Intertwined order in the cuprate high-temperature superconductors

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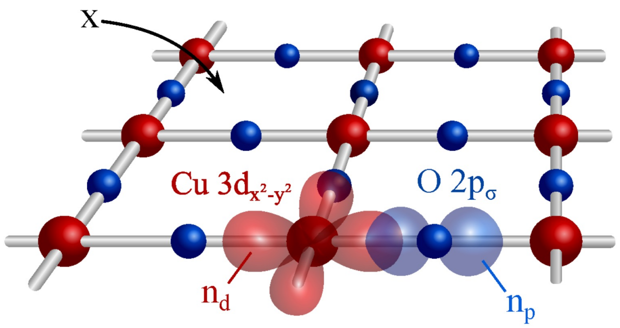
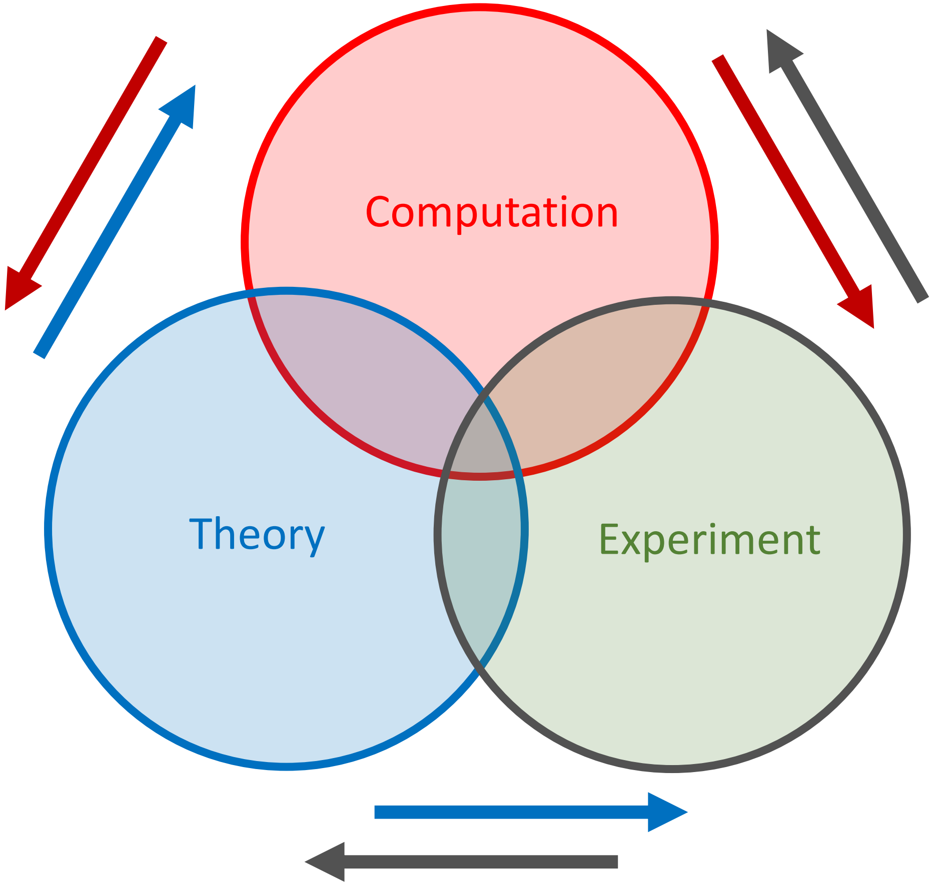
THE UNIVERSITY of TENNESSEE KNOXVILLE

U.S. DEPARTMENT OF ENERGY

My research program

Employ nonperturbative numerical methods: ED, determinant & hybrid QMC, DMRG, VMC. Major users of leadership-class computing.

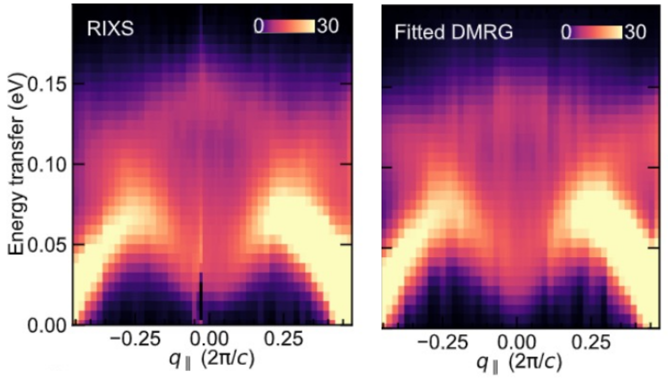




Studying models for strongly correlated quantum materials:

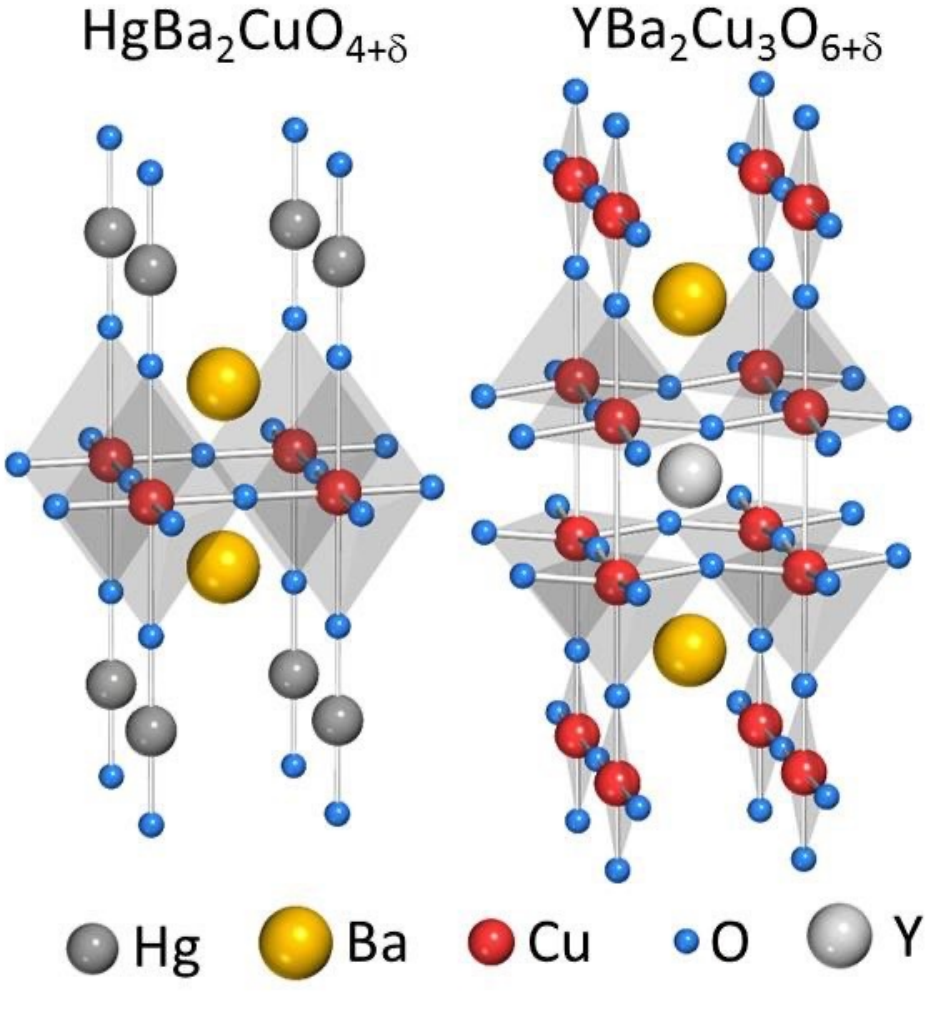
- high-T_c superconductors.
- correlated systems.
- low-dimensional materials.
- e-ph interactions.
- theory of spectroscopy.

Actively collaborating with experimental groups: e.g. ARPES, STM/STS, INS, RIXS.



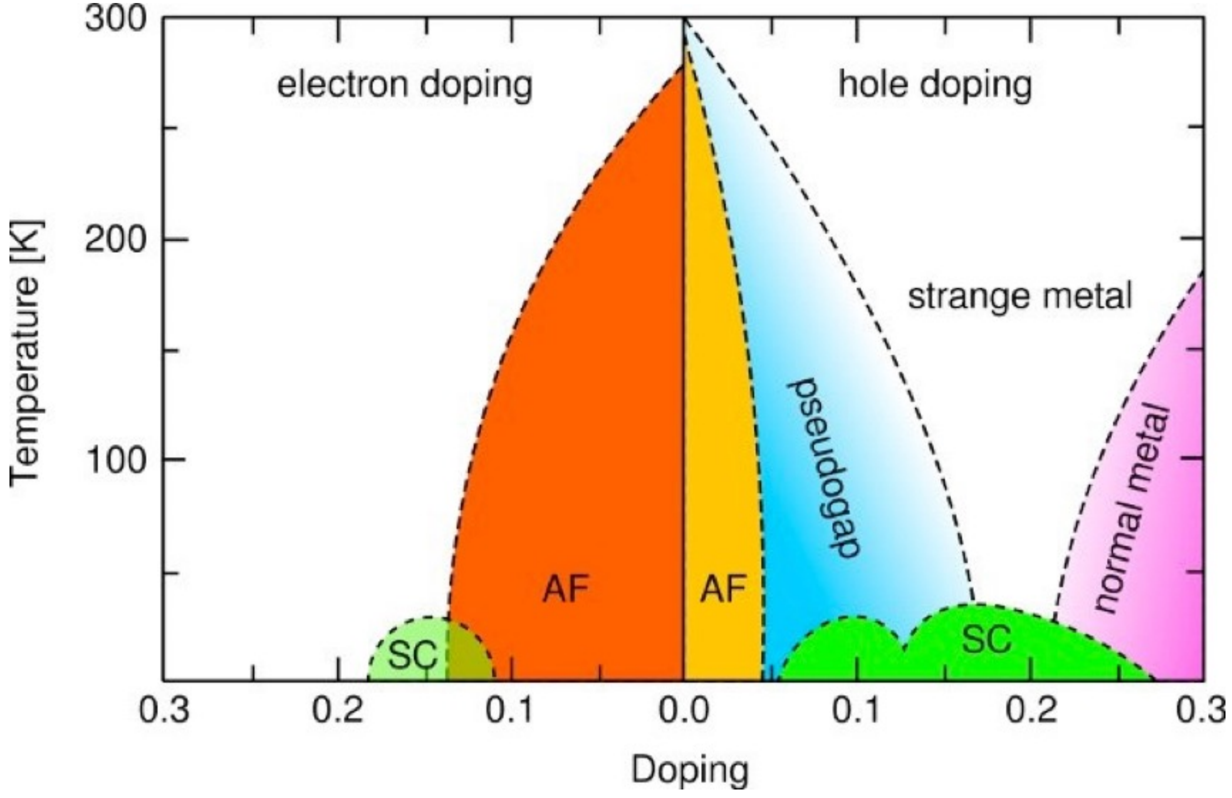
Members: 5+5 PhD, 2 MS, 3+1 Post-docs (past + present); Support from DOE, NSF, ONR

The high-T_c superconducting cuprates



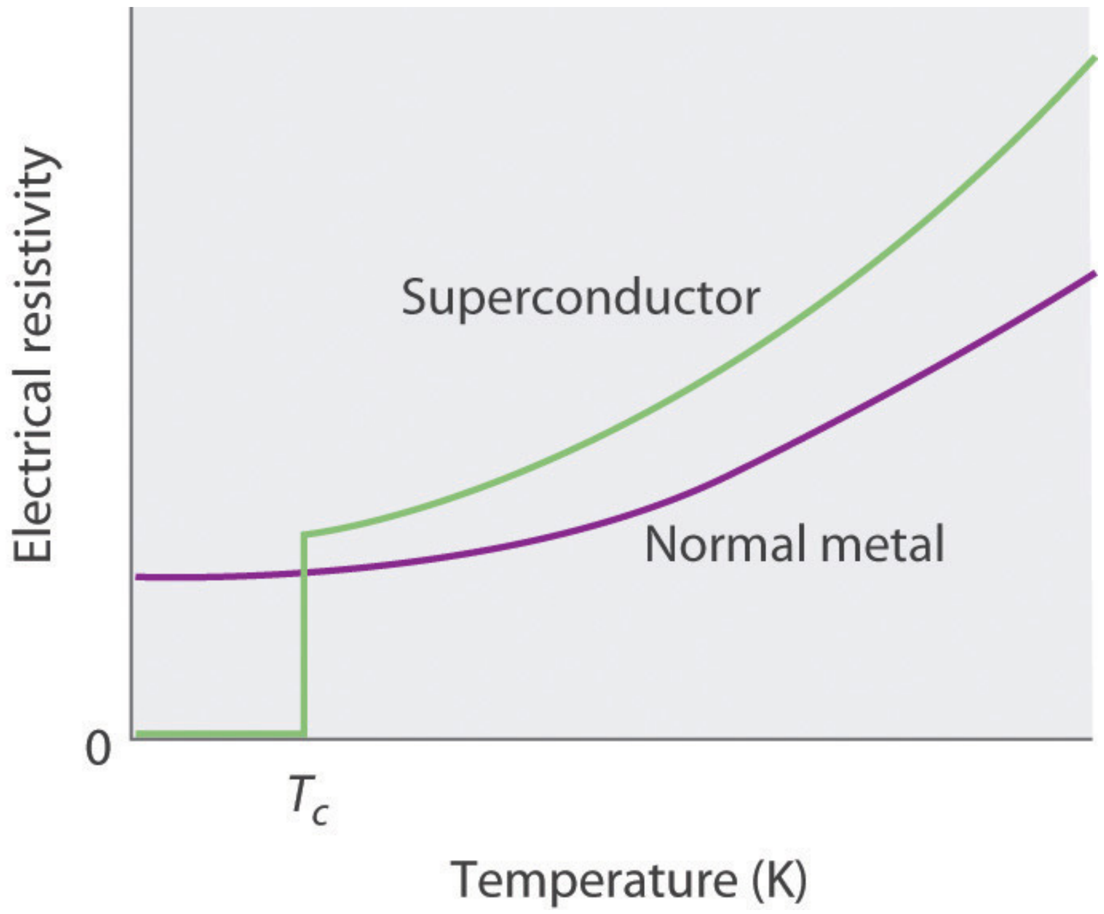
*Reichardt *et al.*, Condens. Matter **3**, 23 (2018).

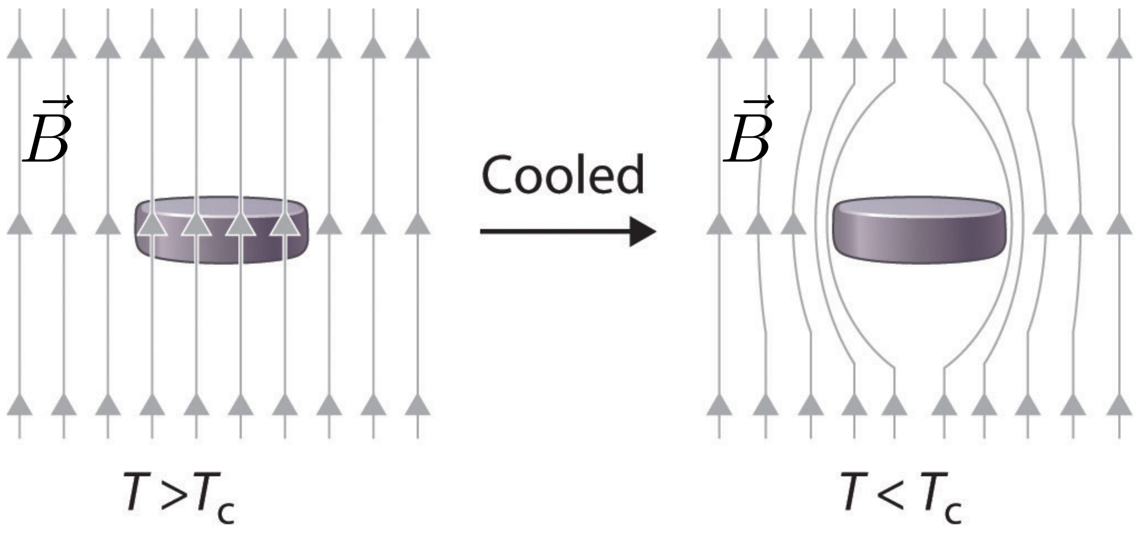
"Universal" temperature-doping phase diagram

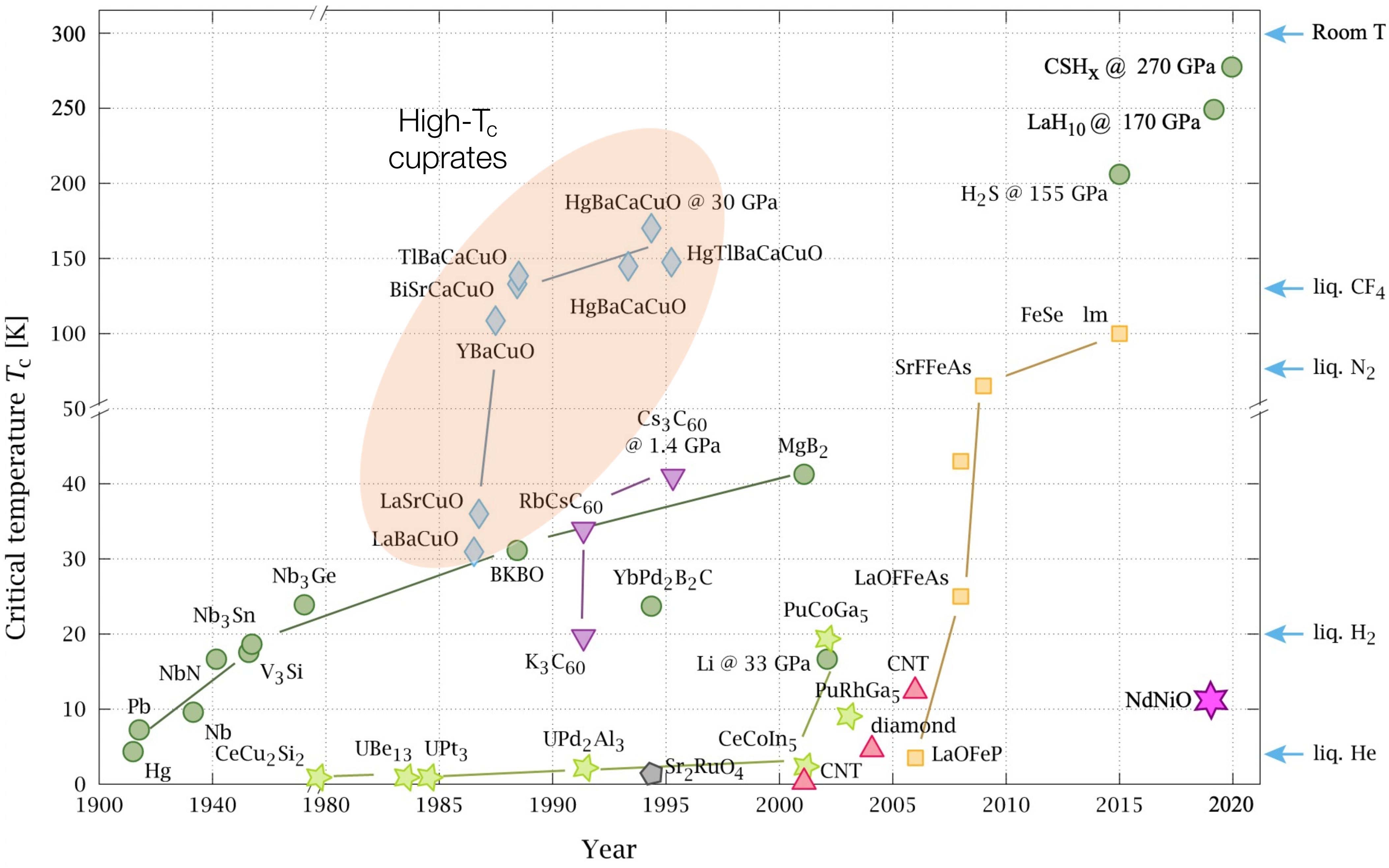


*E. Dagotto, Science **308**, 5732 (2005).

Superconductivity

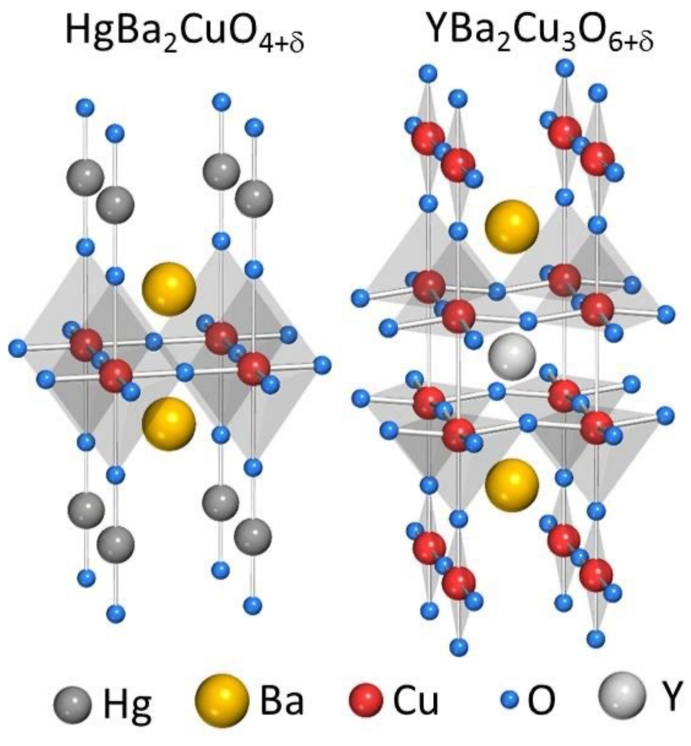




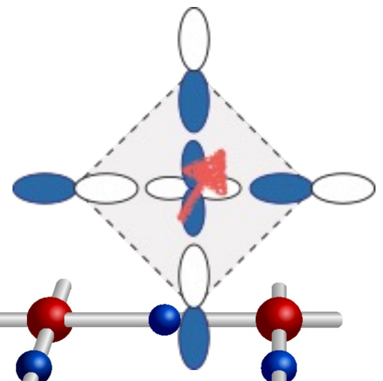


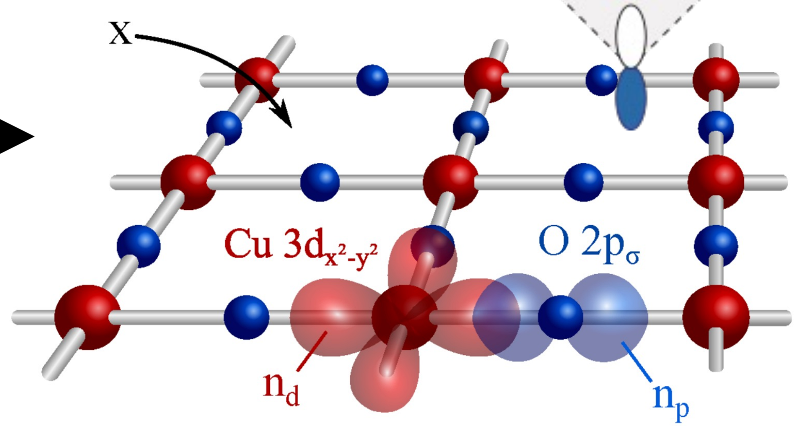
High-Tc cuprates
Room T
liq. CF4
liq. N2
liq. H2
liq. He
CSHx @ 270 GPa
LaH10 @ 170 GPa
H2S @ 155 GPa
FeSe lm
SrFFeAs
LaOFFeAs
LaOFeP
NdNiO
CNT
diamond
PuCoGa5
PuRhGa5
Li @ 33 GPa
CeCoIn5
CNT
MgB2
Cs3C60
@ 1.4 GPa
RbCsC60
K3C60
YbPd2B2C
Sr2RuO4
UPd2Al3
UPt3
UBe13
CeCu2Si2
HgBaCaCuO @ 30 GPa
HgTlBaCaCuO
HgBaCaCuO
TlBaCaCuO
BiSrCaCuO
YBaCuO
LaSrCuO
LaBaCuO
BKBO
Nb3Ge
Nb3Sn
NbN
V3Si
Pb
Nb
Hg
Critical temperature Tc [K]
Year
0
10
20
30
40
50
100
150
200
250
300
1900
1940
1980
1985
1990
1995
2000
2005
2010
2015
2020

From the real material to the Hubbard model

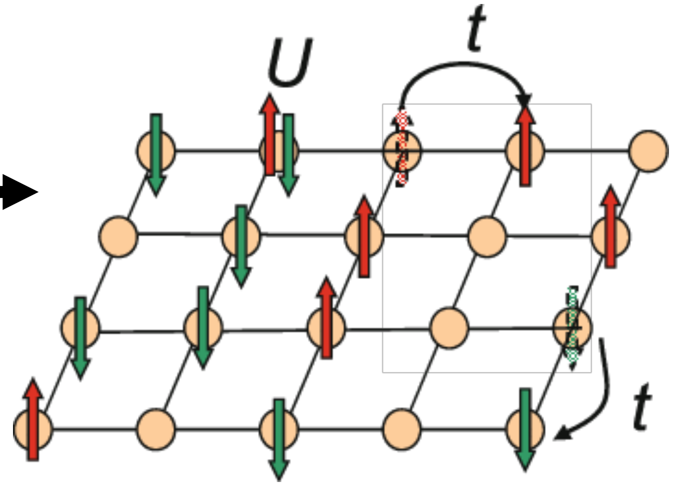


Isolate the CuO_2 plane





remove O from the problem



$$U/t \approx 6-8$$

$$H = -\sum_{\alpha=1}^{N_a} \frac{\hbar^2 \nabla_\alpha^2}{2M_\alpha} + \frac{1}{2}\sum_{\alpha\neq\alpha'} \frac{Z_\alpha Z_{\alpha'} e^2}{|\mathbf{R}_\alpha - \mathbf{R}_{\alpha'}|} - \sum_{\mu=1}^{N_e} \frac{\hbar^2 \nabla_\mu^2}{2m} + \frac{1}{2}\sum_{\mu\neq\mu'} \frac{e^2}{|\mathbf{r}_\mu - \mathbf{r}_{\mu'}|} - \sum_{\mu,\alpha} \frac{Z_\alpha e^2}{|\mathbf{R}_\alpha - \mathbf{r}_\mu|}$$

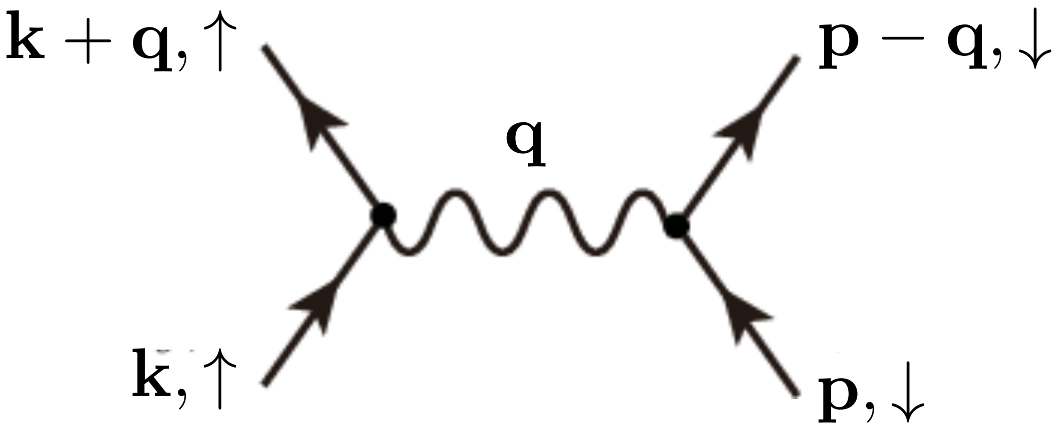
$$\longrightarrow$$

$$H = -\sum_{\mathbf{i},\mathbf{j},\sigma} t_{\mathbf{ij}} \left(c_{\mathbf{i}\sigma}^\dagger c_{\mathbf{j}\sigma} + \mathrm{h.c.} \right) - \mu \sum_{\mathbf{i},\sigma} n_{\mathbf{i}\sigma} + U \sum_{\mathbf{i}} n_{\mathbf{i}\uparrow} n_{\mathbf{i}\downarrow}.$$

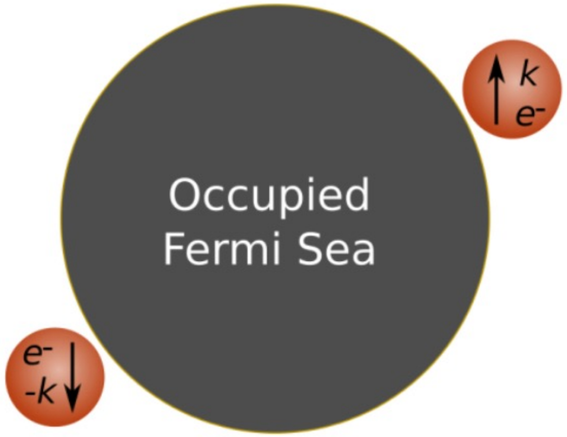
Images from Reichardt *et al.*, Condens. Matter **3**, 23 (2018).

Superconductivity: Cooper pairing

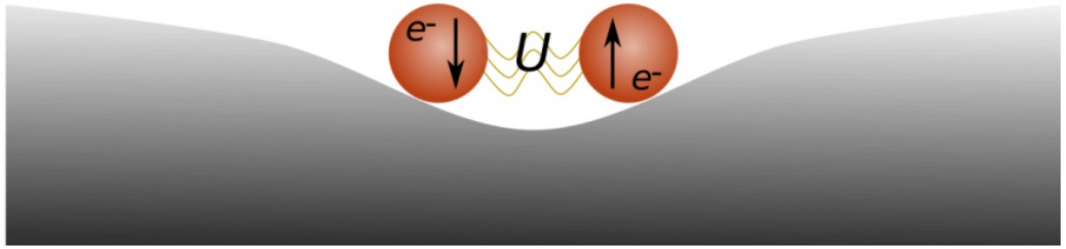
The electron-phonon interaction mediates an effective *attractive* interaction between electrons.



Leon Cooper* showed that two electrons above the Fermi sea will form a bound state.



$$E = 2\epsilon_{\mathrm{F}} - 2\Omega_{\mathrm{D}} e^{-\frac{2}{\lambda}} < 2e_{\mathrm{F}}$$

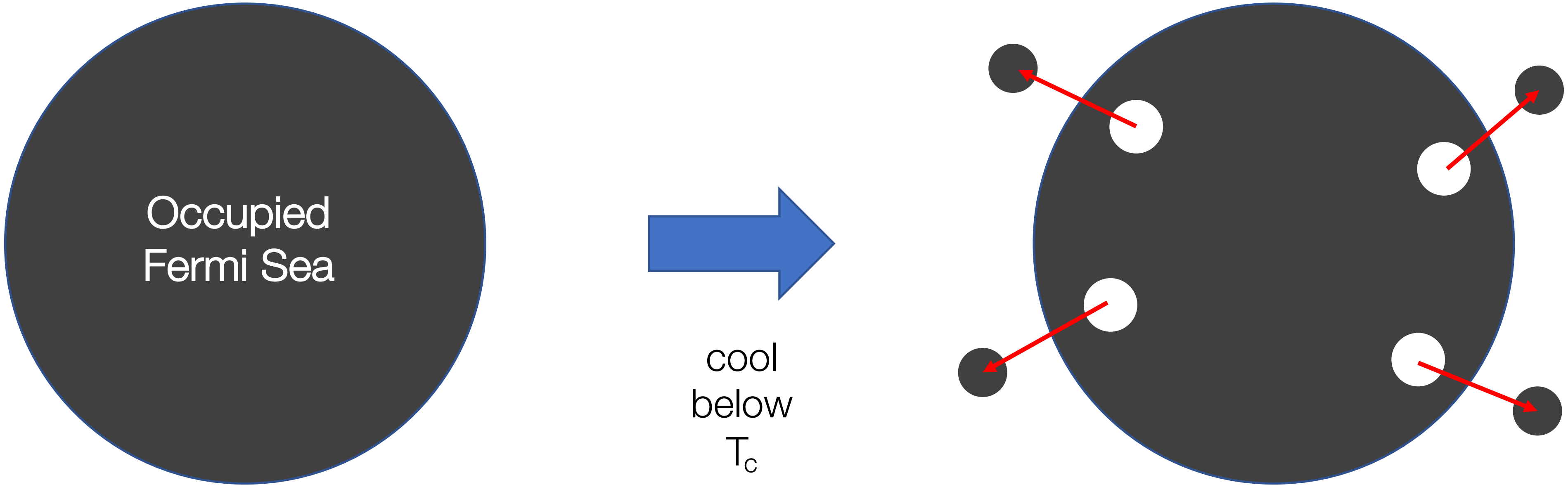


* L. N. Cooper, Phys. Rev. **104**, 1189 (1956).

Superconductors: coherent states of Cooper pairs

The “normal” state” (a metal)

The superconducting state



$|\mathrm{FS}\rangle$

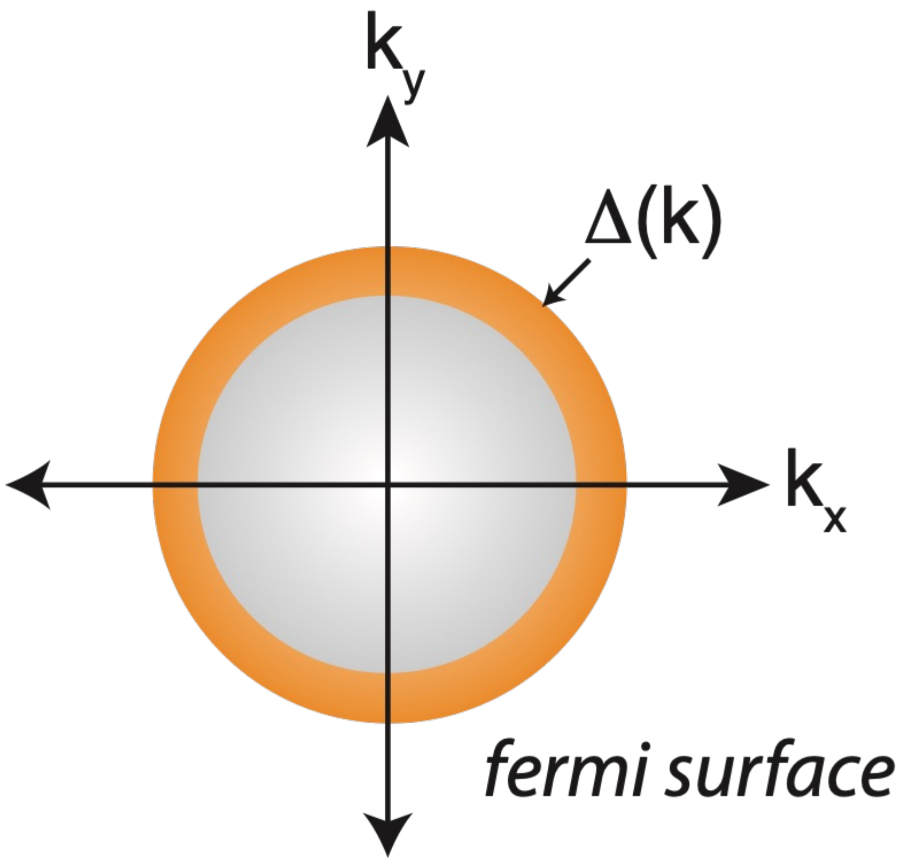
$$|\Psi_{\mathrm{BCS}}\rangle = \prod_{k>k_{\mathrm{F}}} \left(u_{\mathbf{k}} + v_{\mathbf{k}} c^{\dagger}_{\mathbf{k},\uparrow} c^{\dagger}_{-\mathbf{k}},\downarrow \right)$$

$$\times \prod_{k<k_{\mathrm{F}}} \left(u_{\mathbf{k}} c_{-\mathbf{k}} c_{\mathbf{k},\uparrow} + v_{\mathbf{k}},\downarrow \right) |\mathrm{FS}\rangle$$

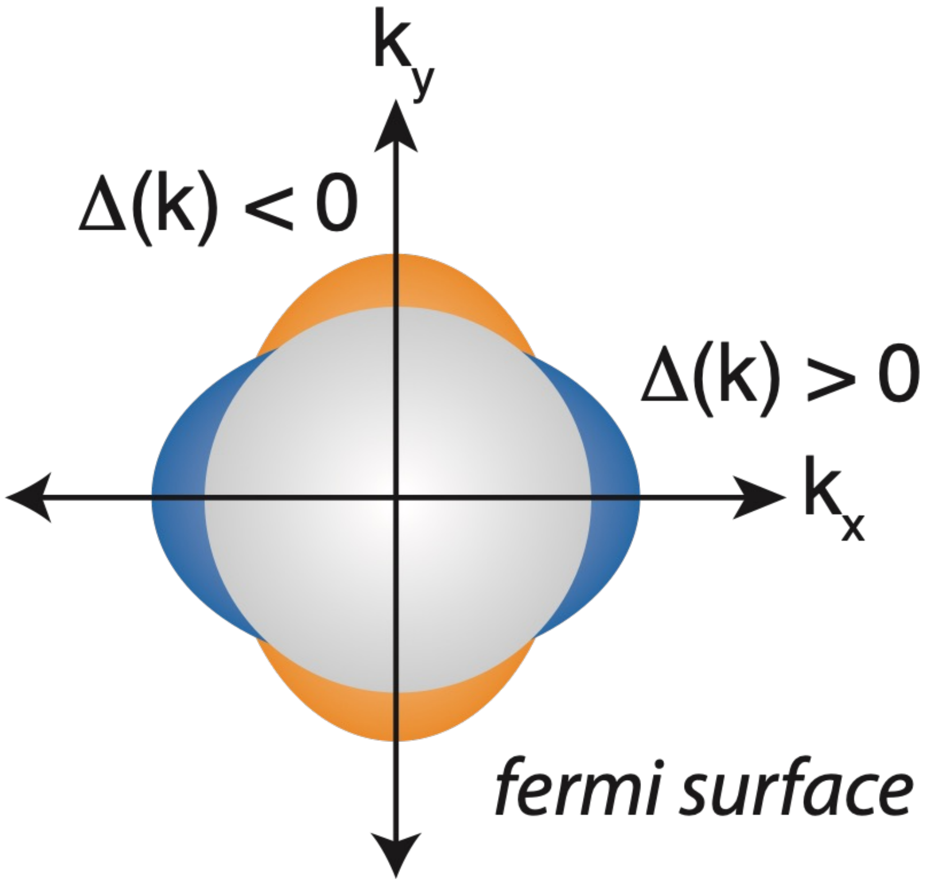
* BCS, Phys. Rev. **106**, 162 (1957); BCS, Phys. Rev. **108**, 1175 (1957)

The superconducting gap symmetry

Attractive interaction (e.g. phonons)
s-wave gap symmetry

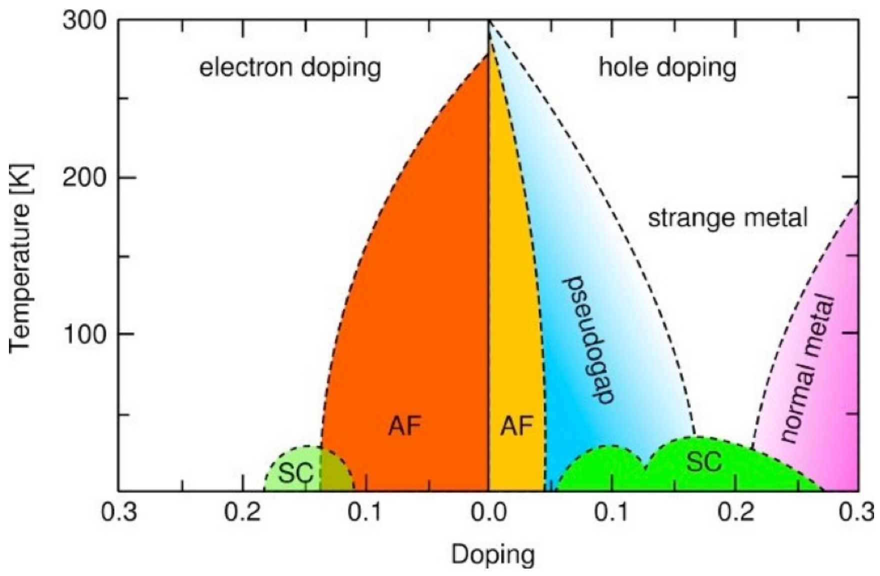


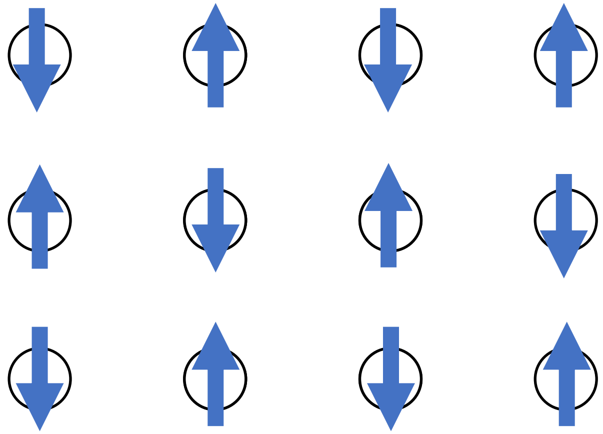
Repulsive interaction (e.g. magnons)
d-wave gap symmetry



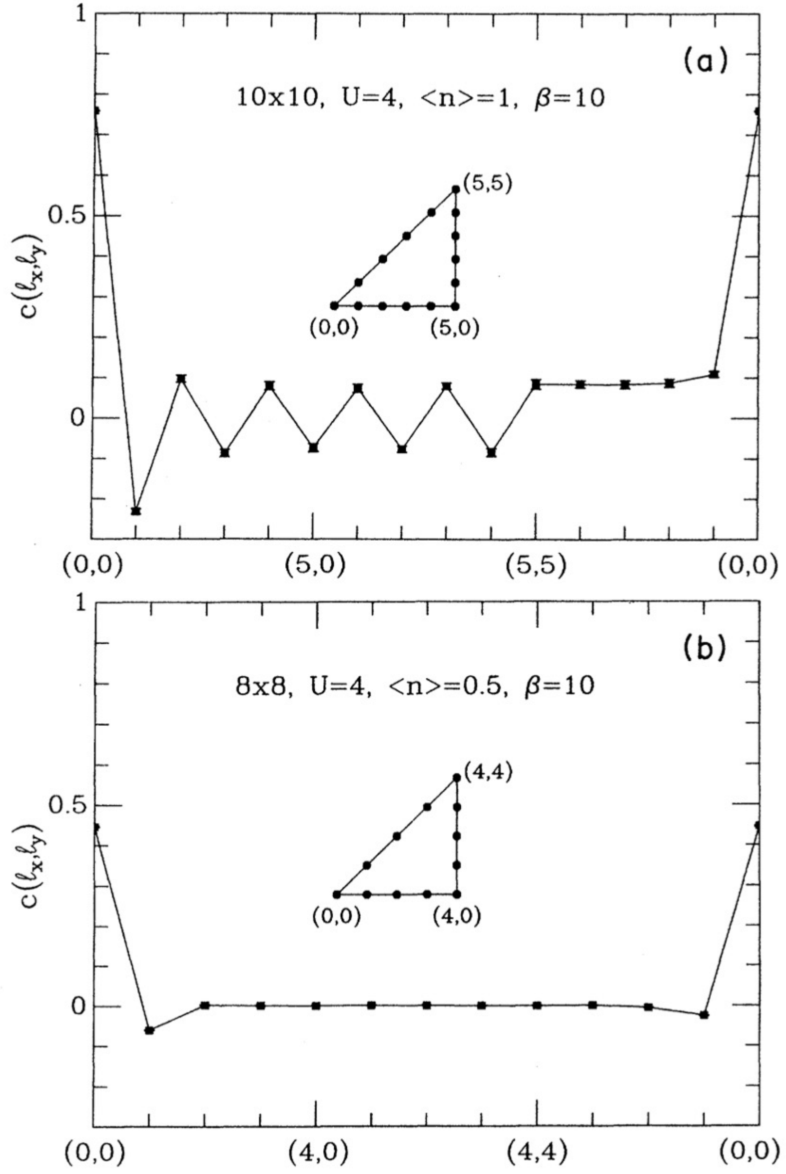
What is the pairing mechanism in the cuprate superconductors?

Early picture: doping a Mott insulator



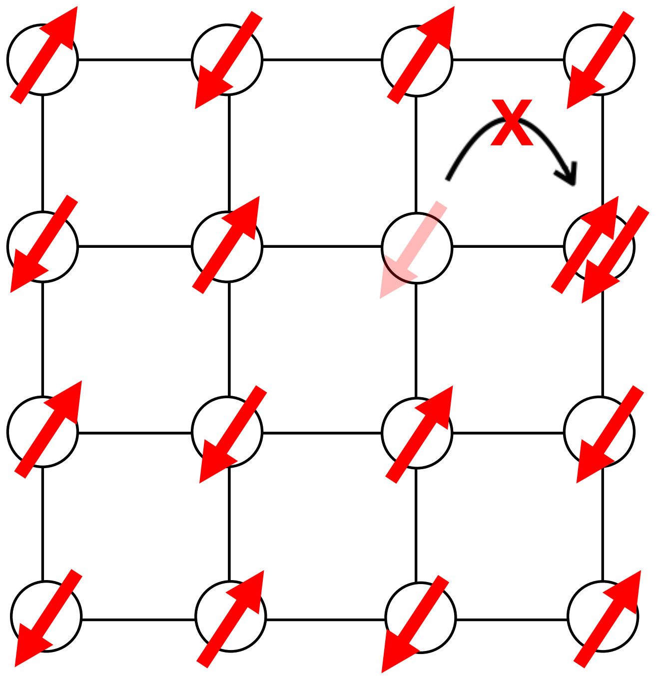


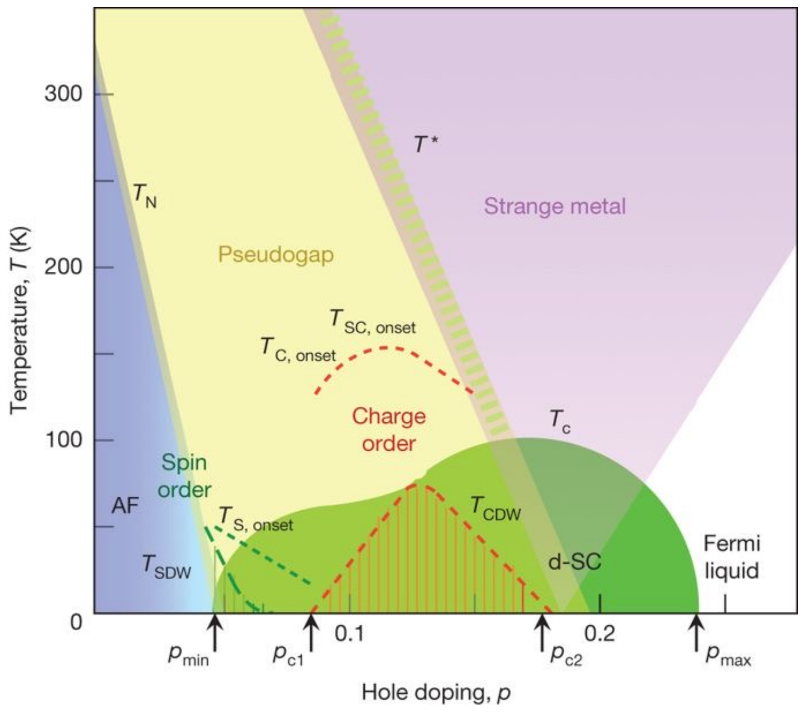
Early DQMC simulations

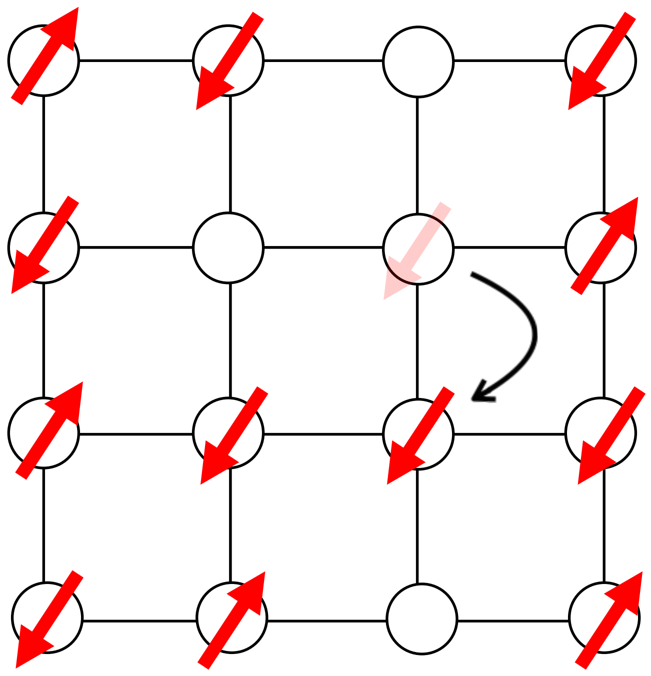


*S.R. White *et al.*, Phys. Rev. B **40**, 506 (1989).

Balancing energies in the cuprates







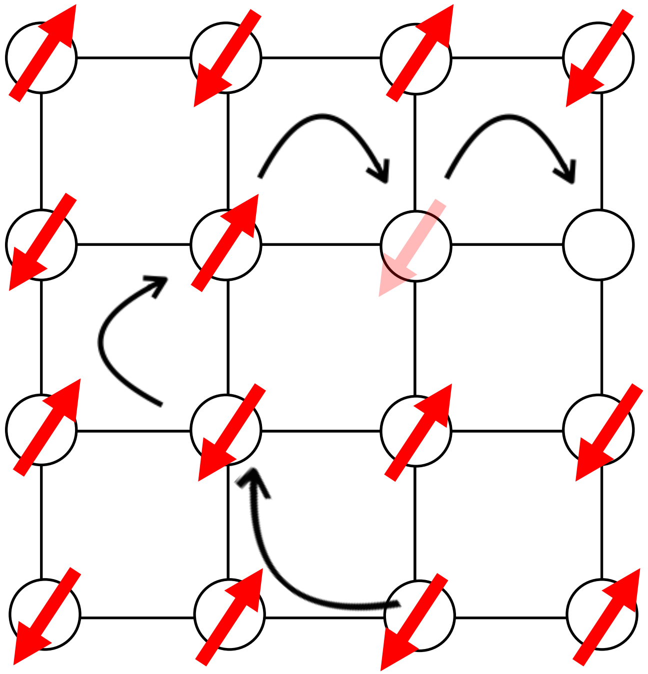
No doped holes:
1 carrier / Cu, AFM Insulator

Many doped holes:
Good conduction

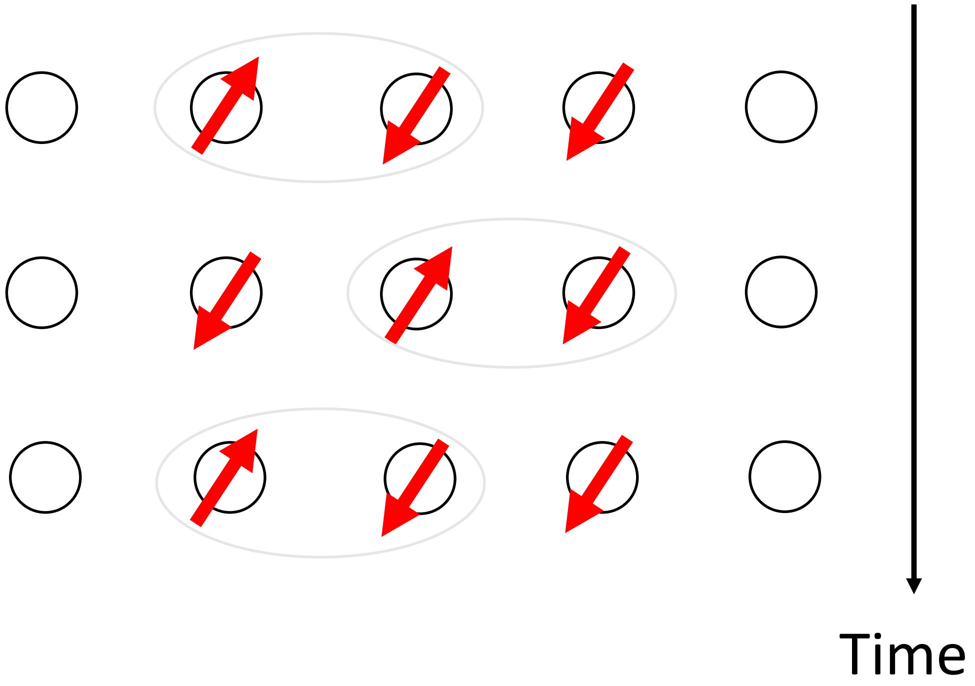
How do we move from one scenario to the other?

Balancing energies in the cuprates

Only Correlated/Collective motion is allowed.



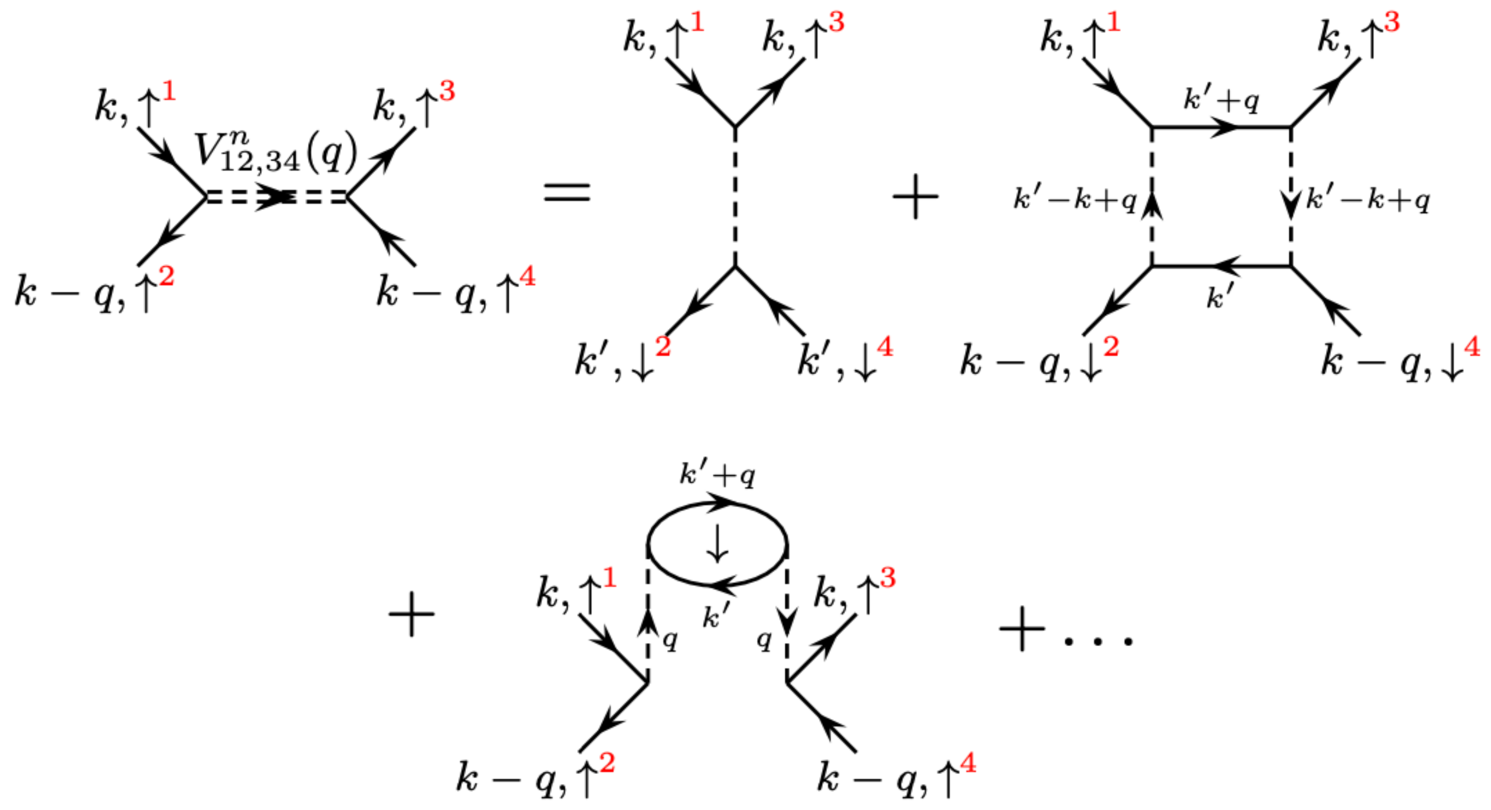
Pairing somehow emerges at small to intermediate doping levels.

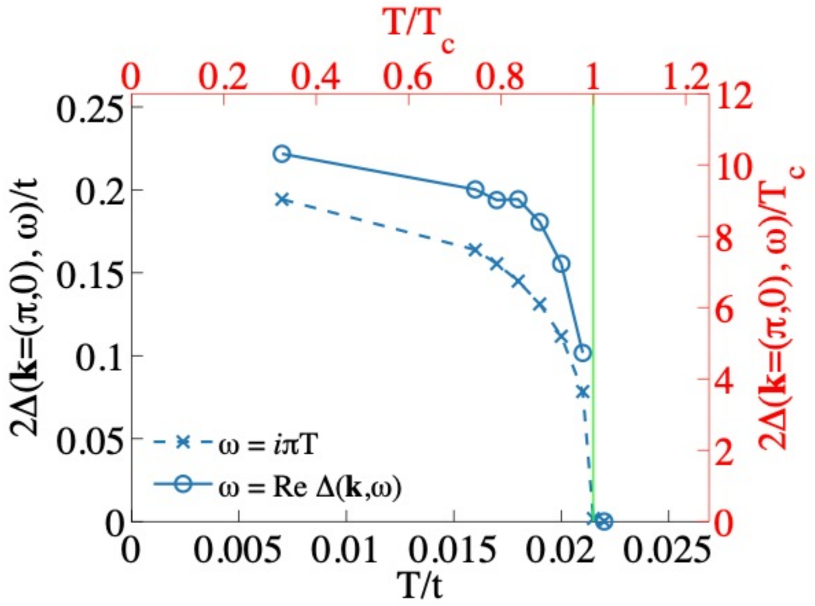


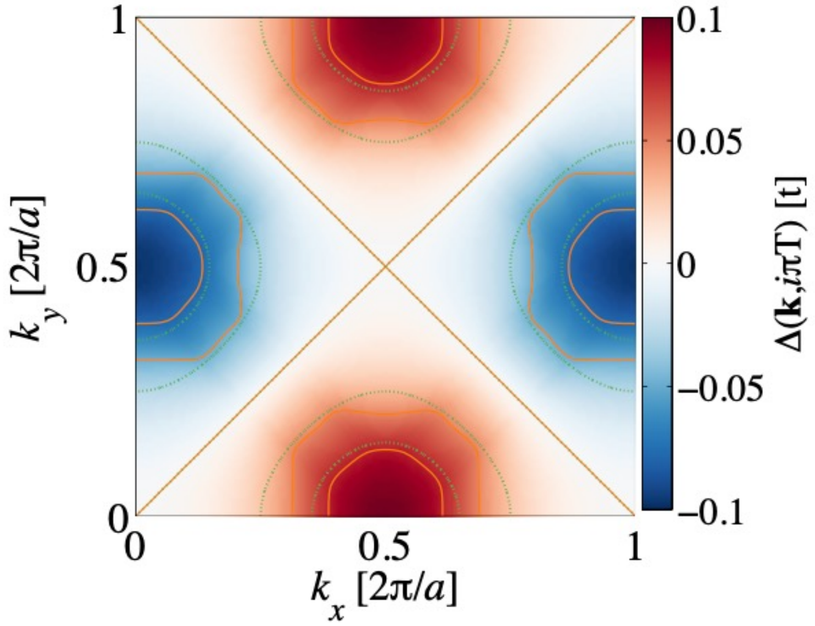
Understanding the spin and charge dynamics in this regime is crucial for understanding superconductivity!

Early picture: doping a Mott insulator

The analog of the Cooper problem, exchange of a spin fluctuations



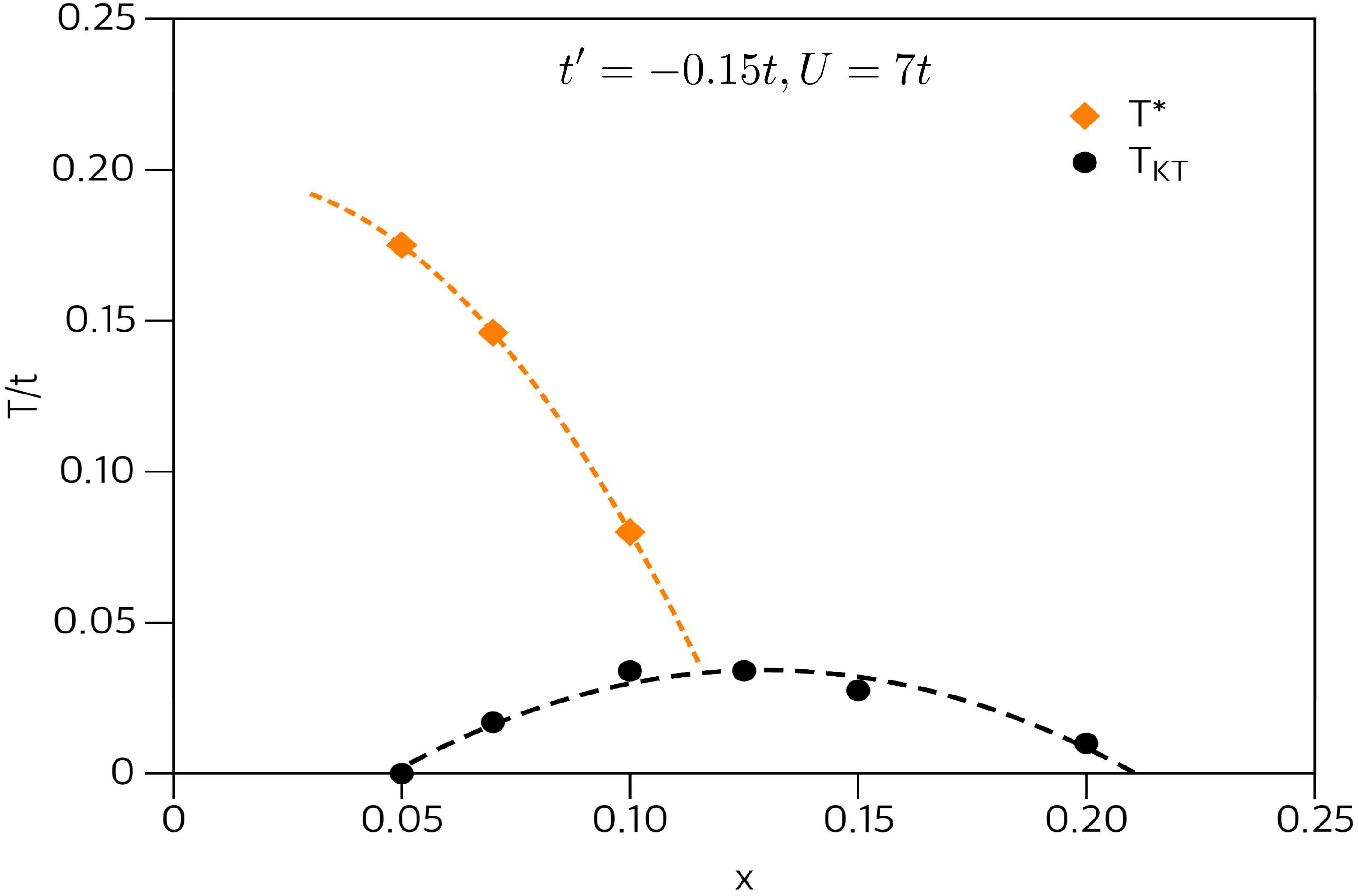




*P. Monthoux and D. J. Scalapino, Phys. Rev. Lett., 472, 1874 (1994).

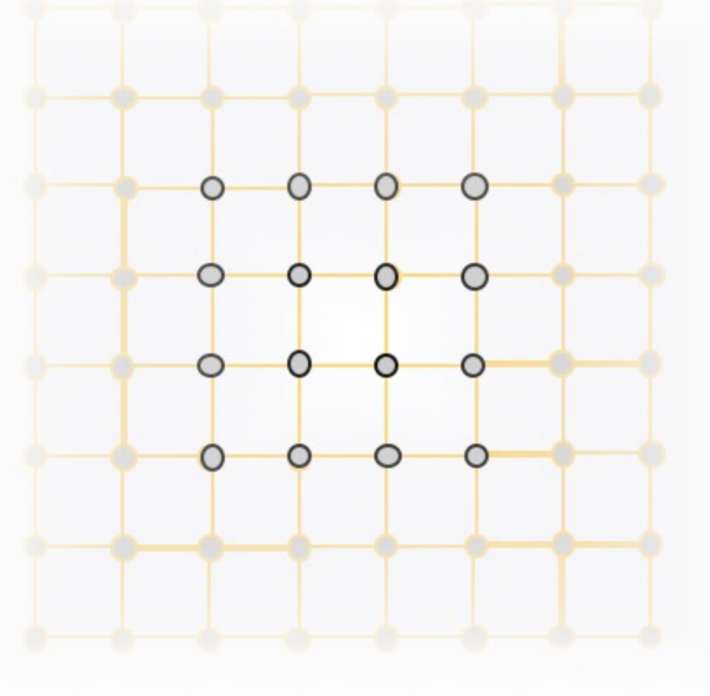
Superconductivity in embedded cluster methods

DCA Phase Diagram, 12 site cluster



*T. A. Maier *et al.*, PRL **95**, 237001 (2005).

DCA: finite cluster in a mean-field

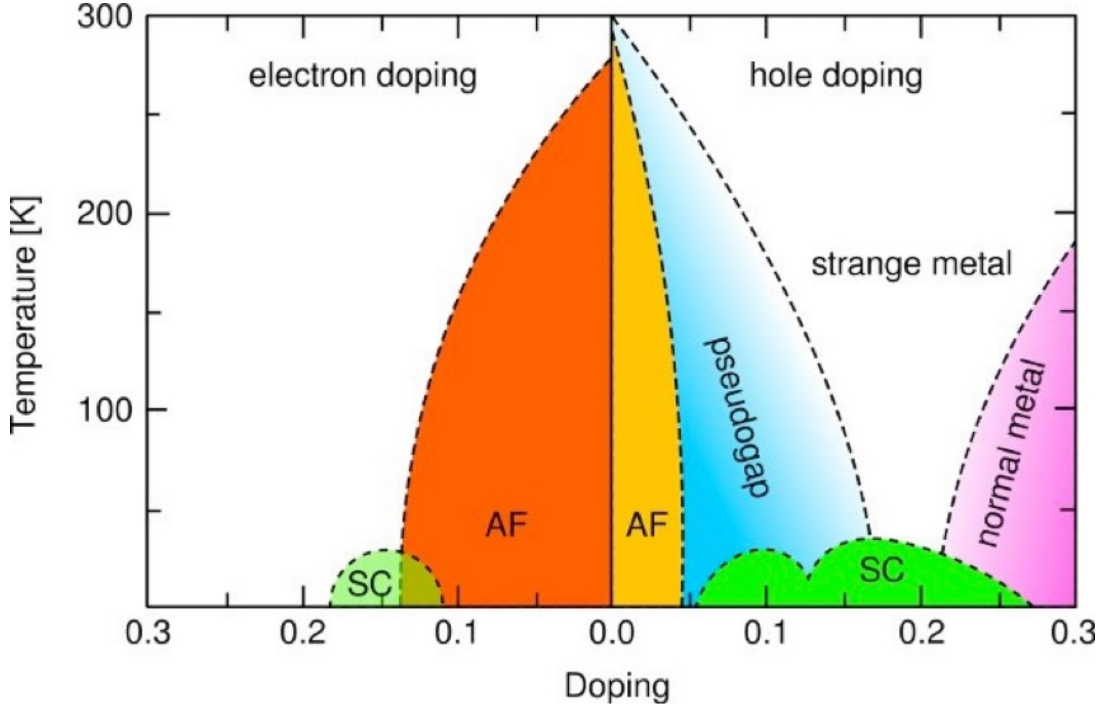


Robust d-wave superconductivity

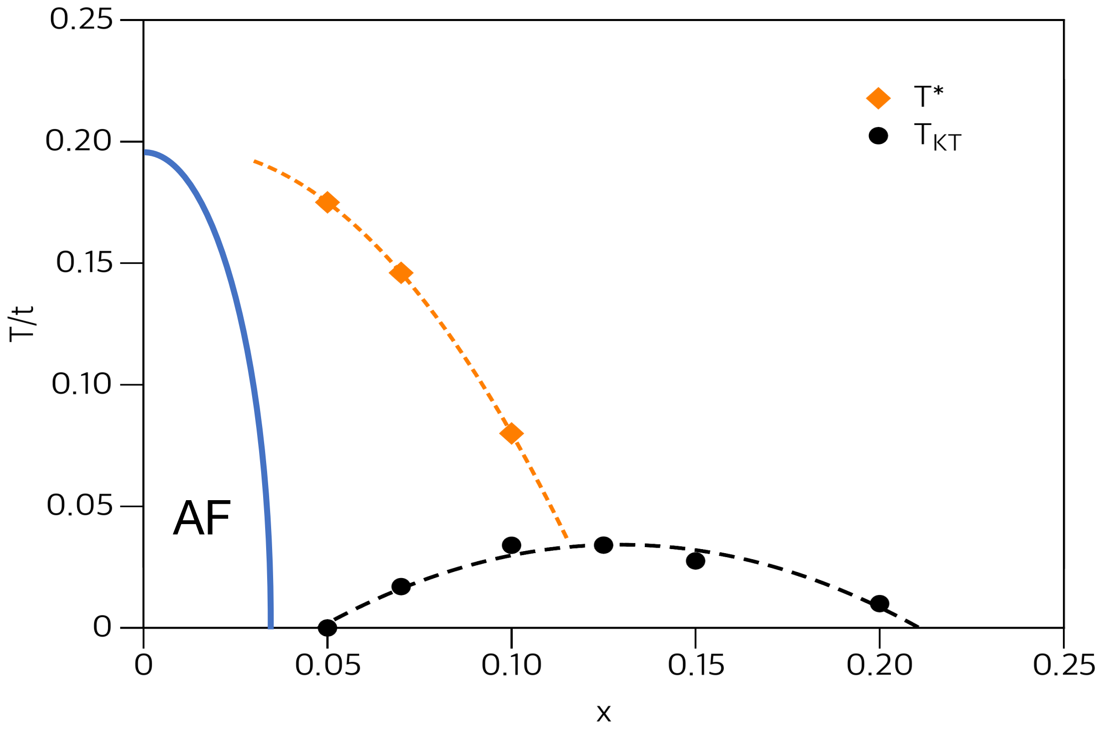
- In clusters up to 30 sites.
- Max T_c/t ~ 0.025 – 0.05, depending on model parameters.

Mission Accomplished!

Cuprate Phase Diagram



Singleband Hubbard Model



Superconductivity: Cooper pairing

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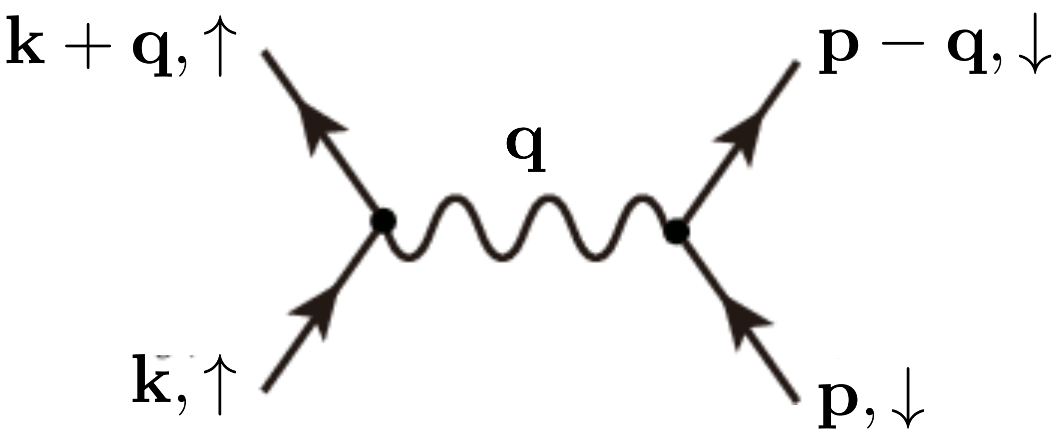

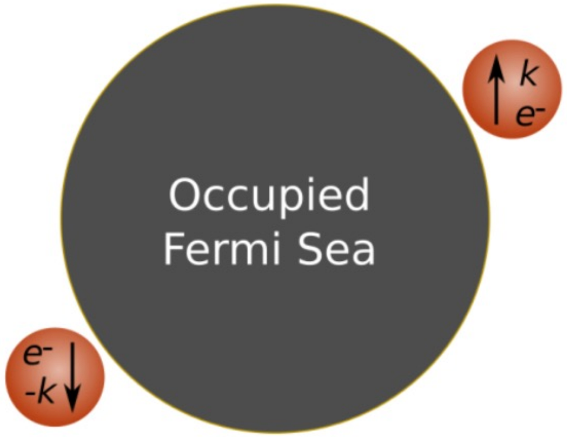

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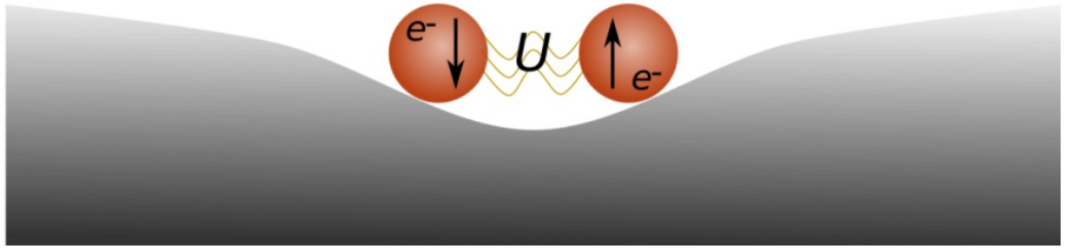
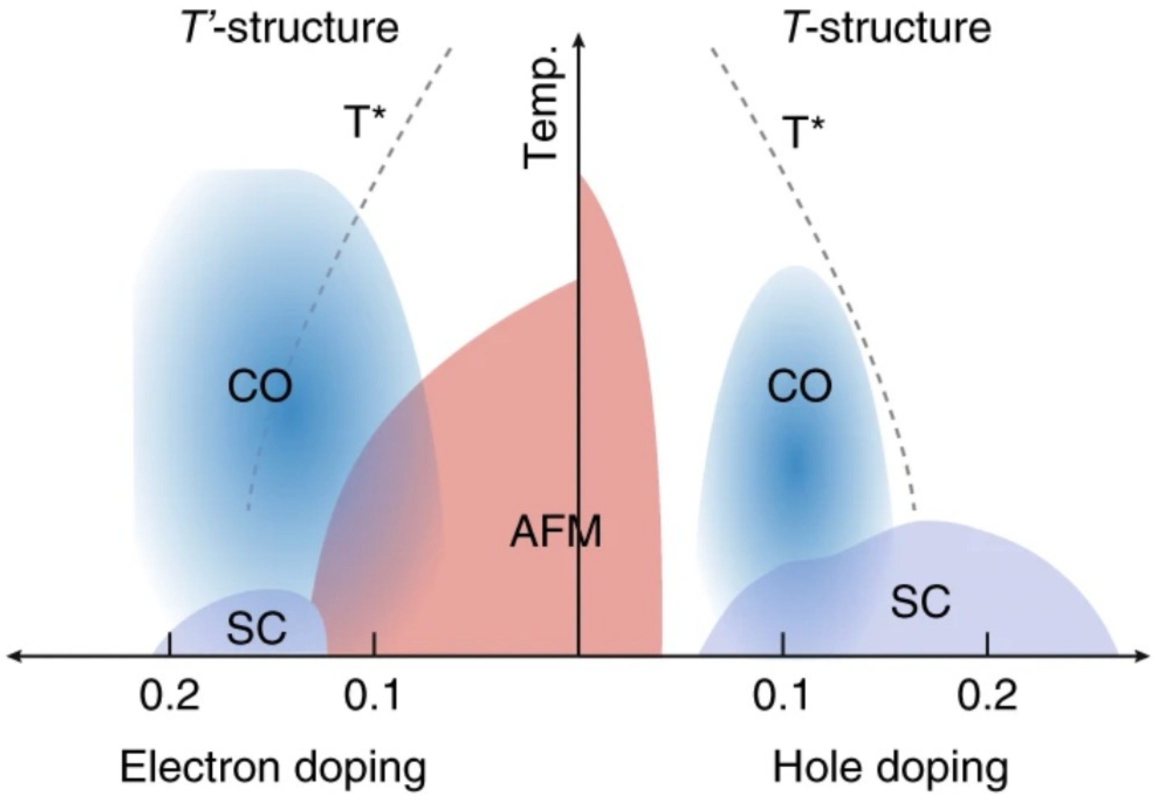


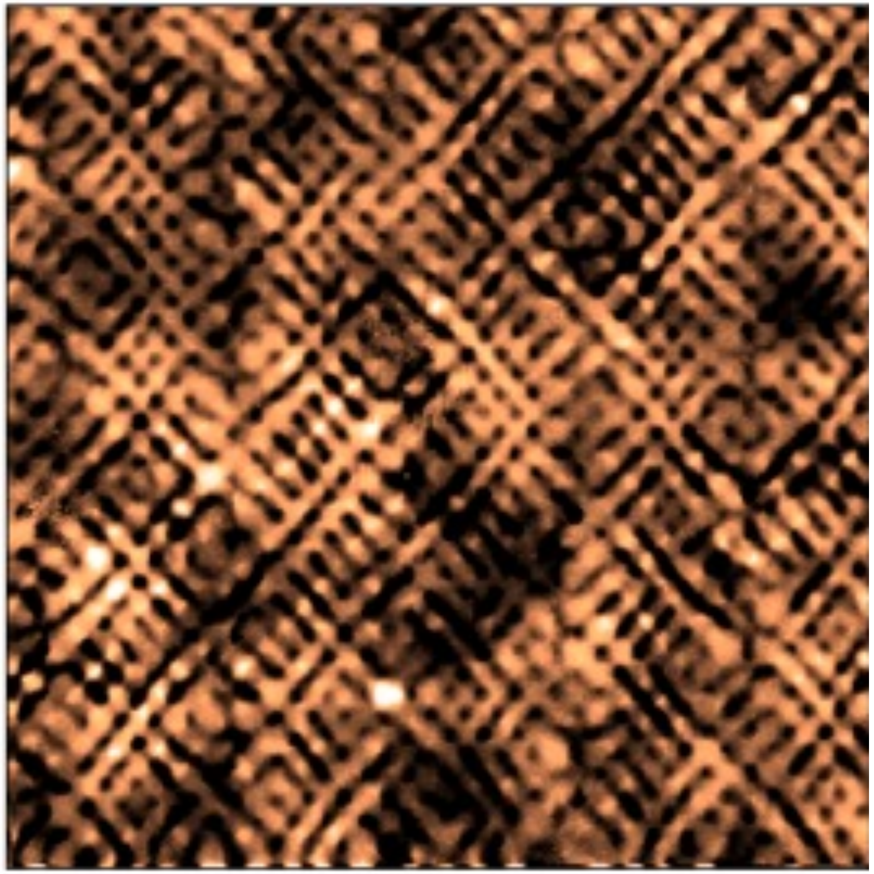
* L. N. Cooper, Phys. Rev. **104**, 1189 (1956).

The high-T_c cuprates today



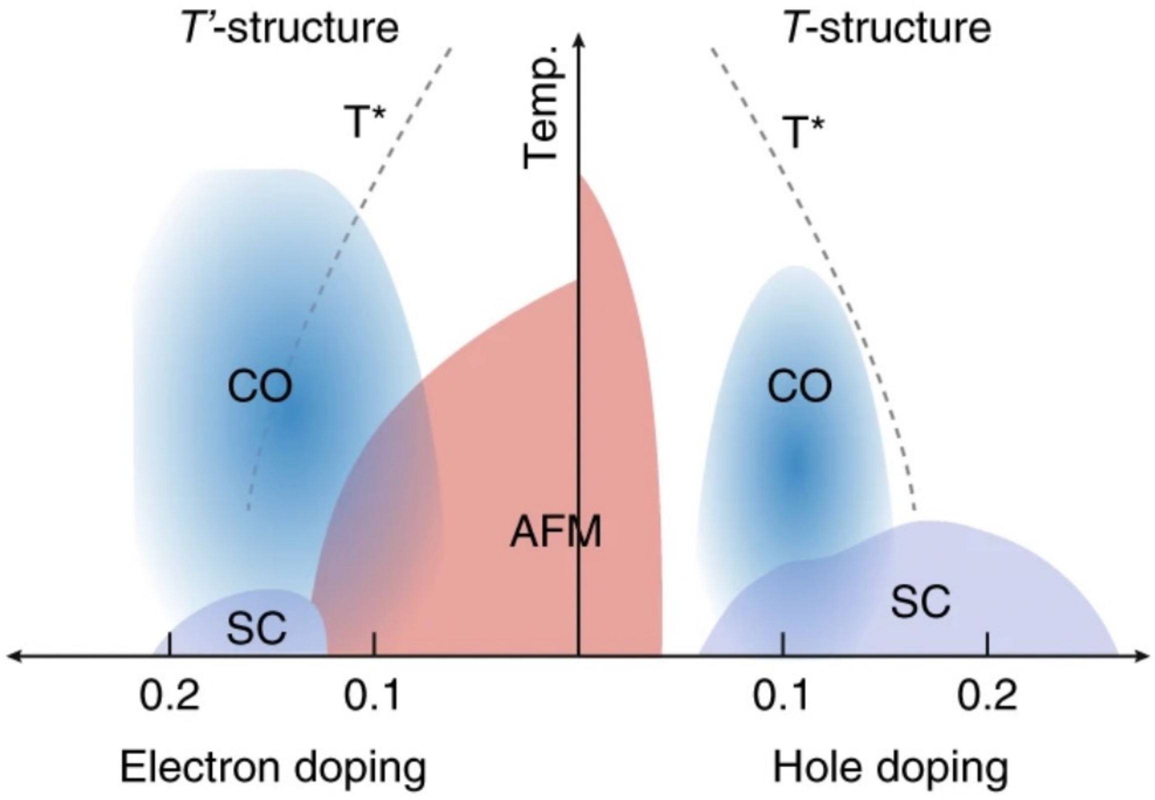
*Kang *et al.*, Nature Physics **15**, 335 (2015).

$Bi_{2.2}Sr_{1.8}Ca_{0.8}Dy_{0.2}Cu_2O_y$

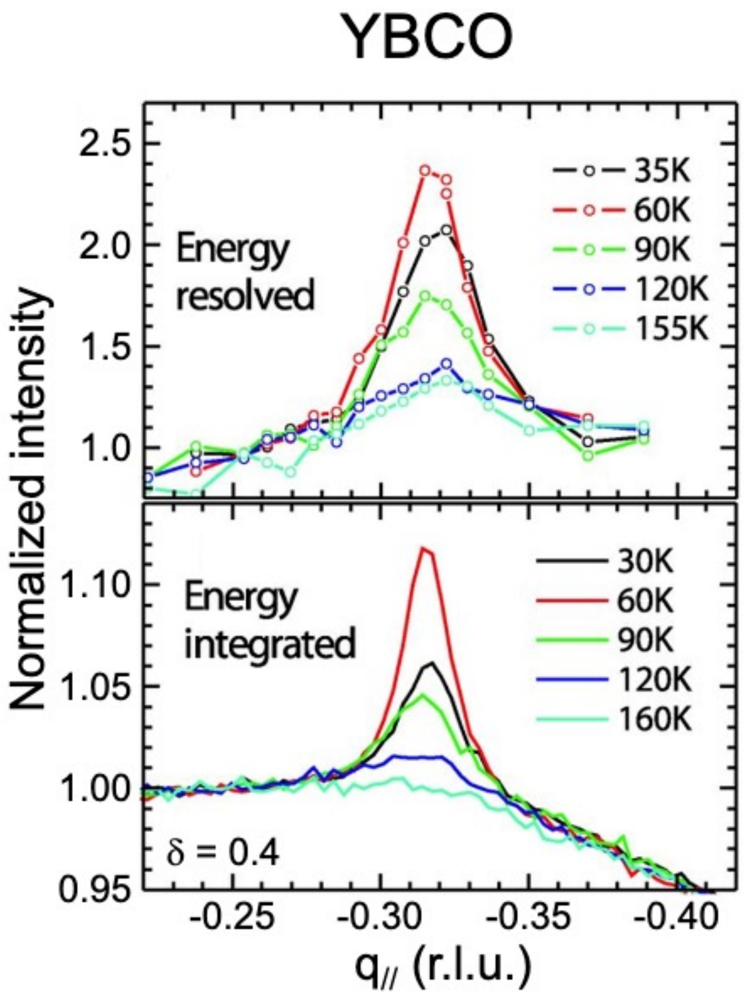


*J. E. Hoffman *et al.*, Science **295**, 466 (2002), and many others!

The high-T_c cuprates today



*Kang *et al.*, Nature Physics **15**, 335 (2015).



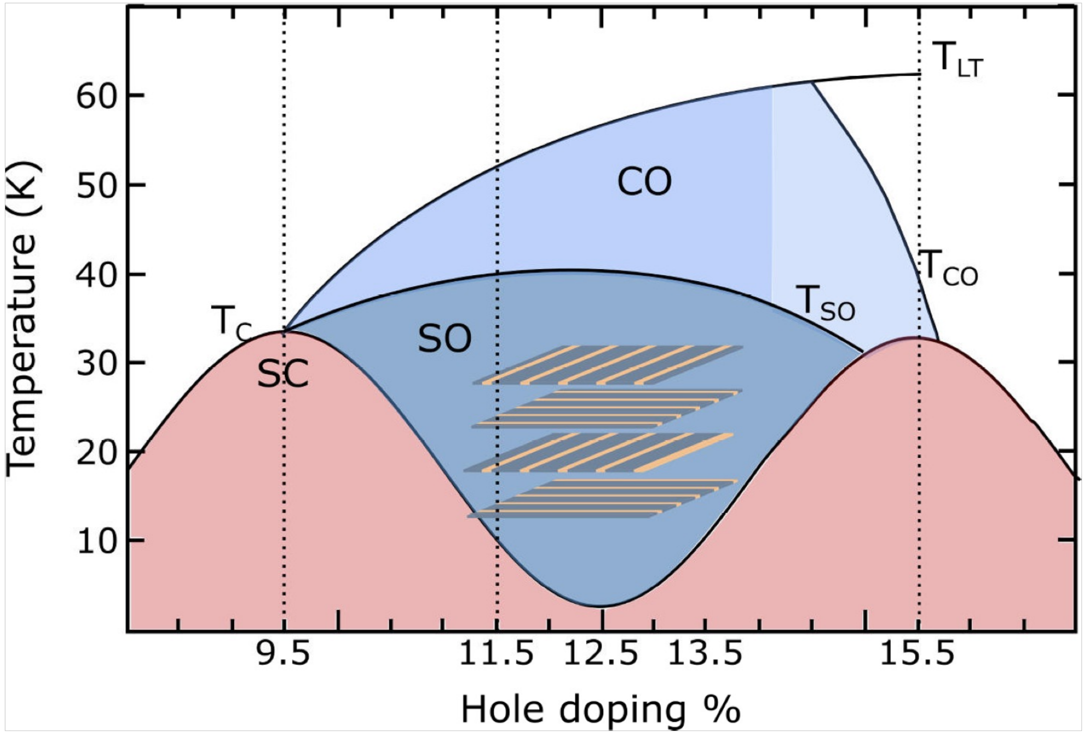
Review, see: R. Arpaia and G. Ghiringhelli, J. Phys. Soc. Jpn. **90**, 111005 (2021)

What is the pairing mechanism in the cuprate superconductors?

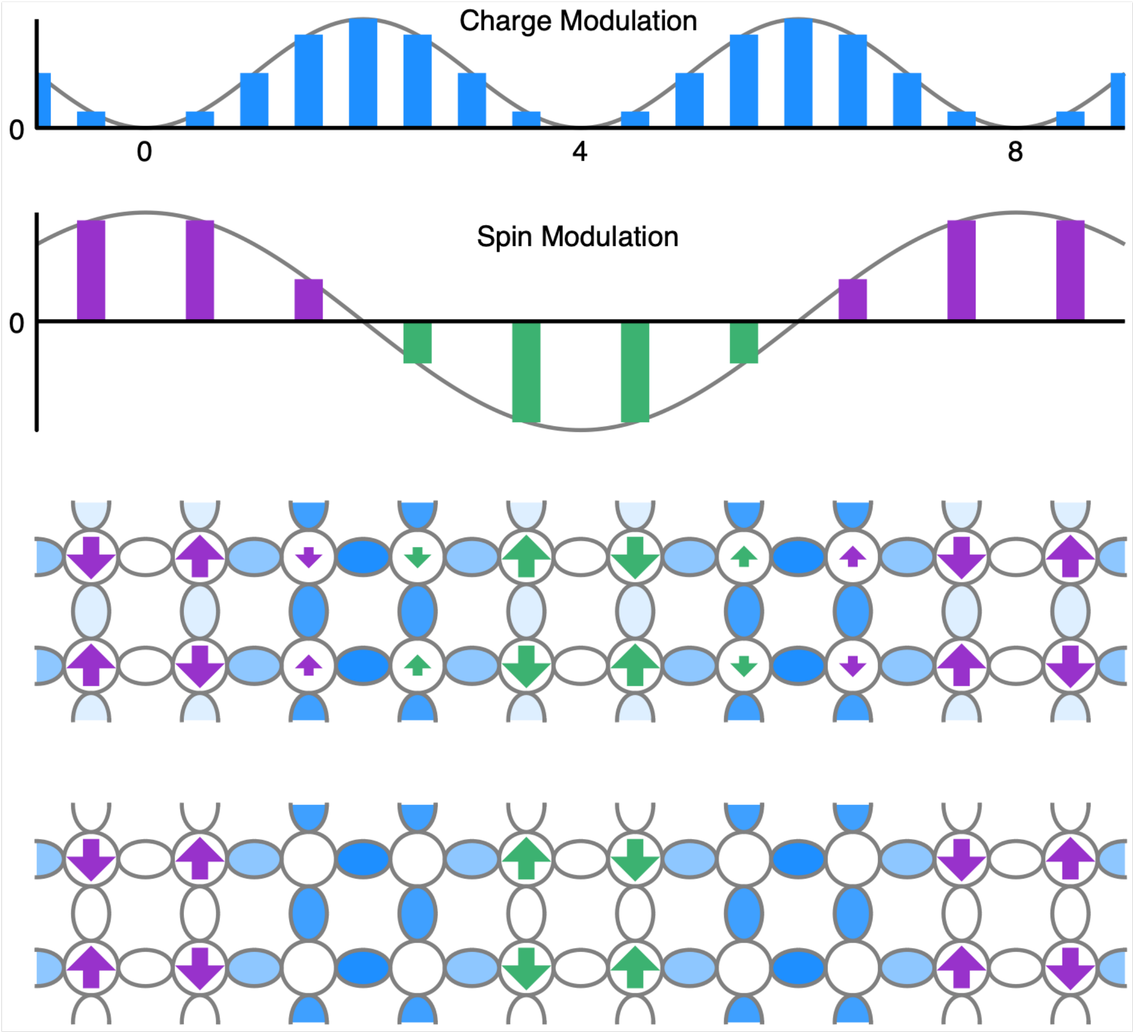
What is the pairing mechanism in the cuprate superconductors?

What is the nature of their normal state?

Spin and charge stripes



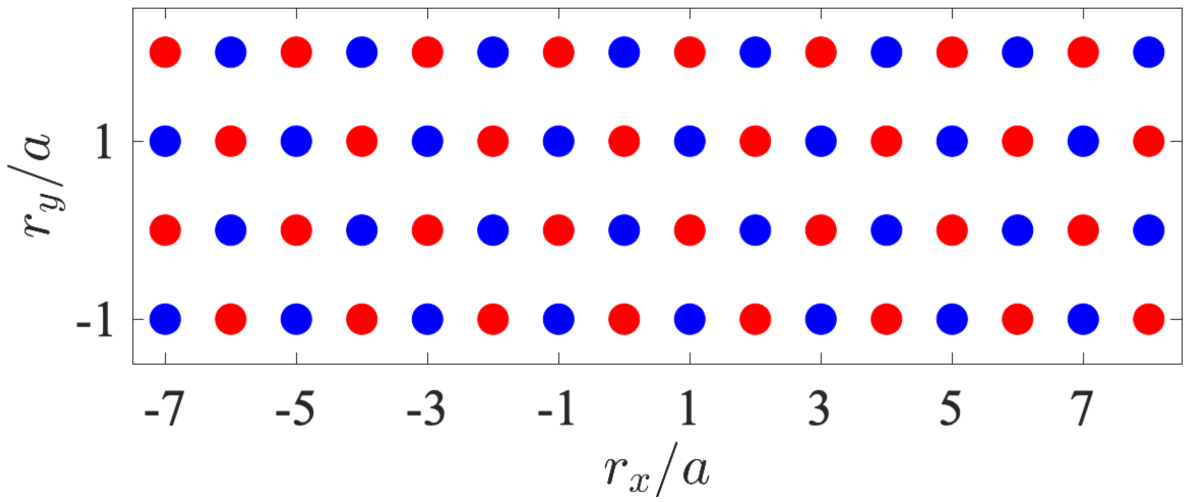
*Rajasekaran *et al.*, Science **359**, 6375 (2018).

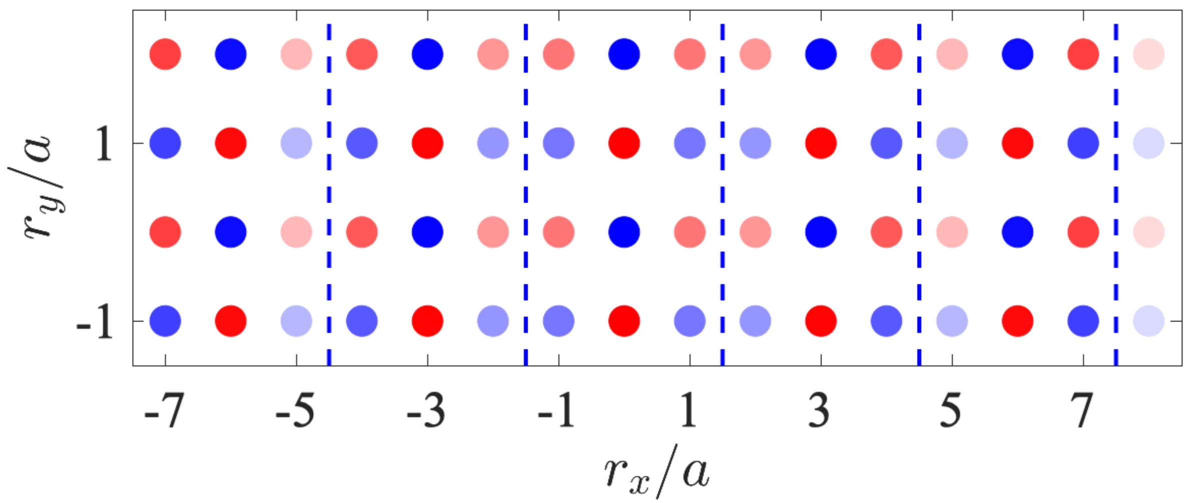


* J. Tranquada, Adv. Phys. **69**, 437-509 (2020).

A quick note on notation

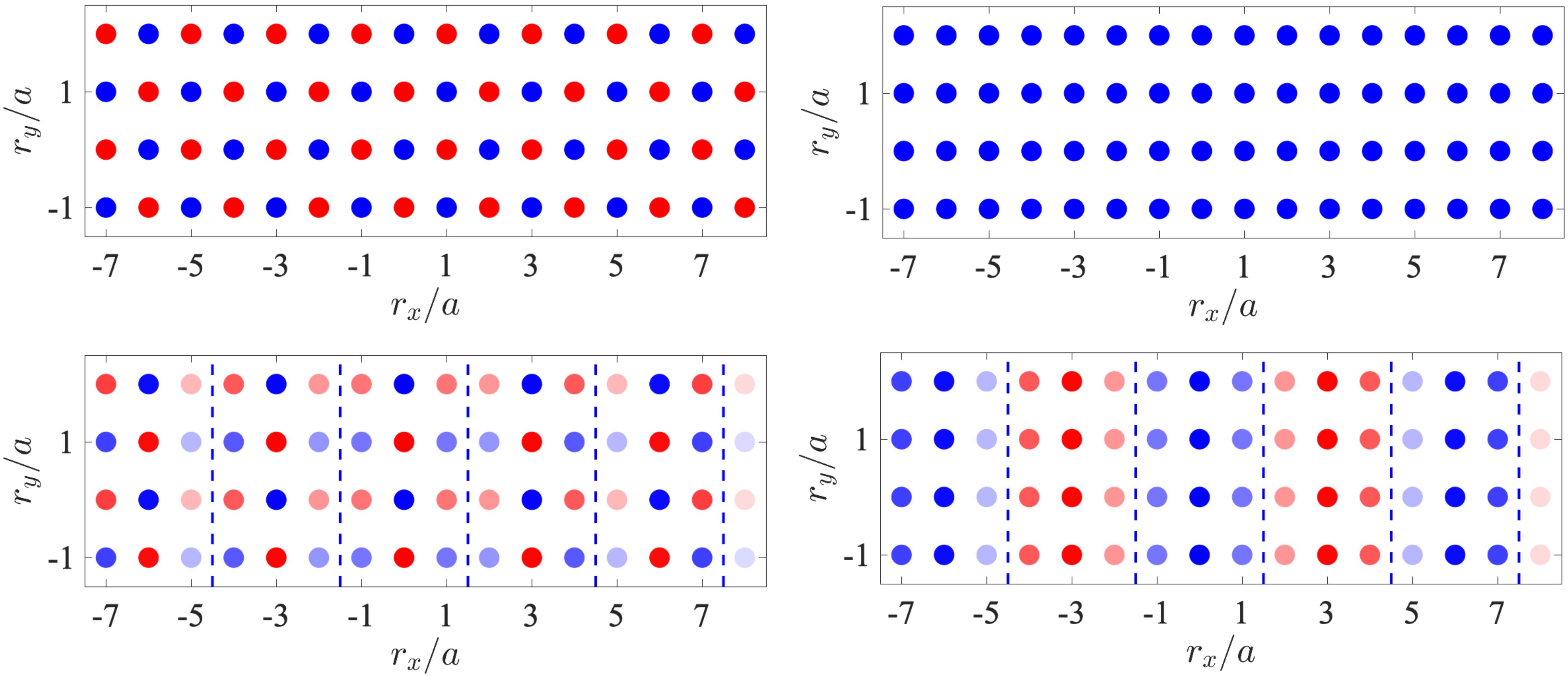
$$S(\mathbf{r}) = \langle S_z(\mathbf{r}) S_z(0) \rangle$$



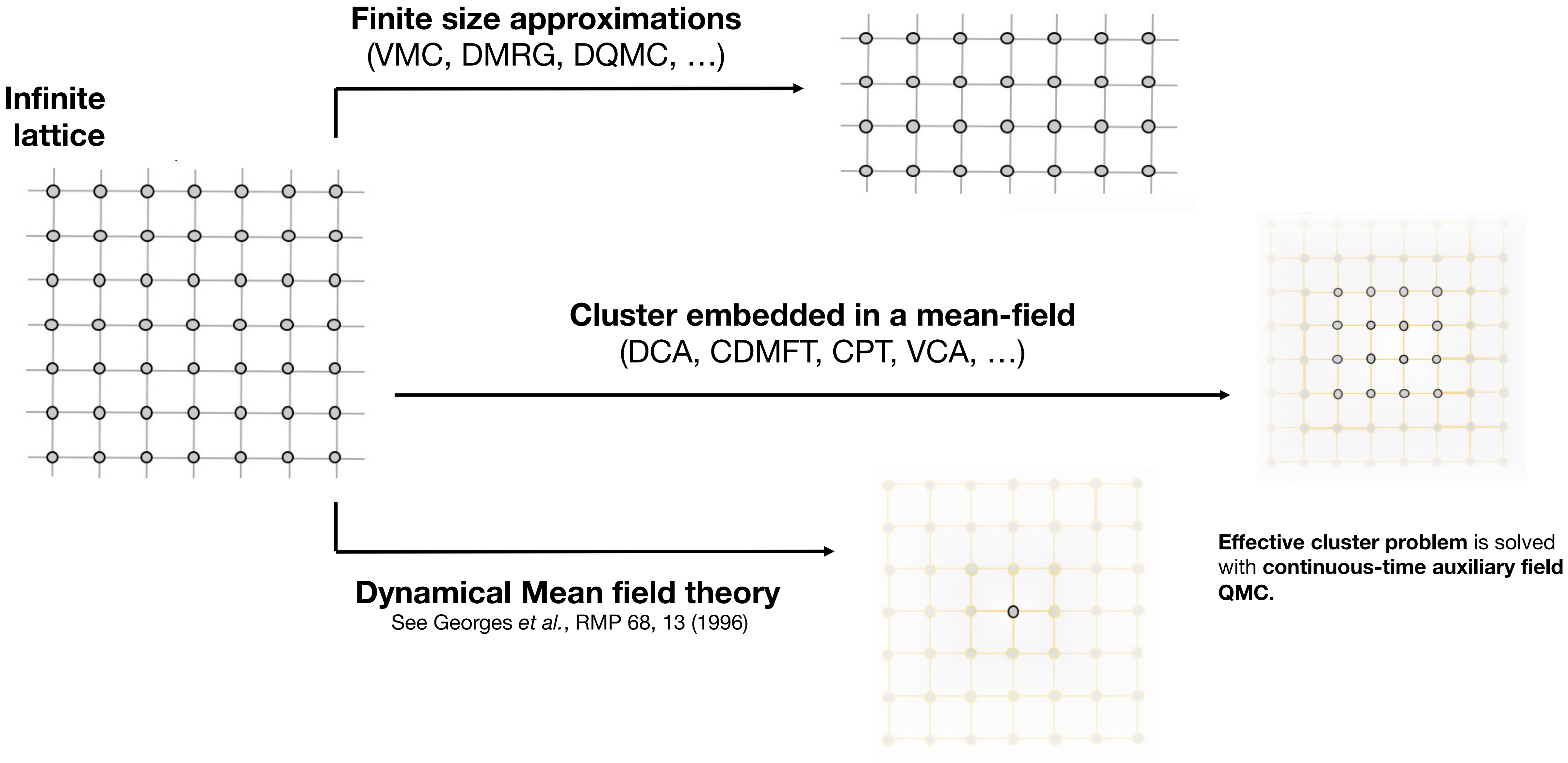


A quick note on notation

$$S(\mathbf{r}) = \langle S_z(\mathbf{r}) S_z(0) \rangle \longrightarrow S_{\text{stag}}(\mathbf{r}) = (-1)^{r_x + r_y} \langle S_z(\mathbf{r}) S_z(0) \rangle$$

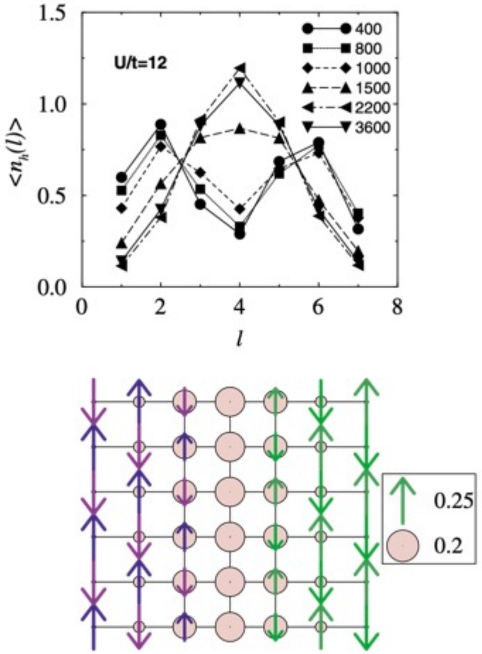


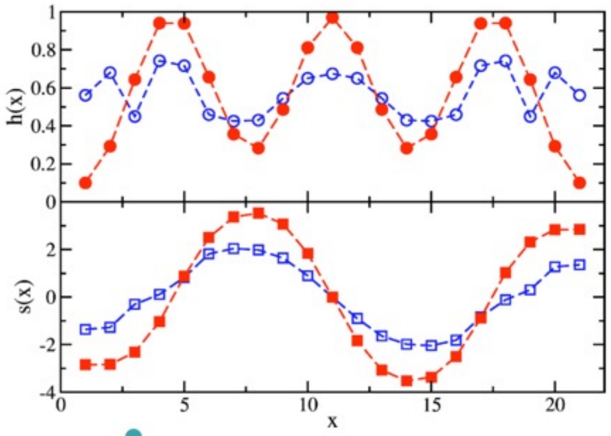
Finite cluster vs quantum embedding methods

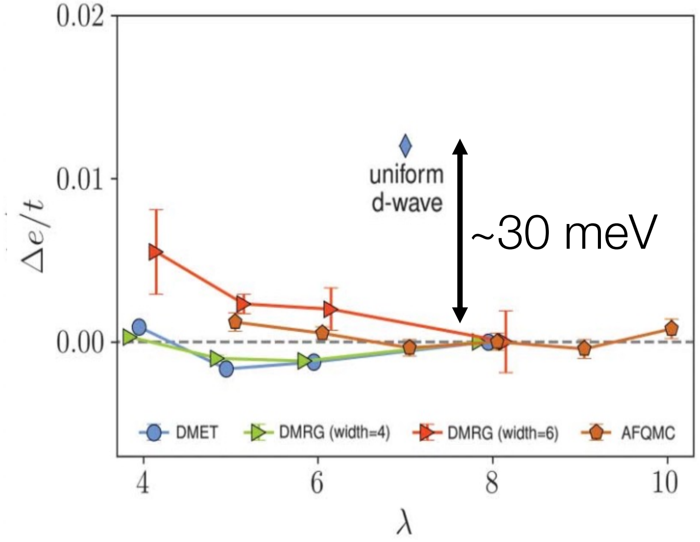


Leadership class computing facilities

Stripes in finite size cluster methods @ T = 0







2003

DMRG on

7 x 6 Hubbard ladder

White & Scalapino, PRL '03

2005

DMRG on

21 x 6 Hubbard ladder

Hager et al., PRB '05

⇒ Stripes

2018

DMRG (and other methods)

on up to

64 x 7 Hubbard ladders

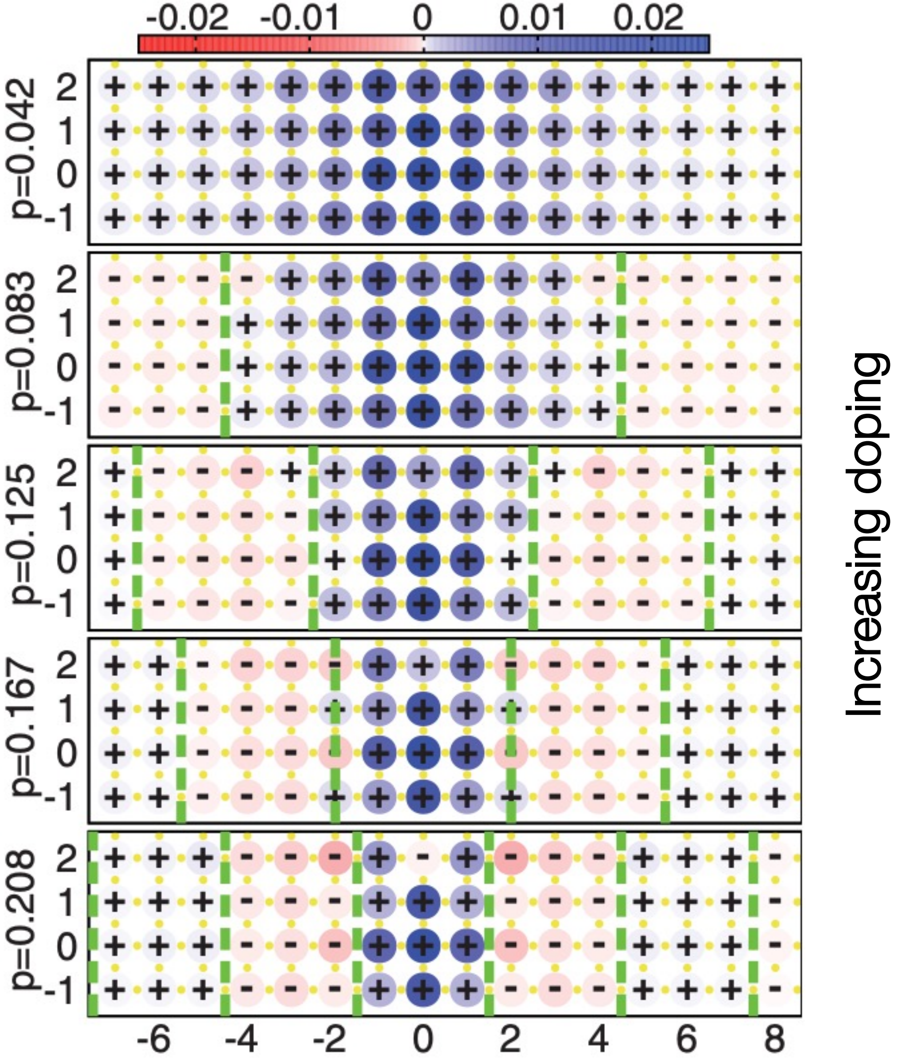
Zheng et al., Science '17

⇒ Filled charge & spin stripes

(t'=0)

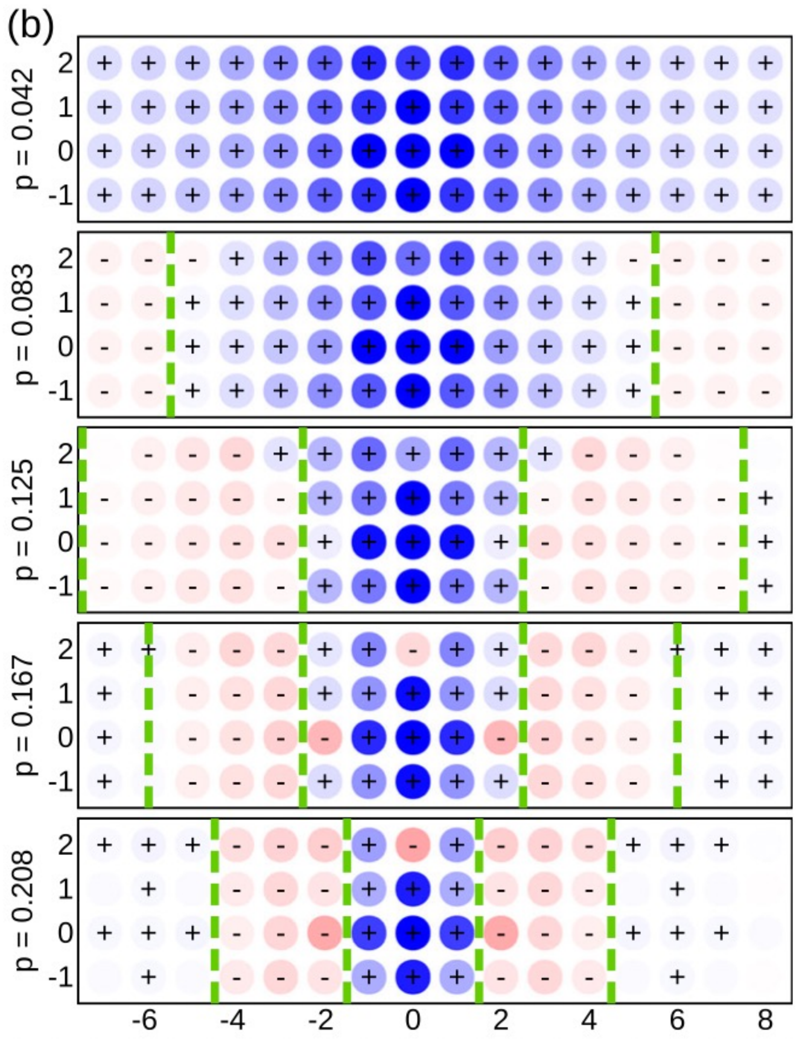
Stripes in finite size cluster methods @ finite T

three-band Hubbard



*E. Huang, … SJ, *et al.*,
Science **358**, 1161 (2017).

single-band Hubbard



*E. Huang *et al.*,
Quant. Mat. **3**, 22 (2018).

PHYSICAL REVIEW X **10,** 031016 (2020)

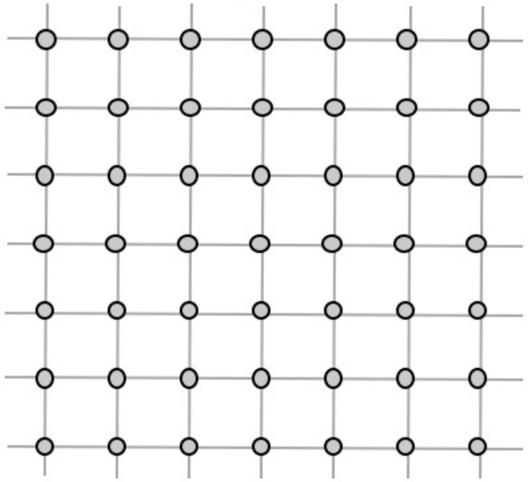
Absence of Superconductivity in the Pure Two-Dimensional Hubbard Model

Mingpu Qin,[1,2,*] Chia-Min Chung,[3,4,*] Hao Shi,[5] Ettore Vitali,[6,2] Claudius Hubig,[7]
Ulrich Schollwöck,[3,4] Steven R. White,[8] and Shiwei Zhang[5,2]

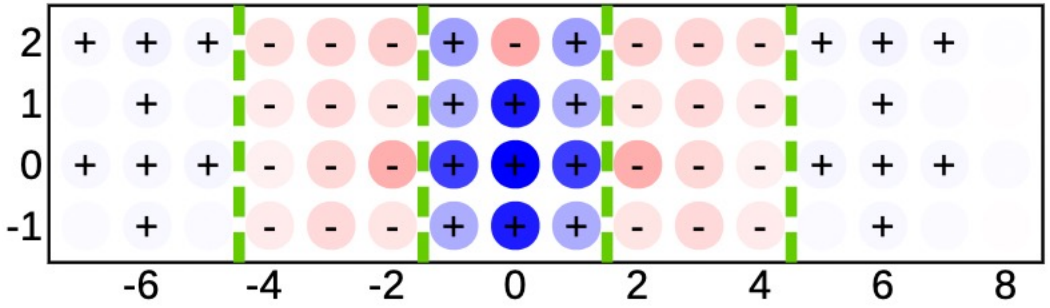
(Simons Collaboration on the Many-Electron Problem)

Finite cluster vs quantum embedding methods

Infinite lattice



Finite size approximations
(VMC, DMRG, DQMC, …)

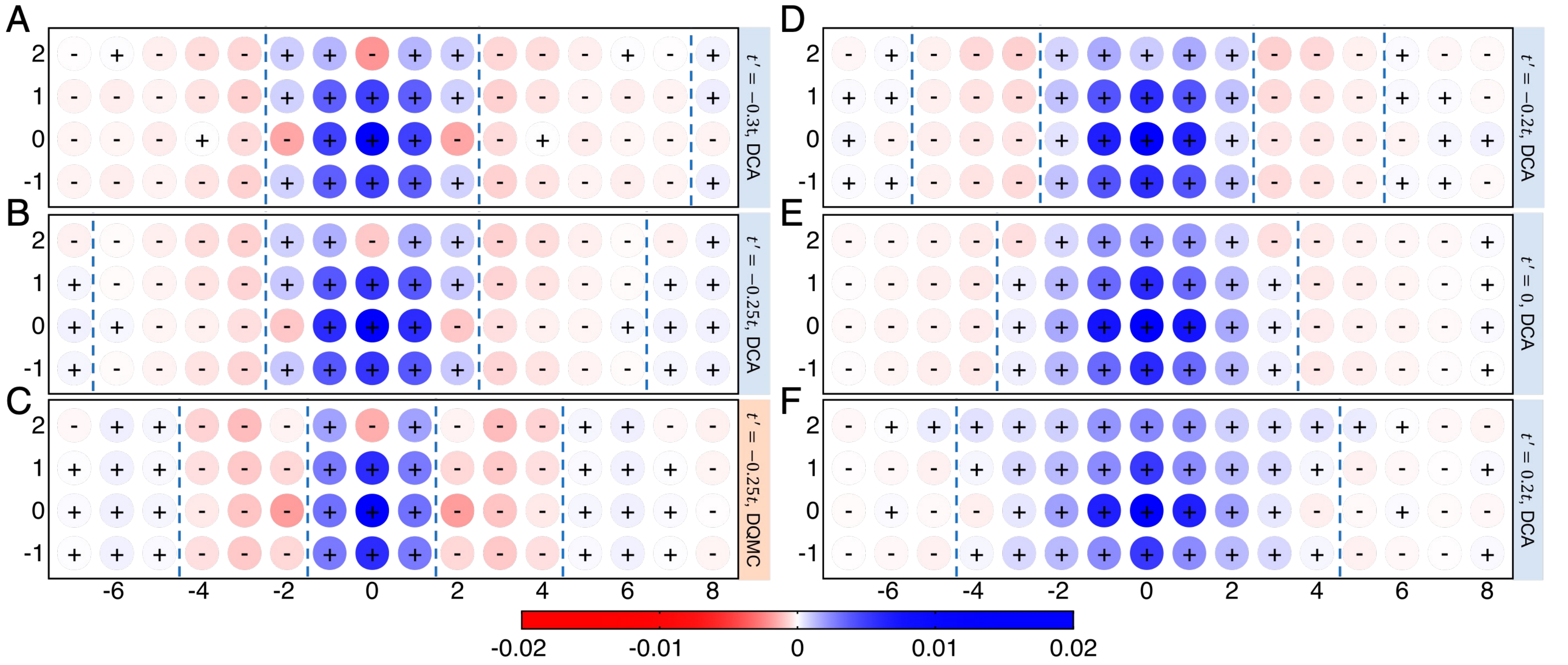


Cluster embedded in a mean-field
(DCA, CDMFT, CPT, VCA, …)

DCA spin stripe correlations in 16 x 4 clusters

Time-integrated (zero frequency) staggered spin correlations:

$$S_{\mathrm{stag}}(\mathbf{r}, \omega = 0) = (-1)^{(r_x + r_y)} \frac{1}{N} \sum_{\mathbf{i}} \int_0^{\beta} \langle \hat{S}^z_{\mathbf{r}+\mathbf{i}}(\tau) \hat{S}^z_{\mathbf{i}}(0) \rangle d\tau$$



$$U/t = 6$$

$$\beta t = 6$$

$$\langle n \rangle = 0.8$$

DCA spin stripes

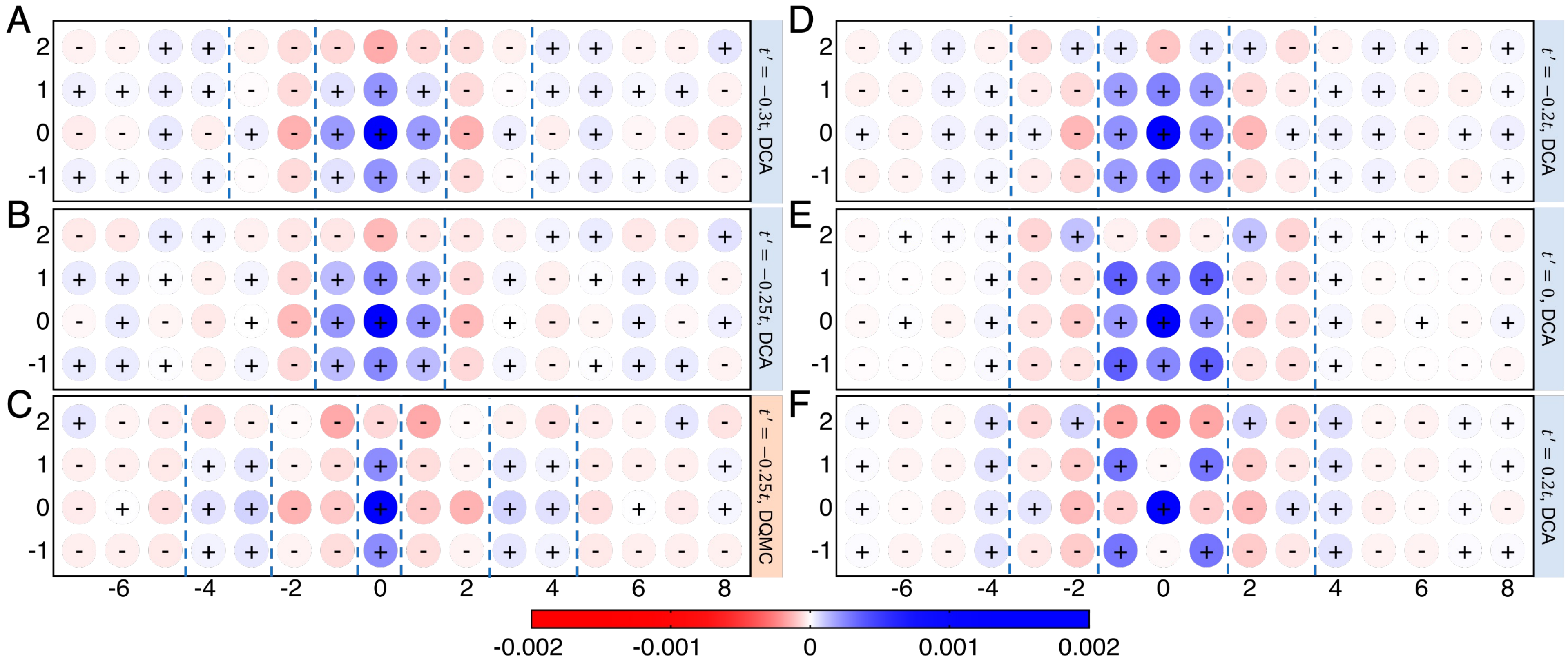
- Clearly present at $\langle n \rangle = 0.8$
- Note as strong as in DQMC
- Periodicity and strength depend on t'/t

*P. Mai *et al.*, PNAS **119,** e2112806119 (2022).

DCA charge stripe correlations in 16 x 4 clusters

Time-integrated (zero frequency) density-density correlations:

$$N(\mathbf{r}, \omega = 0) = \frac{1}{N} \int_0^\beta \left[\langle n_{\mathbf{r}+\mathbf{i}}(\tau) n_{\mathbf{i}}(0) \rangle - \langle n_{\mathbf{r}+\mathbf{i}}(\tau) \rangle \langle n_{\mathbf{i}}(0) \rangle \right] d\tau.$$



$$U/t = 6$$

$$\beta t = 6$$

$$\langle n \rangle = 0.8$$

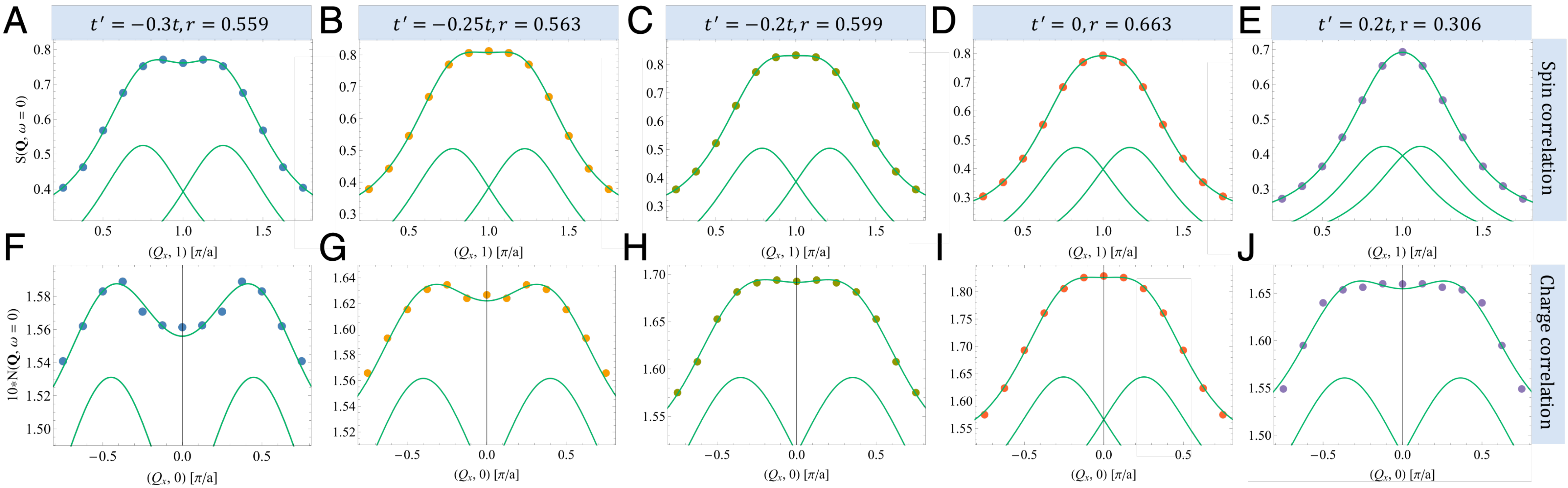
DCA charge stripes

- Clearly present at $\langle n \rangle = 0.8$.
- Less dependence on t'/t.

*P. Mai *et al.*, PNAS **119**, e2112806119 (2022).

DCA spin (top) and charge (bottom) structure factors

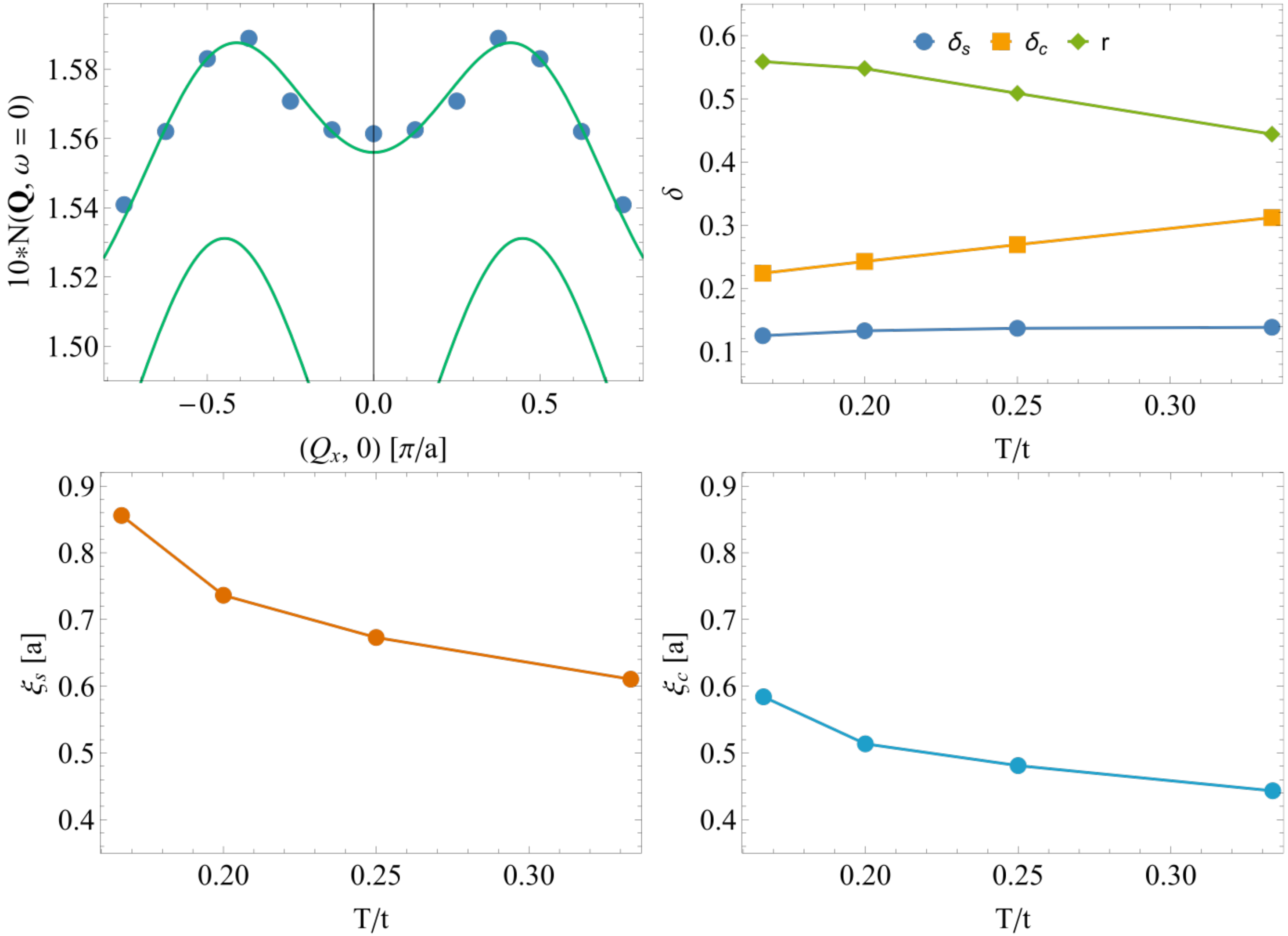
$$S(\mathbf{Q}, \omega = 0) = \frac{1}{N} \sum_{i,j} e^{\mathrm{i}\mathbf{Q}\cdot\mathbf{r}_{i,j}} S(\mathbf{r}_{i,j}, \omega = 0) \qquad N(\mathbf{Q}, \omega = 0) = \frac{1}{N} \sum_{i,j} e^{\mathrm{i}\mathbf{Q}\cdot\mathbf{r}_{i,j}} N(\mathbf{r}_{i,j}, \omega = 0)$$



*P. Mai *et al.*, PNAS **119**, e2112806119 (2022).

Evolution of the charge & spin correlations

Spin and charge commensurabilities: $\delta_c = Q_c,\ \delta_s = \pi - Q_s,\ r = \delta_s/\delta_c \approx 0.5$



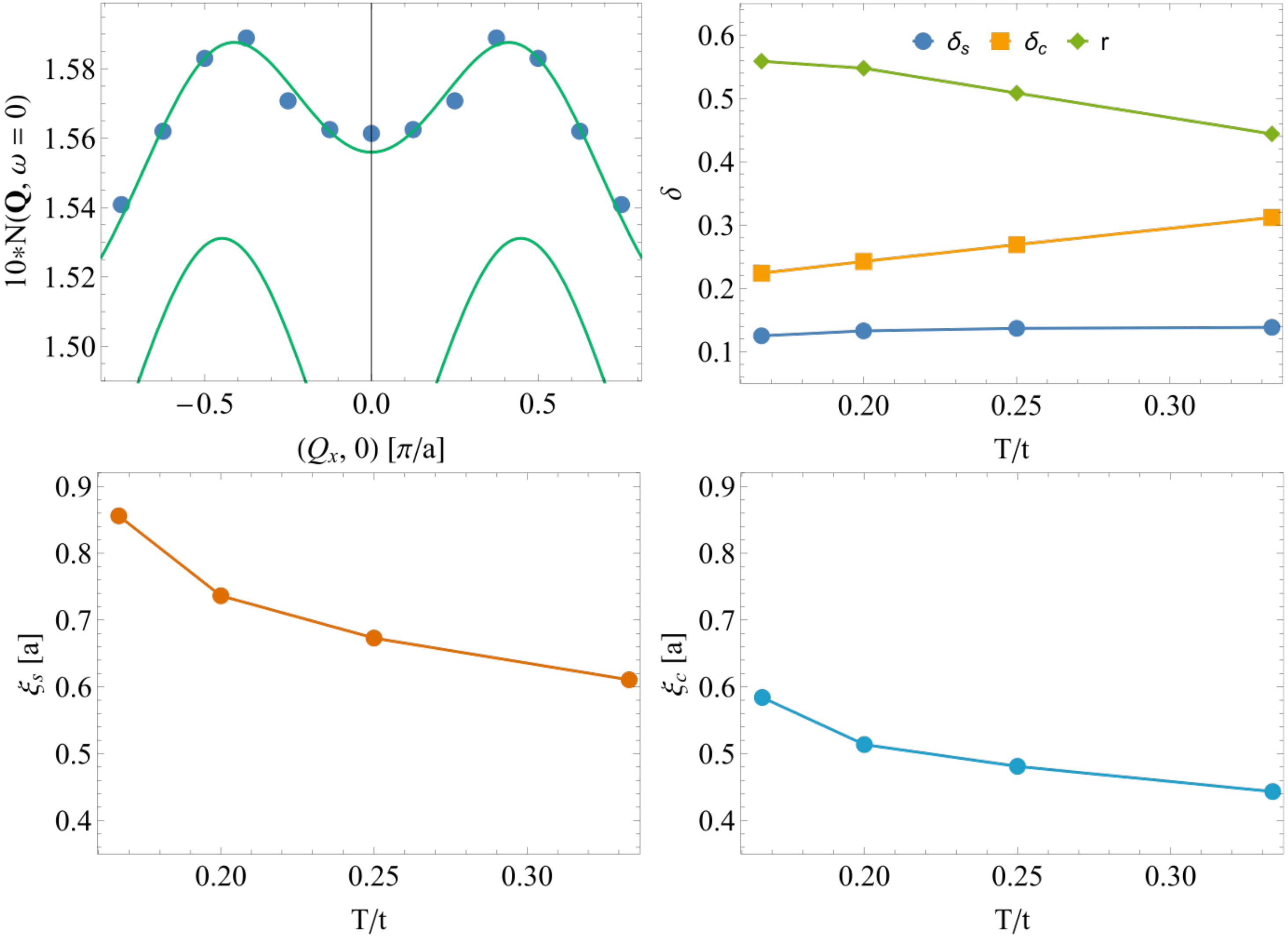
Spin & charge stripes

- Charge incommensurability locks in at twice that of the spin stripes
- Charge stripes emerge at lower energy scales as spin stripes in hole-doped case

*P. Mai et al., PNAS **119**, e2112806119 (2022).

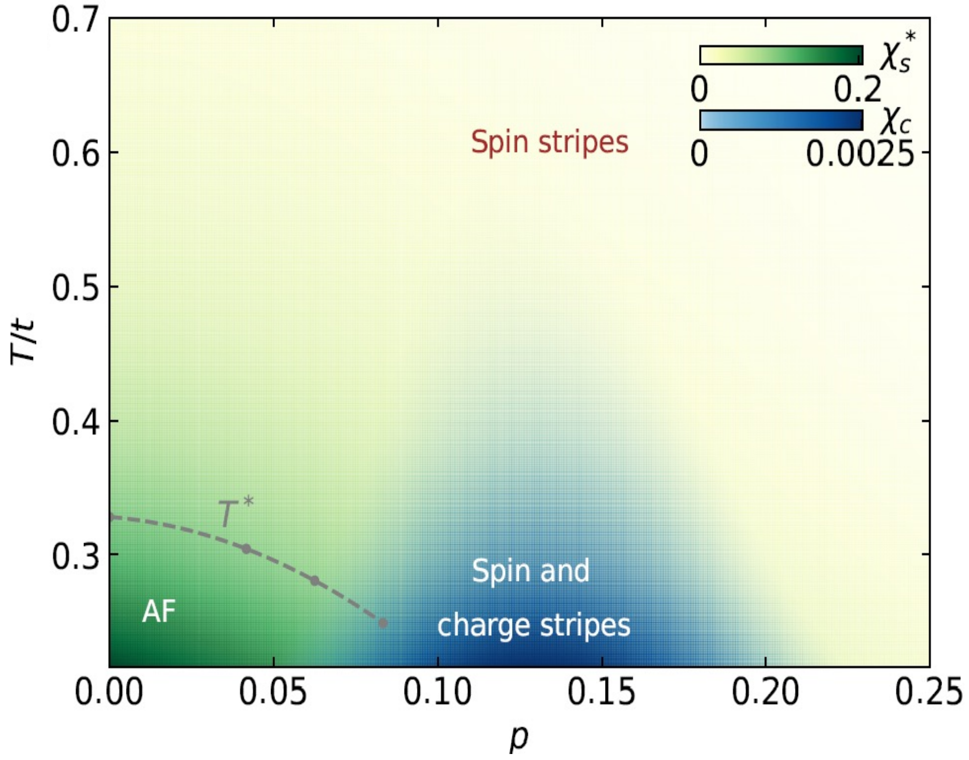
Evolution of the charge & spin correlations

Spin and charge commensurabilities: $\delta_c = Q_c,\ \delta_s = \pi - Q_s,\ r = \delta_s/\delta_c \approx 0.5$



*P. Mai *et al.*, PNAS **119**, e2112806119 (2022).

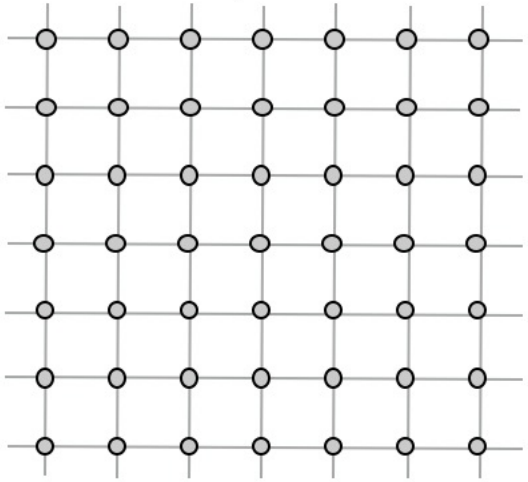
DQMC (finite T)



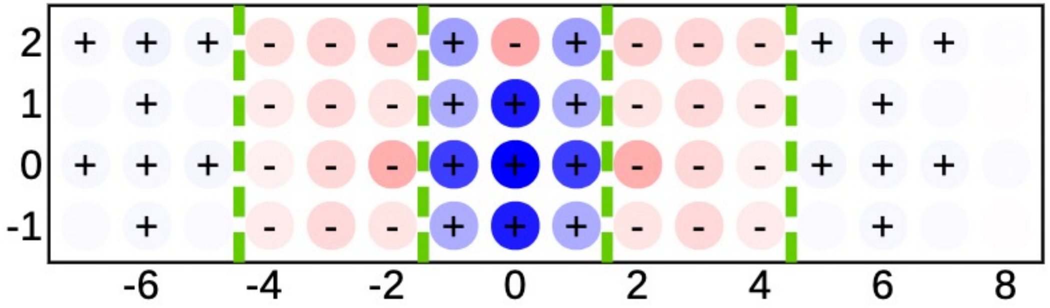
*E. Huang, … **SJ**, et al., arXiv:2202.08845 (2022).

Finite cluster vs quantum embedding methods

Infinite lattice



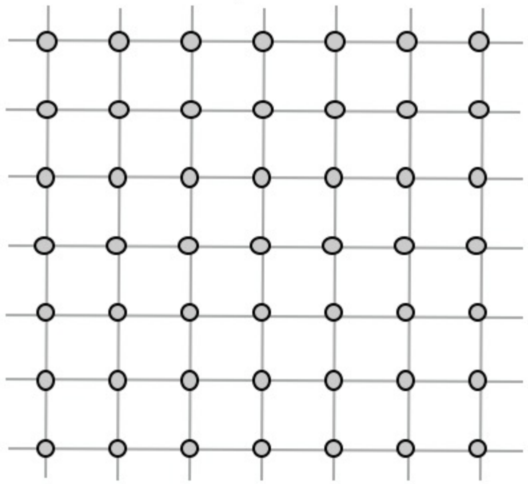
Finite size approximations
(VMC, DMRG, DQMC, …)



Cluster embedded in a mean-field
(DCA, CDMFT, CPT, VCA, …)

Finite cluster vs quantum embedding methods

Infinite lattice



Finite size approximations
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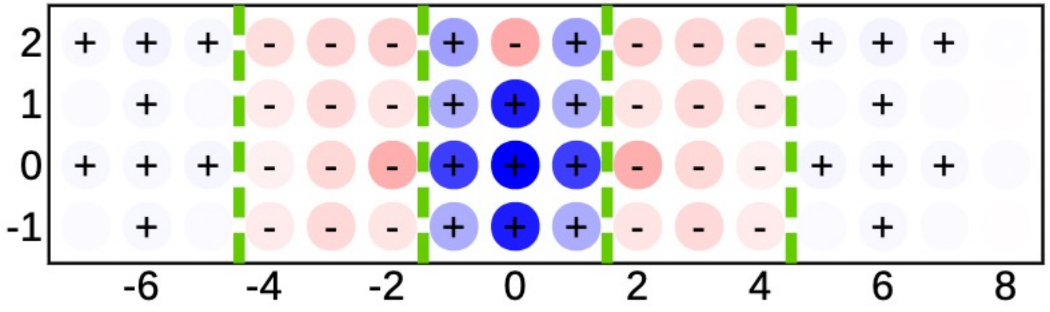

Cluster embedded in a mean-field
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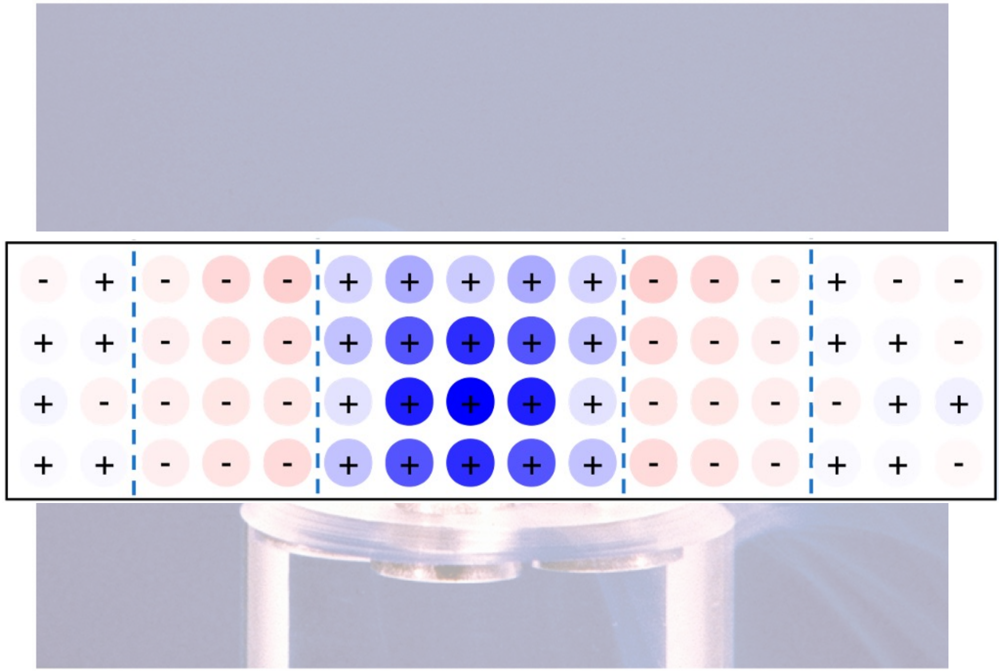
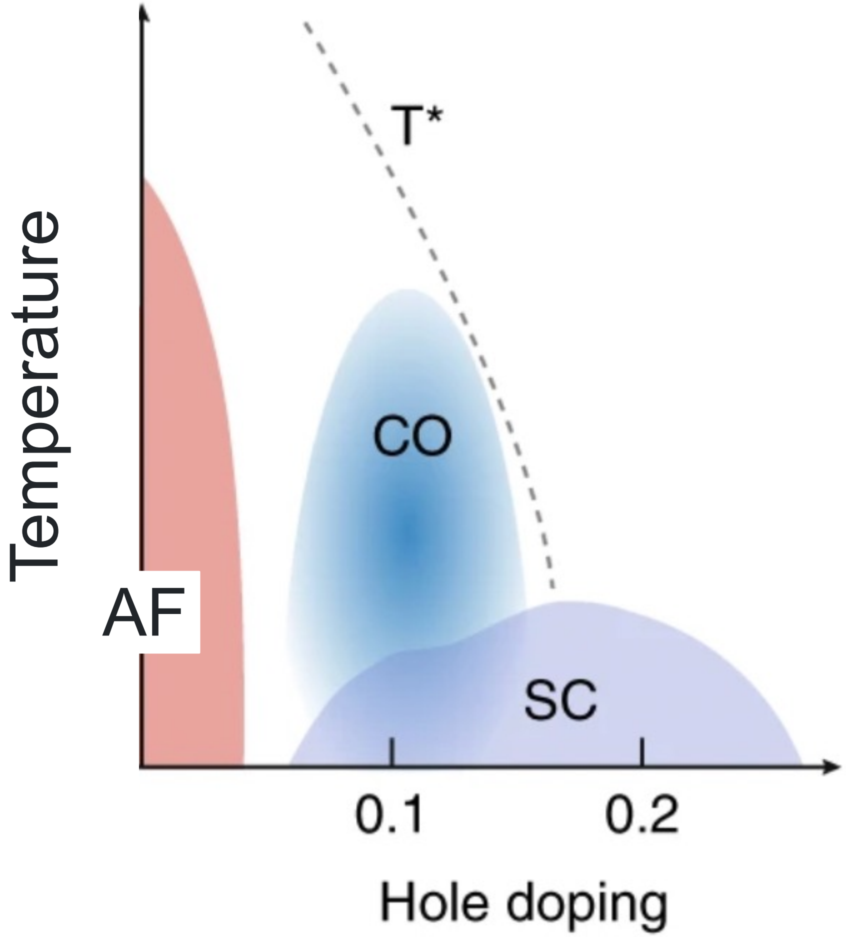
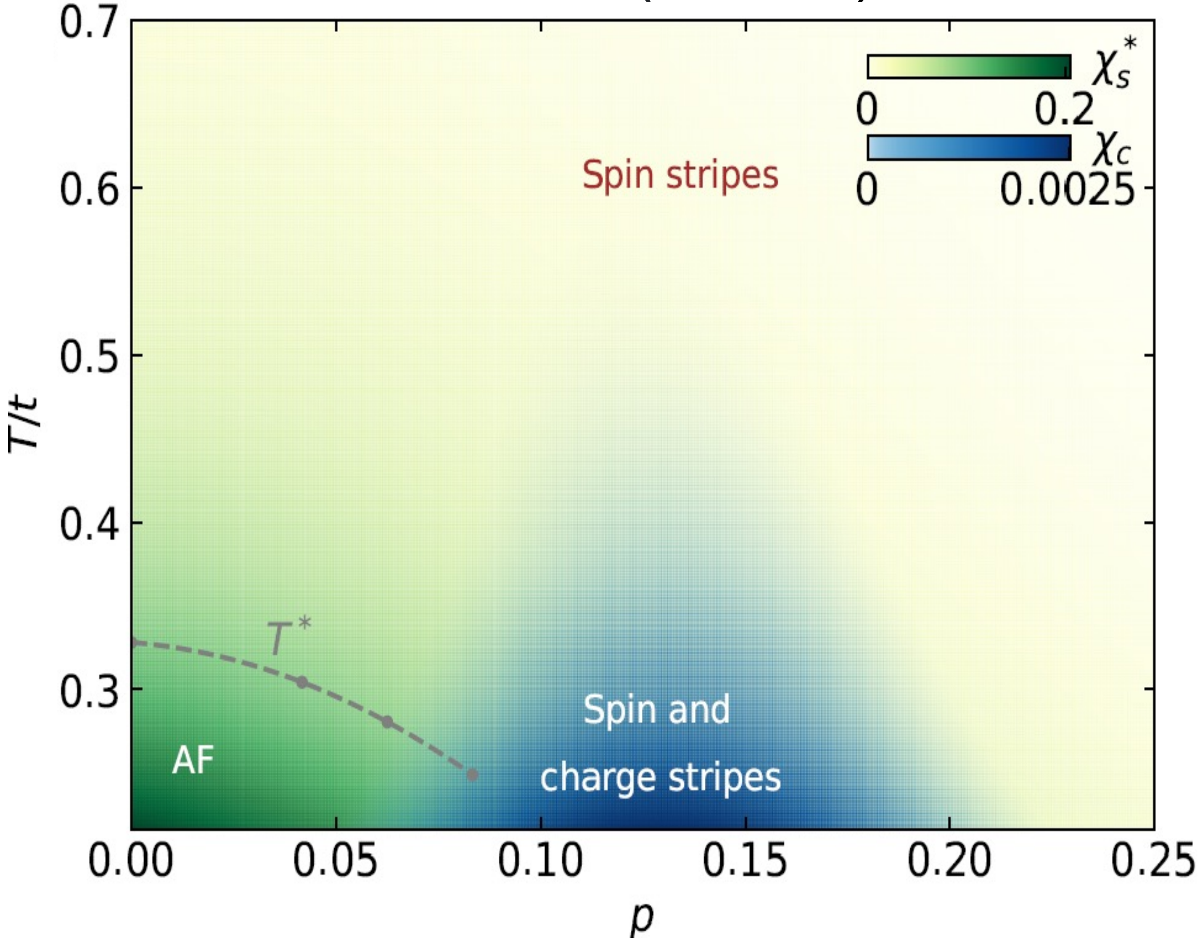


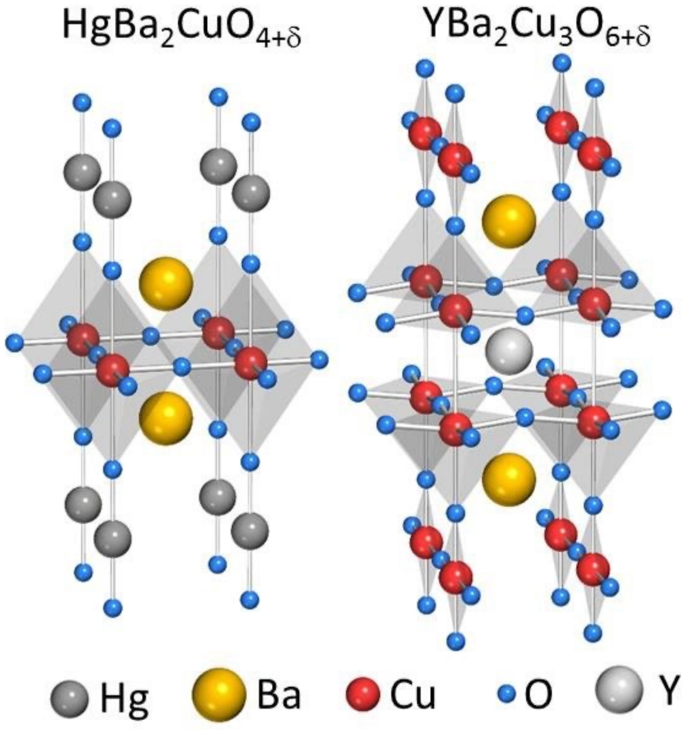
Which comes first, the spin or the charge?



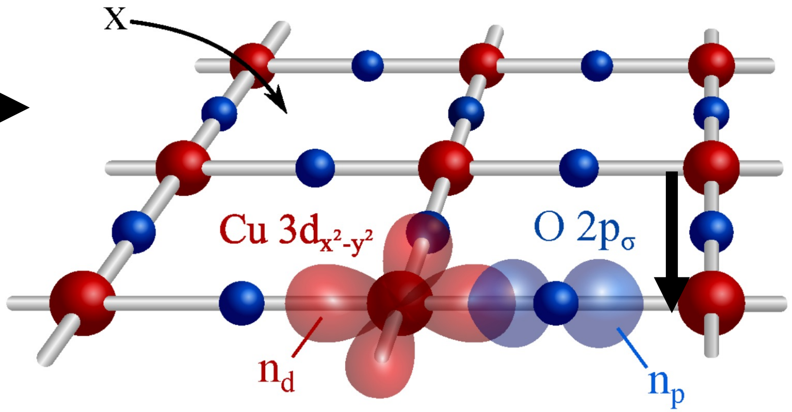
DQMC (finite T)



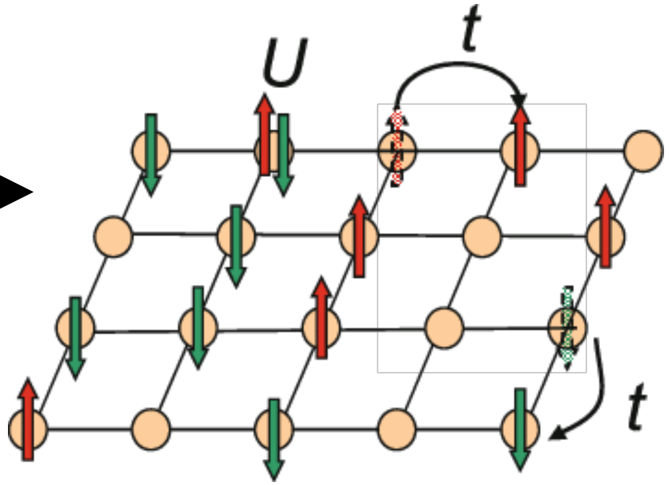
*E. Huang, … **SJ**, et al., arXiv:2202.08845 (2022).



Isolate the CuO_2 plane

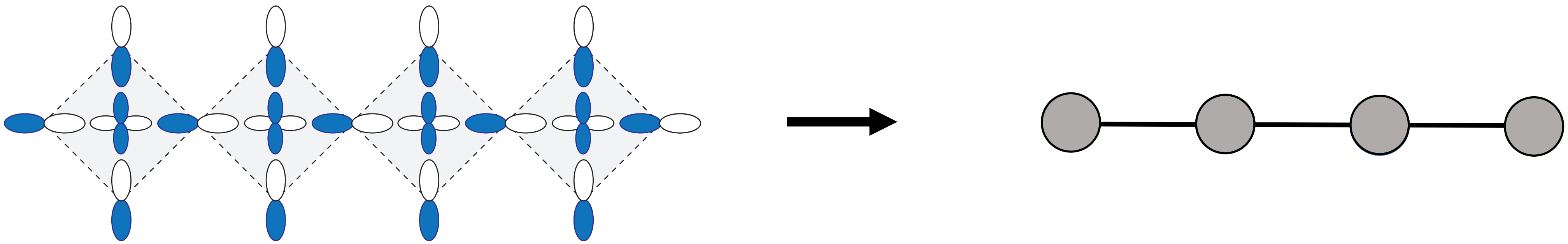


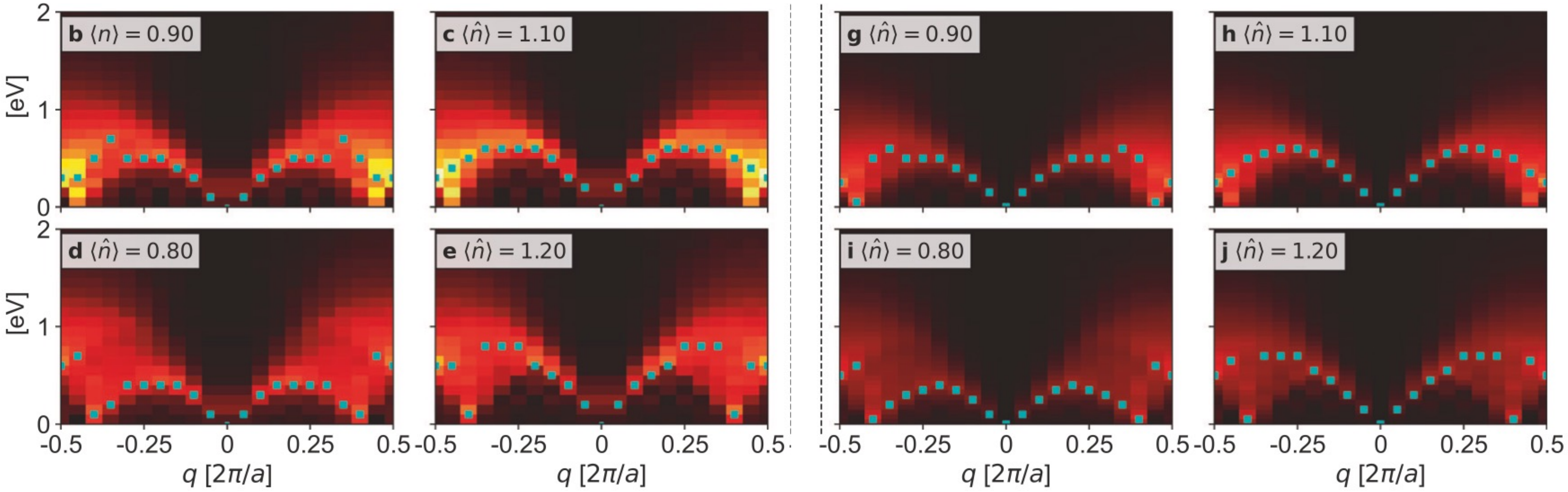
“Combine” the Cu and O orbitals



Did we lose something along the way?

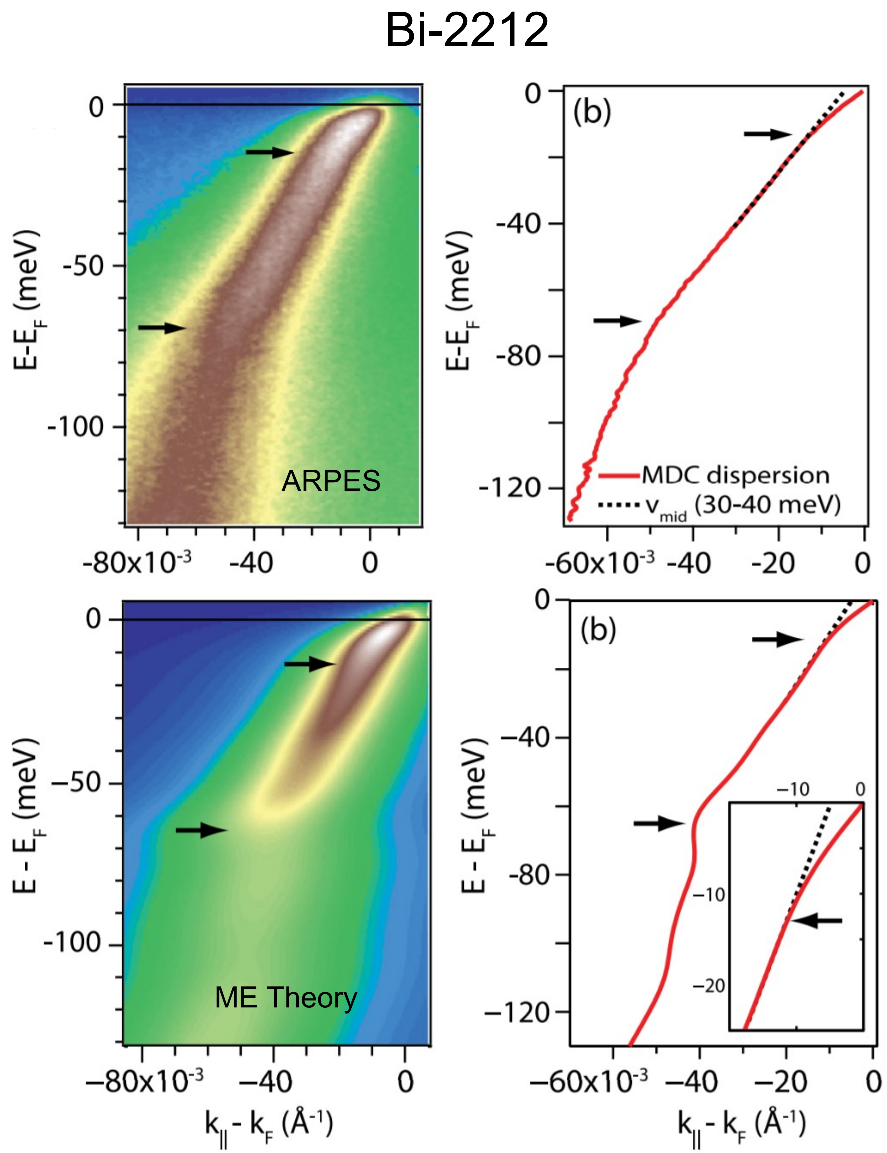
Spin dynamics of Sr_2CuO_3, a cuprate spin chain





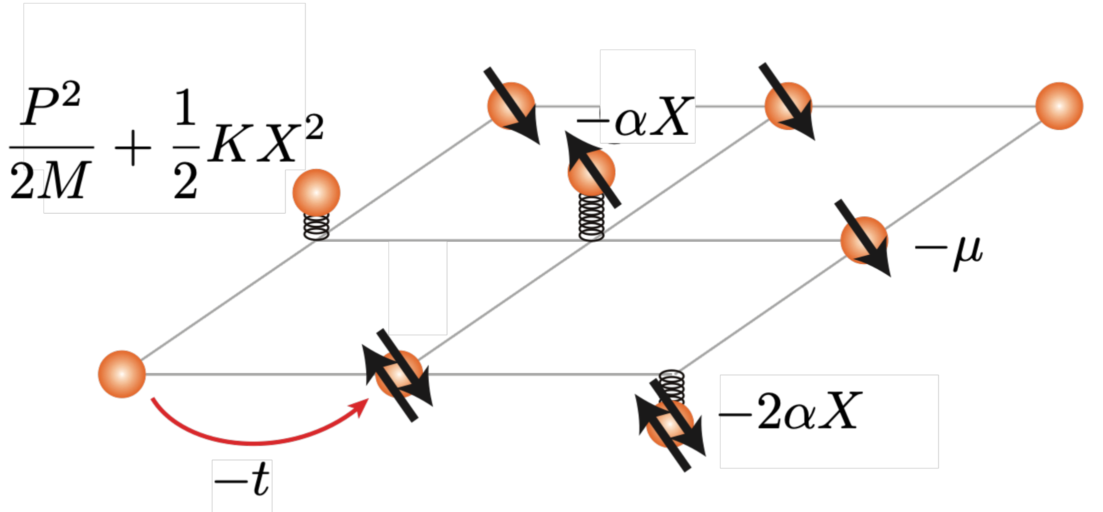
*S. Li et al., Commun. Phys. **4**, 217 (2021).

Evidence for strong electron-phonon coupling



*S. Johnston *et al.* PRL **108**, 166404 (2012).

Single-band Hubbard-Holstein model:

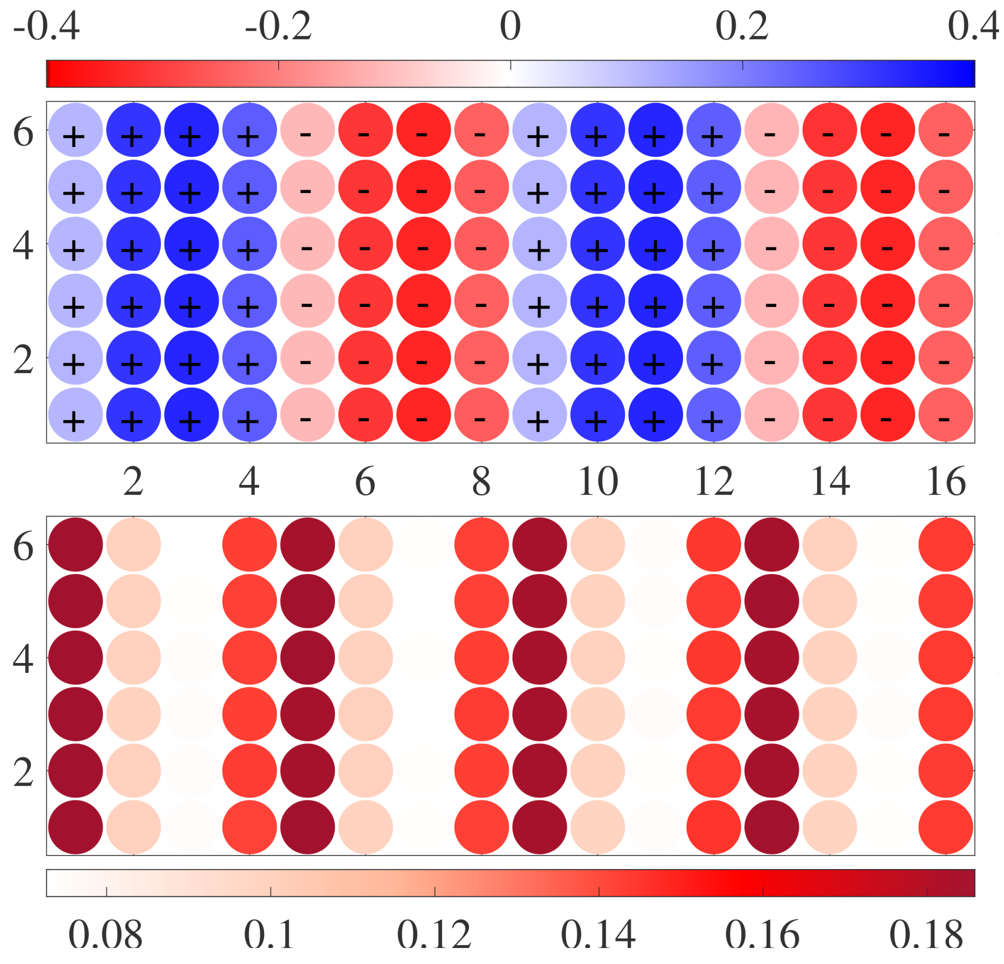


Variational Monte Carlo (VMC):

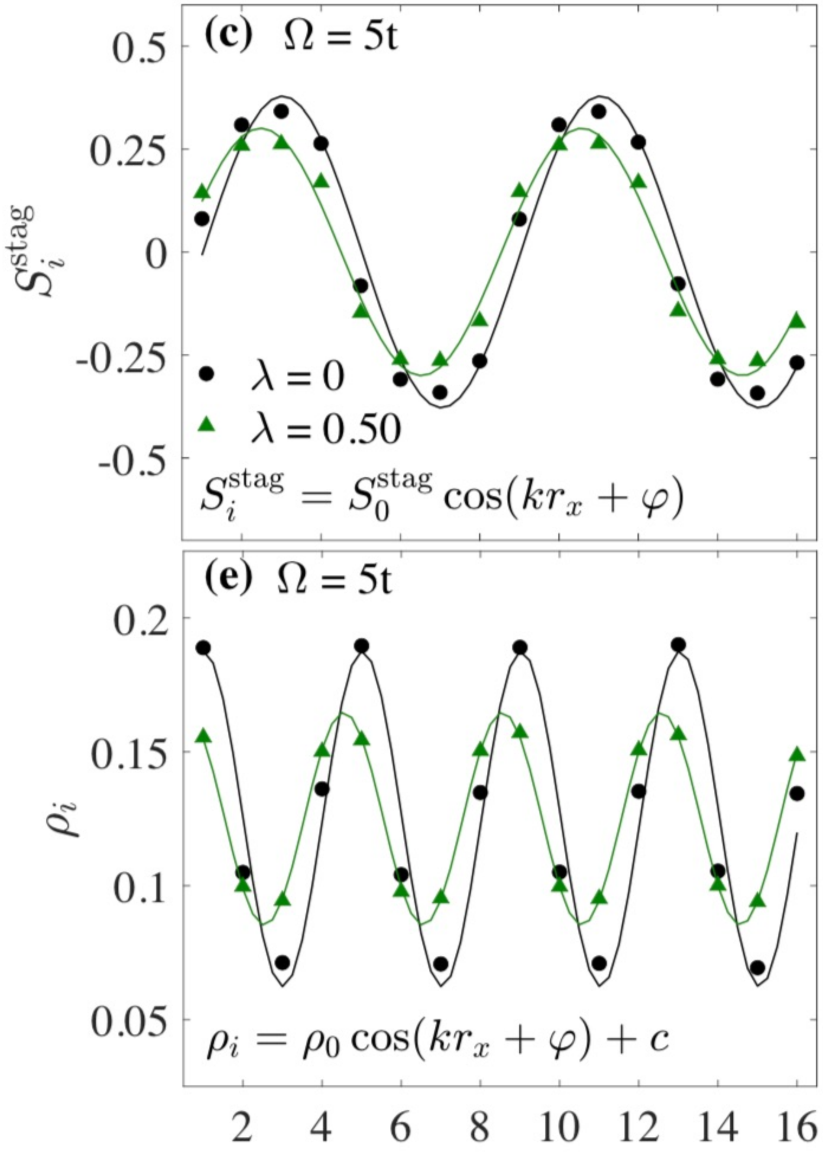
- Zero temperature method [see S. Karakuzu *et al.*, PRB **96**, 205145 (2017)]
- Markov chain Monte Carlo to optimize $\psi(\boldsymbol{r}_1, \ldots, \boldsymbol{r}_N)$
- Solved on 16 x 6 clusters

Variational Monte Carlo Results

$t'/t = -0.25,\ U = 8t,\ \langle n \rangle = 0.875, \lambda = g^2/(4t\Omega) = 0$

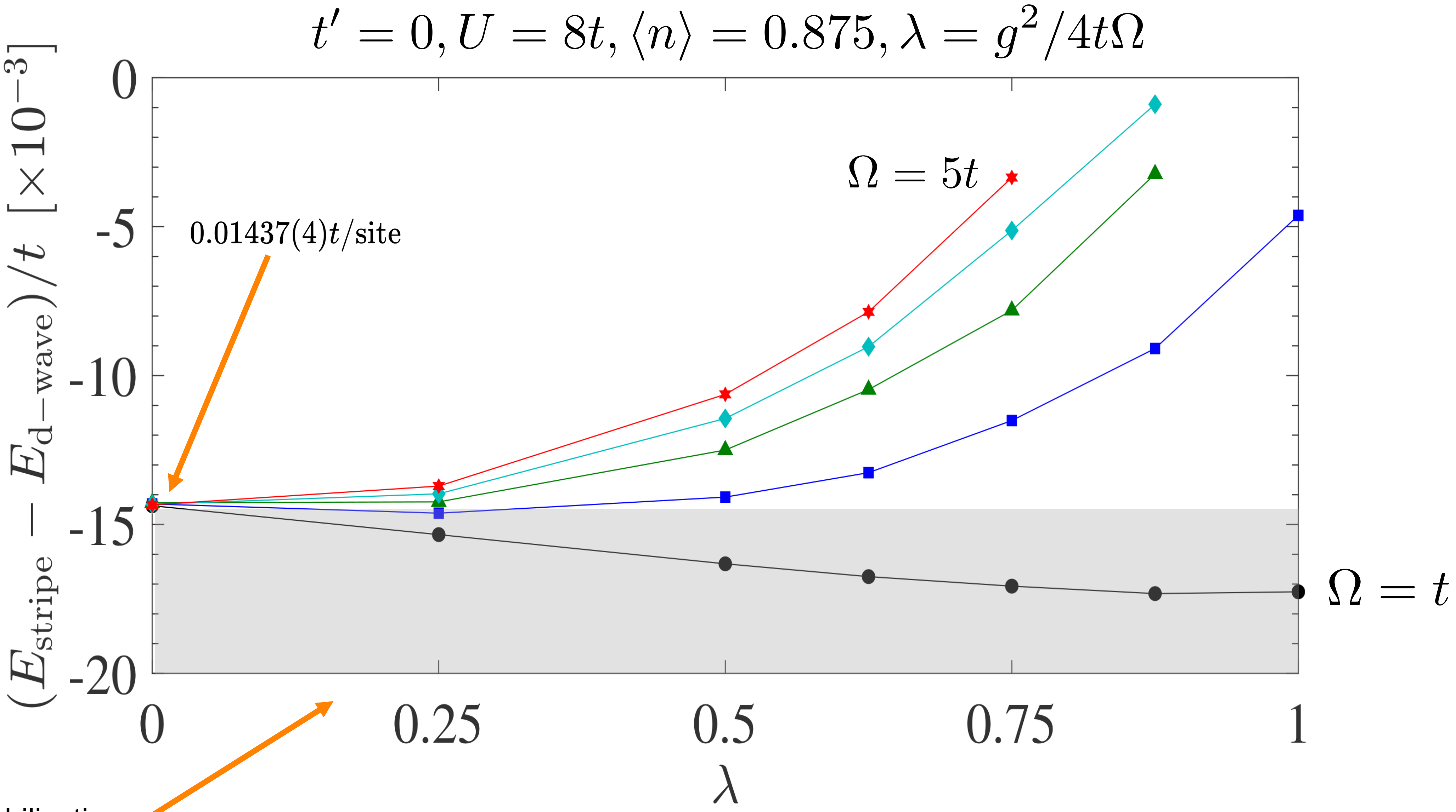


$\Omega = 5t$



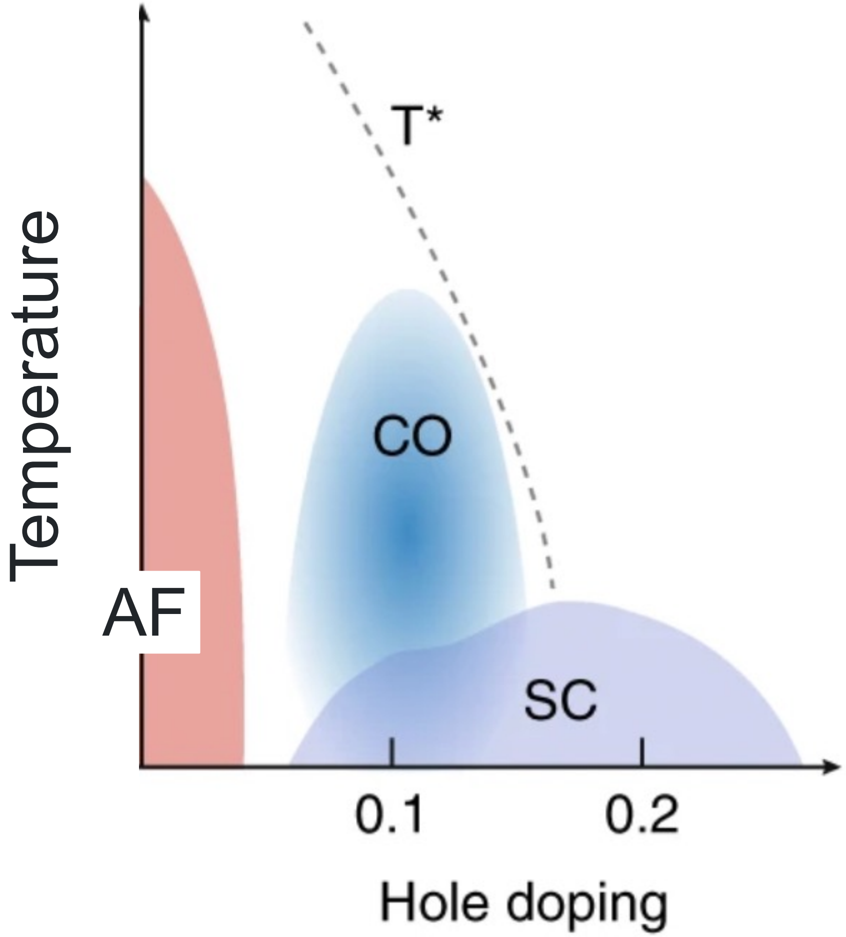
Results consistent with K. Ido *et al.* PRB **97**, 045138 (2018)

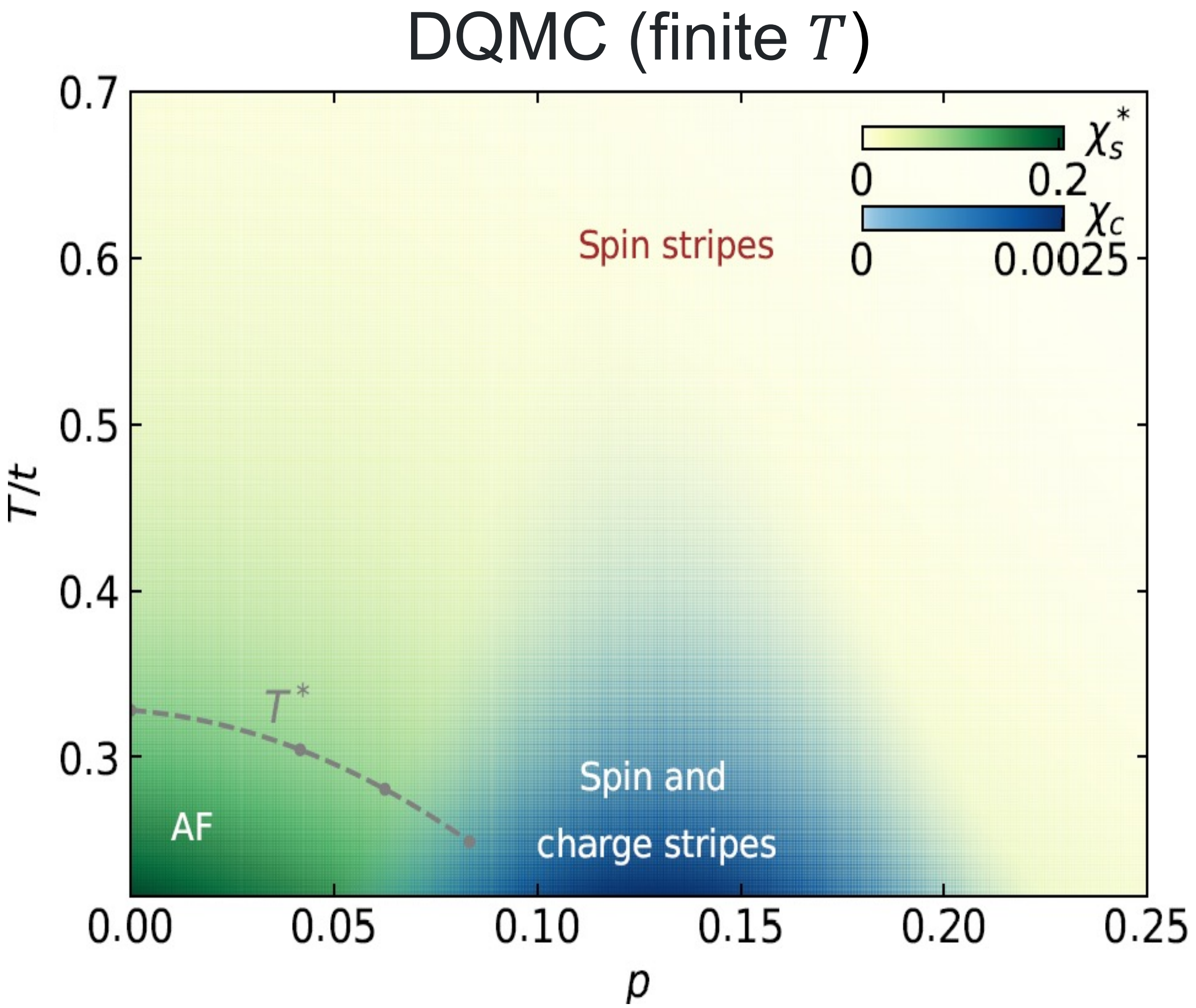
Variational Energies



Stripe Stabilization

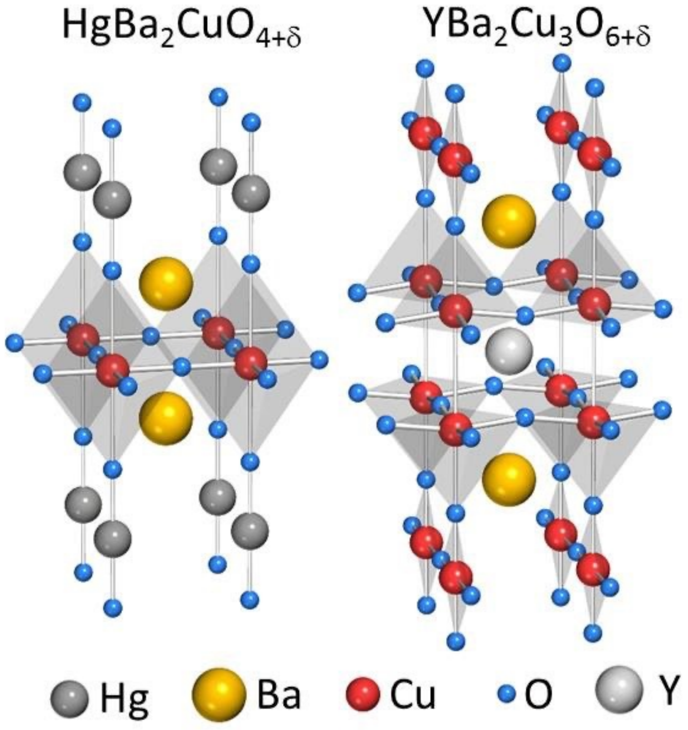
Which comes first, the spin or the charge?



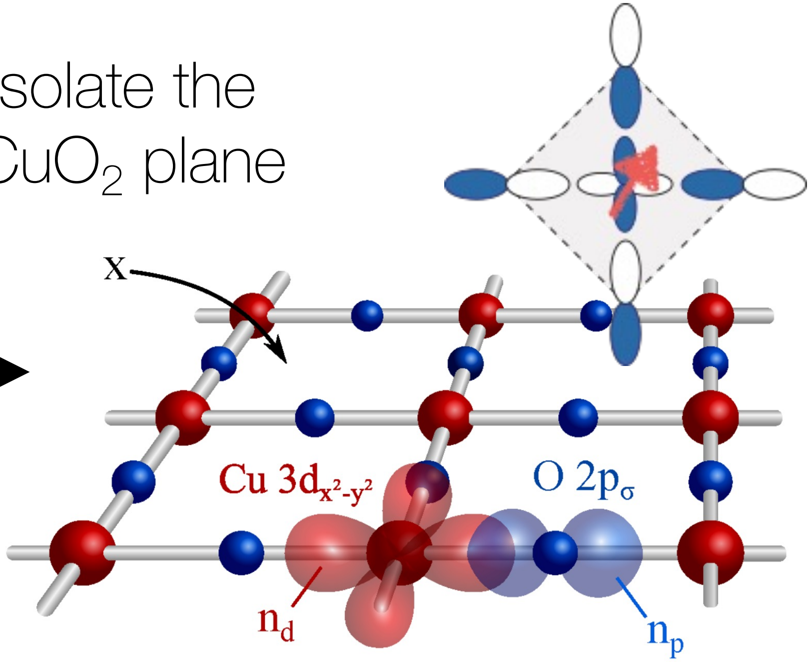


*E. Huang, … **SJ**, et al., arXiv:2202.08845 (2022).

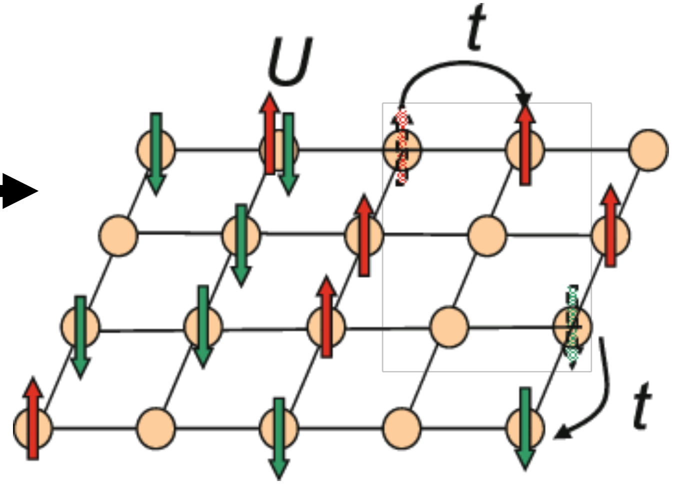
From the real material to the Hubbard model



Isolate the CuO_2 plane



remove O from the problem



$$H = -\sum_{\alpha=1}^{N_a} \frac{\hbar^2 \nabla_\alpha^2}{2M_\alpha} + \frac{1}{2}\sum_{\alpha \neq \alpha'} \frac{Z_\alpha Z_{\alpha'} e^2}{|\mathbf{R}_\alpha - \mathbf{R}_{\alpha'}|} - \sum_{\mu=1}^{N_e} \frac{\hbar^2 \nabla_\mu^2}{2m} + \frac{1}{2}\sum_{\mu \neq \mu'} \frac{e^2}{|\mathbf{r}_\mu - \mathbf{r}_{\mu'}|} - \sum_{\mu,\alpha} \frac{Z_\alpha e^2}{|\mathbf{R}_\alpha - \mathbf{r}_\mu|}$$

$$H = -\sum_{\mathbf{i},\mathbf{j},\sigma} t_{\mathbf{ij}} \left(c_{\mathbf{i}\sigma}^\dagger c_{\mathbf{j}\sigma} + \mathrm{h.c.} \right) - \mu \sum_{\mathbf{i},\sigma} n_{\mathbf{i}\sigma} + U \sum_{\mathbf{i}} n_{\mathbf{i}\uparrow} n_{\mathbf{i}\downarrow}.$$

Images from Reichardt *et al.*, Condens. Matter 3, 23 (2018).

Artificial intelligence and data science enabled predictive modeling of collective phenomena in strongly correlated quantum materials

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UTK

Shaozhi Li
UTK
(now ORNL)

Giovanni Balduzzi
ETH Zürich
(now industry)

References:

1. P. Mai *et al.*, PNAS **119,** e2112806119 (2022).
2. S. Karakuzu *et al.*, arXiv:2205.15464 (2022).
3. P. Mai *et al.*, in preparation (2022).

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