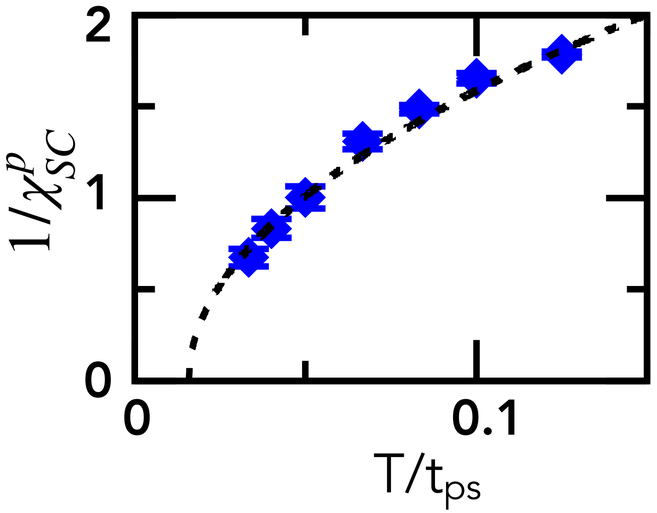
Li et al., arXiv:1901.07612

Polaronic liquid and superconductivity at finite doping

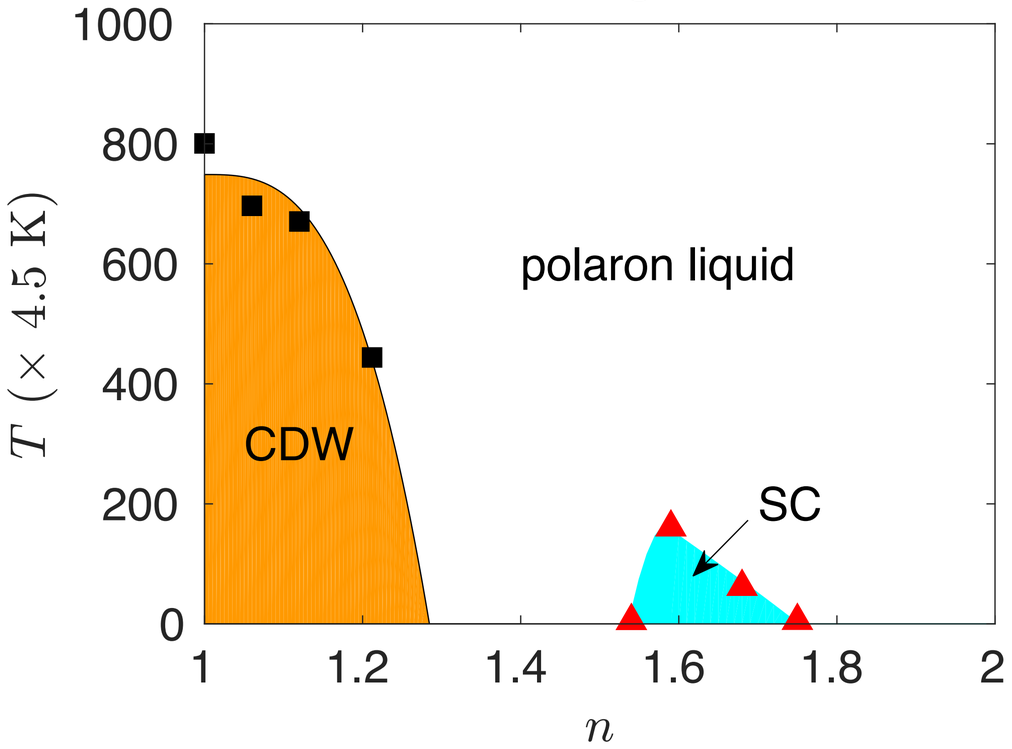
Staggered polaron correlations



S-wave pairfield susceptibility



Phase diagram



- Bipolaronic state at half-filling that melts into a polaron-liquid-like metallic state with a superconducting ground state upon doping.

Li et al., arXiv:1901.07612

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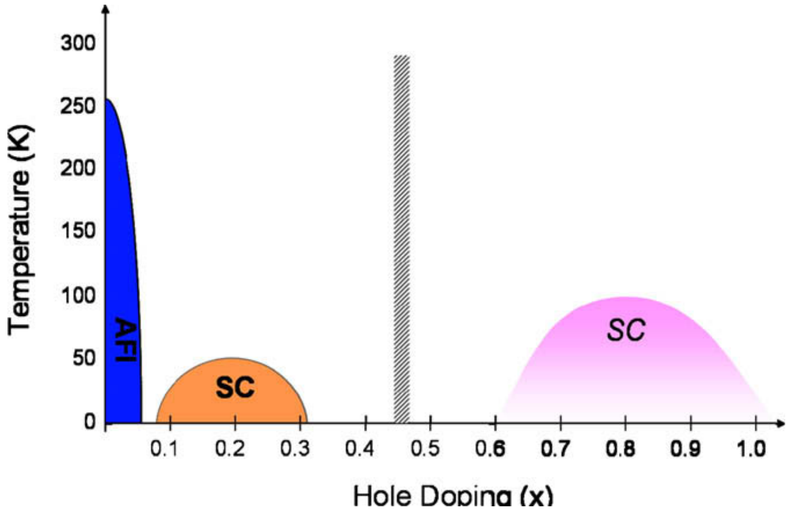
Two pairing domes as Cu^{2+} varies to Cu^{3+}

Highly (hole) overdoped curates

- $Sr_2CuO_{4-\delta}$ (T_c ~ 95 K; isostructural to 214 La_2CuO_4)
- $Cu_{0.75}Mo_{0.25}Sr_2YCu_2O_{7.54}$ (T_c ~ 84 K)
- $Ba_2CuO_{4-\delta}$ (T_c ~ 70 K)
- Monolayer CuO_2 films (T_c ~ 100 K)
- High pressure oxidized synthesis
- Reduced Cu - apical O spacing
- $d_{3z^2-r^2}$ orbital important, in addition to $d_{x^2-y^2}$ orbital

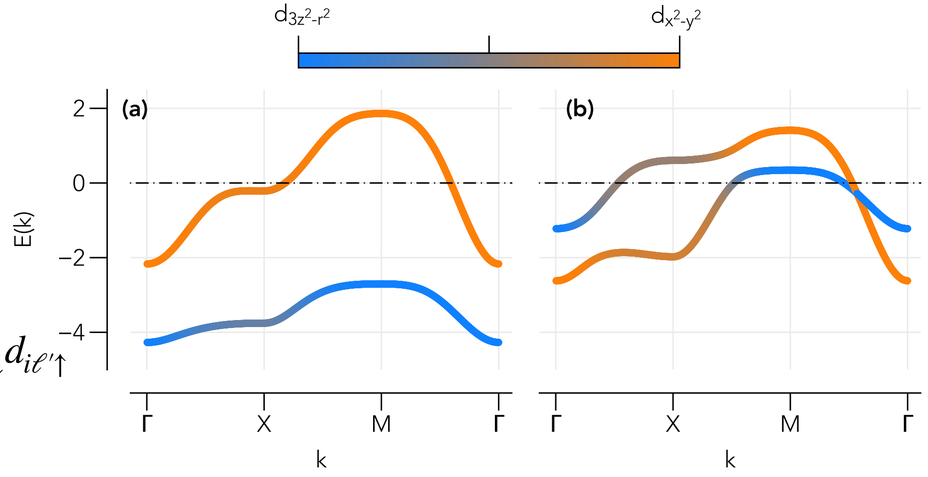


From Geballe & Marezio, Physica C '09

Two-orbital tight-binding Hubbard-Hund model

$$H_0(x) = \sum_{k\sigma} \sum_{\ell\ell'} \left(\xi_{\ell\ell'}(k) + \left(\varepsilon_\ell(x) - \mu \right) \delta_{\ell\ell'} \right) d^\dagger_{\ell\sigma}(k) d_{\ell'\sigma}(k)$$

$$H_1 = U \sum_{i,\ell} n_{i\ell\uparrow} n_{i\ell\downarrow} + U' \sum_{i,\ell'<\ell} n_{i\ell} n_{i\ell'} + J \sum_{i,\ell'<\ell\sigma,\sigma'} \sum d^\dagger_{i\ell\sigma} d^\dagger_{i\ell'\sigma'} d_{i\ell\sigma'} d_{i\ell'\sigma} + J' \sum_{i,\ell'\neq\ell} d^\dagger_{i\ell} d^\dagger_{i\ell\downarrow} d_{i\ell'\downarrow} d_{i\ell'\uparrow}$$

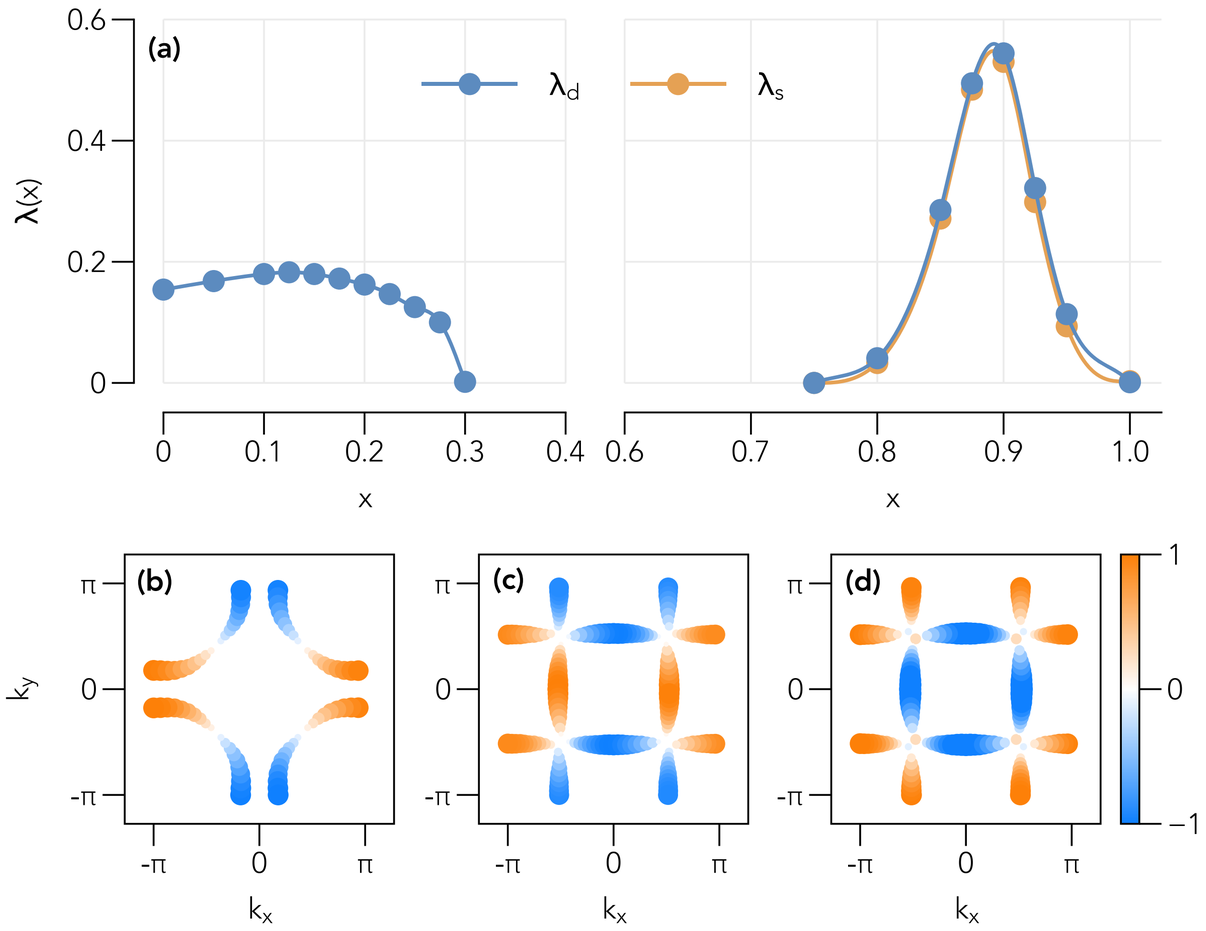


Maier, Berlijn & Scalapino, PRB '19

Random Phase Approximation (Weak Coupling) Prediction

Two pairing domes

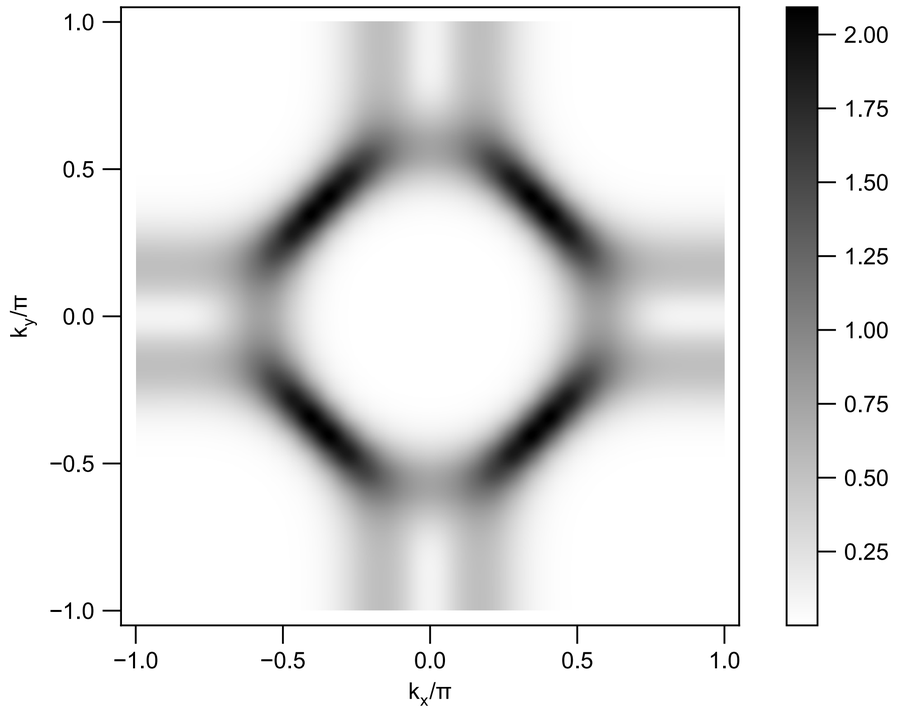
- ***First dome at small doping x*** describes superconductivity in usual, lightly doped cuprates
- ***Second dome at much larger doping*** describes recently observed superconductivity in highly overdoped cuprates
- ***Second dome has much larger pairing strength***
- For large doping, mixing of different *d*-orbitals allows for strong nesting between electron and hole Fermi surfaces
- Possibility of Bardassis-Schrieffer mode in Raman scattering due to near degeneracy of *d- and s*± pairing channels



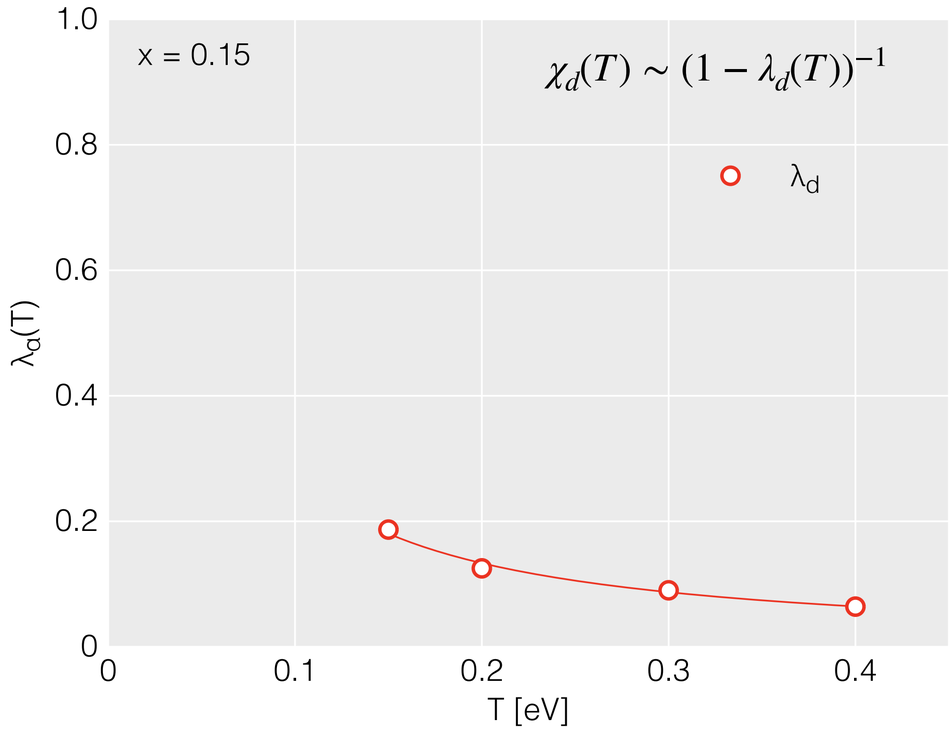
Maier, Berlijn & Scalapino, PRB '19

Dynamic Cluster Quantum Monte Carlo results: $\langle n \rangle = 2.85$ $(x = 0.15)$

Fermi surface ($|\nabla n(k)|$)



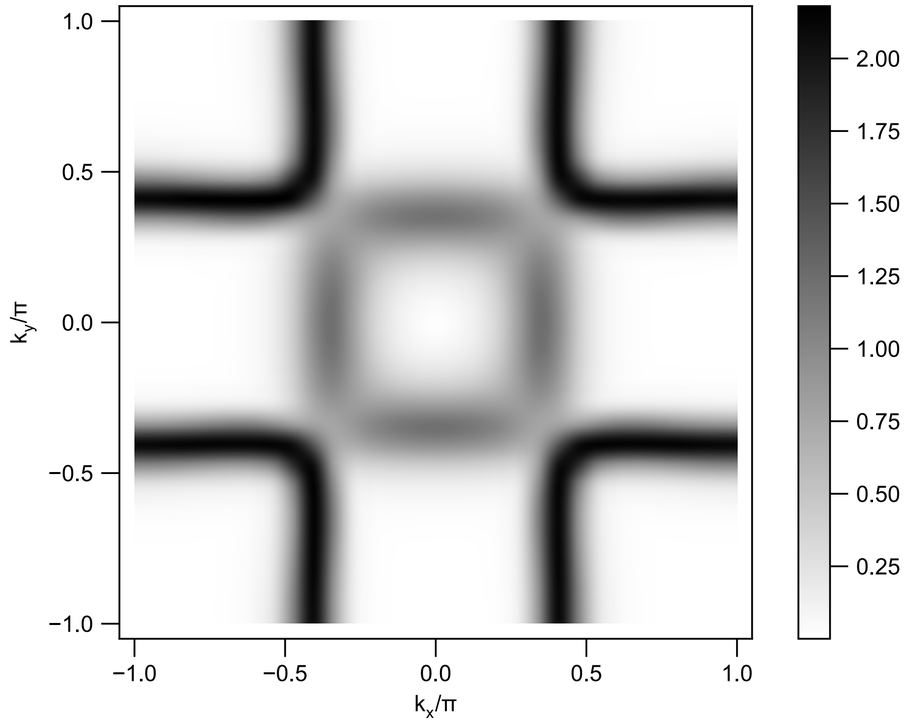
Leading eigenvalue of Bethe-Salpeter equation)



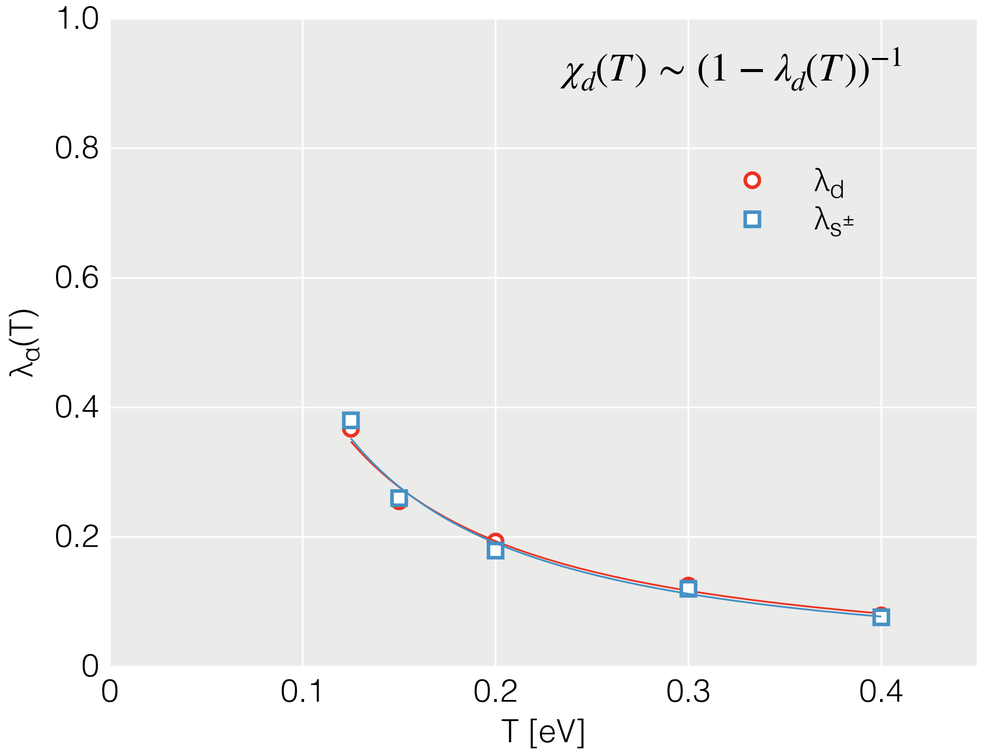
Parameters: 4 x 4 cluster; $U = 4, U' = 2, J = J' = 0$; Doping x=0.15

Dynamic Cluster Quantum Monte Carlo results: $\langle n \rangle = 2.0$ $(x = 1)$

Fermi surface ($|\nabla n(k)|$)

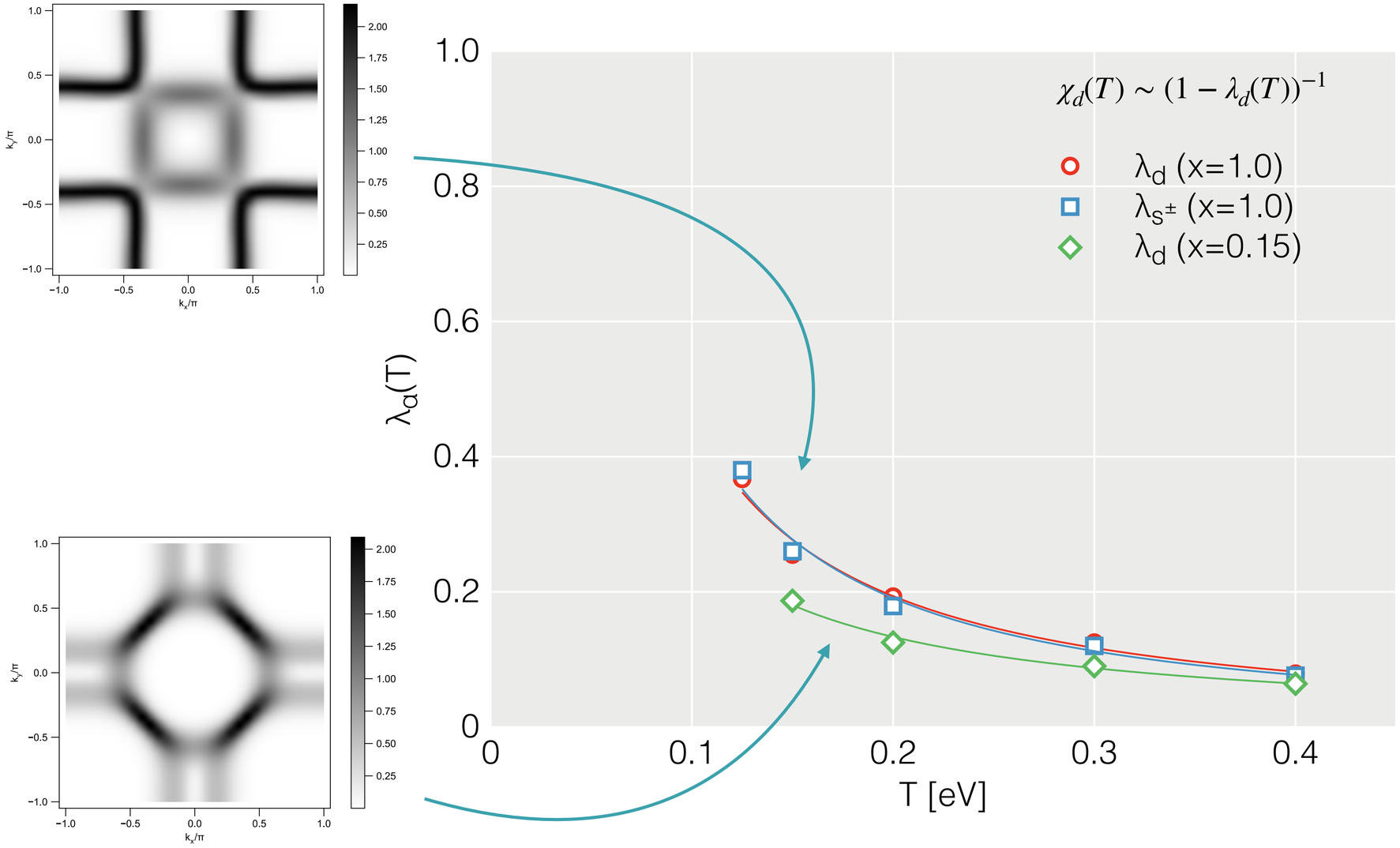


Leading eigenvalue of Bethe-Salpeter equation)



Parameters: 4 x 4 cluster; $U = 4, U' = 2, J = J' = 0$; Doping x=1

Pairing in 2-orbital model for cuprates



- Pairing is stronger in heavily overdoped region with hole and electron Fermi surface pockets
- *d*-wave and $s^\pm$ - pairing almost degenerate in heavily overdoped region

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