Enhanced charge excitations in electron-doped cuprates by resonant inelastic x-ray scattering

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Theory

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Experiment

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M. Fujita, T. Sasaki, K. Tsutsumi, K. Sato (Tohoku Univ.)
M. Yoshida, M. Kurooka, J. Mizuki (Kwansei Gakuin Univ.)
R. Kajimoto (J-PARC Center)
K. Ikeuchi (CROSS)
K. Yamada (KEK)
M. Minola, G. Dellea, C. Mazzoli, G. Ghiringhelli,
L. Braicovich (Politecnico di Milano)
K. Kummer (ESRF)

Outline

1. Electron-hole asymmetry in cuprates

2. Charge degree of freedom in electron-doped cuprates

Resonant inelastic x-ray scattering (RIXS)

Incoherent charge excitation t-t'-J model

Charge order

3. Q-dependent "coherent" charge excitations

Theoretical prediction t-t'-U Hubbard model

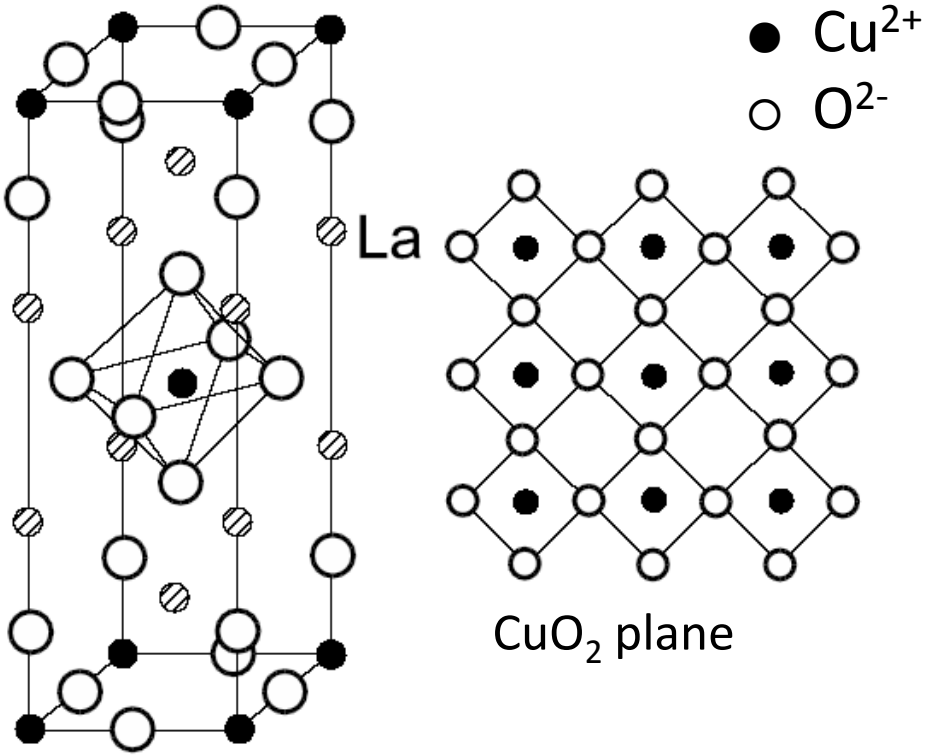
Density-matrix renormalization group (DMRG)

Low-energy charge mode, detectable by RIXS

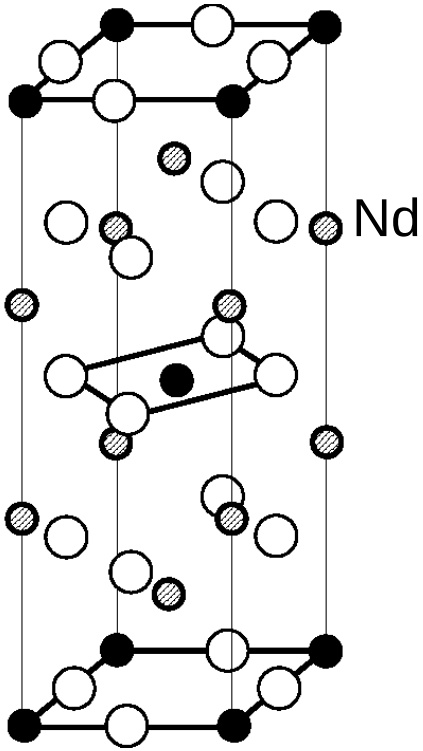
K. Ishii, M. Fujita, T. T. *et al*., Nat. Commun. **5**, 3714 (2014)
T. T., K. Tsutsui, M. Mori, S. Sota, S. Yunoki, Phys. Rev. B **92**, 014515 (2015)
T.T., J. Electro. Spector. and Related Phenome., **200**, 209 (2015)

Crystal structure of La_2CuO_4 and Nd_2CuO_4

La_2CuO_4 (hole-doping)



Nd_2CuO_4 (electron-doping)



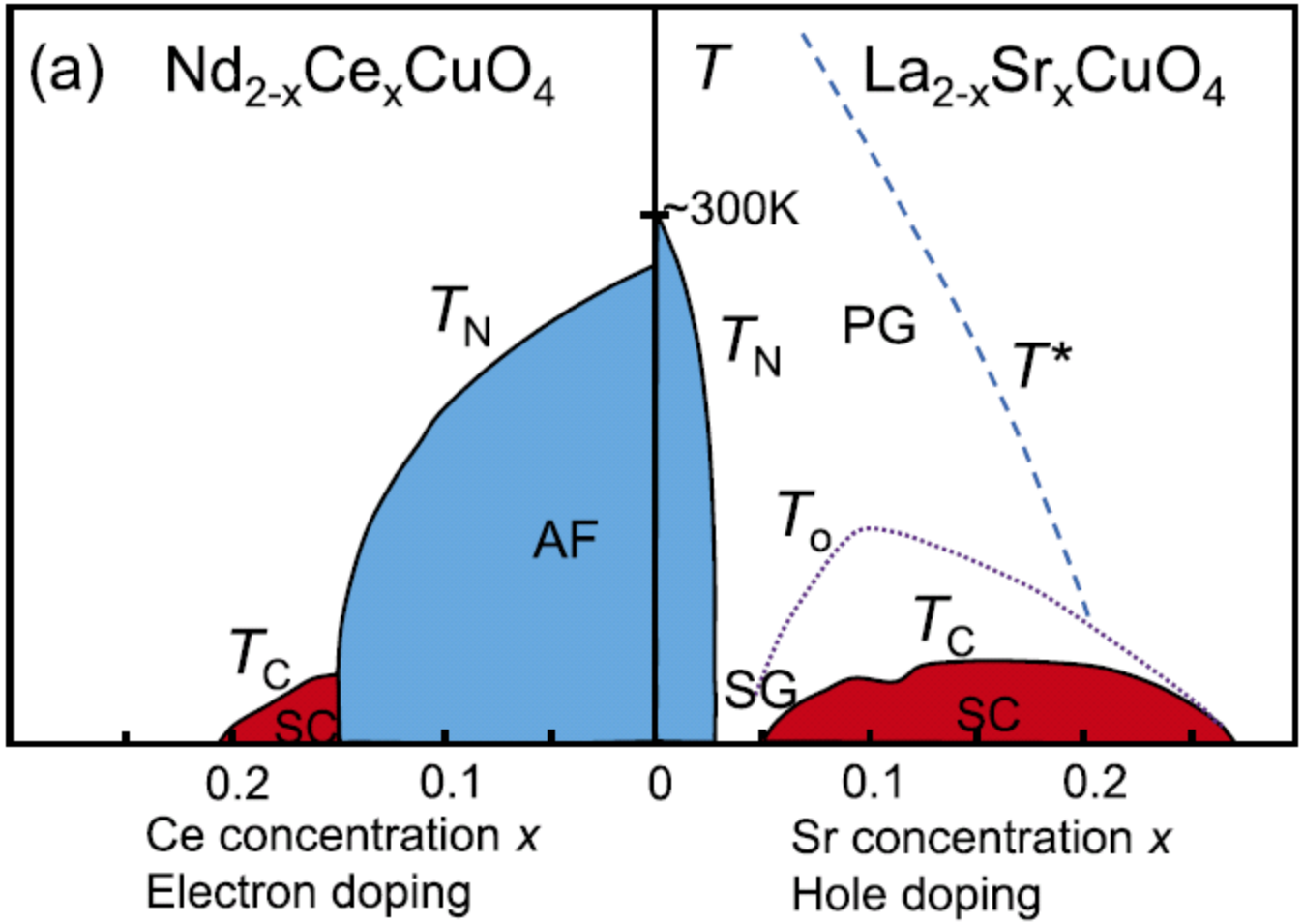
Cu^{2+} $3d^9$ 1 hole on each x^2-y^2 orbital

localized spin → antiferromagnetic exchange interaction

J~1400K

Phase diagrams

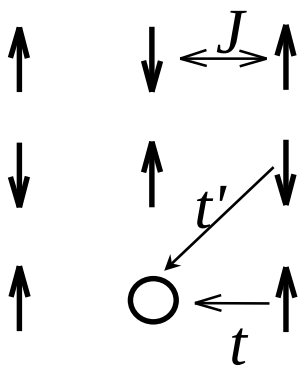
T.T., JJAP **51**, 010004 (2012)



- 3d x^2-y^2 : single-band material
- doped-Mott insulator; d-wave super.
- asymmetric: electron v.s. hole

t-t'-J model

$$H_{tJ} = -t \sum_{<i,j>_1,\sigma} \tilde{c}^{+}_{i,\sigma}\tilde{c}_{j,\sigma} - t' \sum_{<i,j>_2,\sigma} \tilde{c}^{+}_{i,\sigma}\tilde{c}_{j,\sigma} + J \sum_{i\delta} \mathbf{S}_i \cdot \mathbf{S}_{i+\delta}$$



No double occupation

hole-doping

Lower Hubbard band

$t > 0,\ t' < 0$

electron-doping

Upper Hubbard band

$t < 0,\ t' > 0$

$J = 0.4$

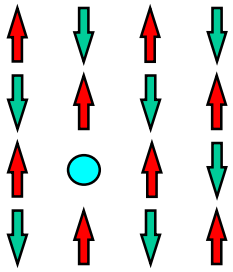
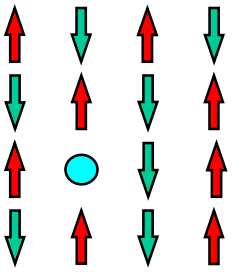
hole $La_{2-x}Sr_xCuO_4$: $t = 1,\ t' = -0.12$

$Bi_2Sr_2Ca_{1-y}R_yCu_2O_{8+z}$: $t = 1,\ t' = -0.34$

electron $Nd_{2-x}Ce_xCuO_4$ (NCCO): $t = -1,\ t' = 0.25$

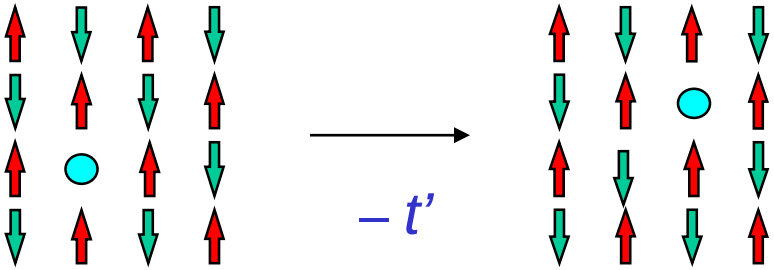
(J = 0.14 eV, t = 0.35 eV)

Effect of next-nearest-neighbor hopping *t'* on AF correlation

$|G.S\rangle \sim c_1 \times$  $+ \; c_2 \times$  $+ \cdots$

The larger $|c_1|$ is, the stronger the AF correlation is.

What is the effect of *t'* on the magnitude of c_1?



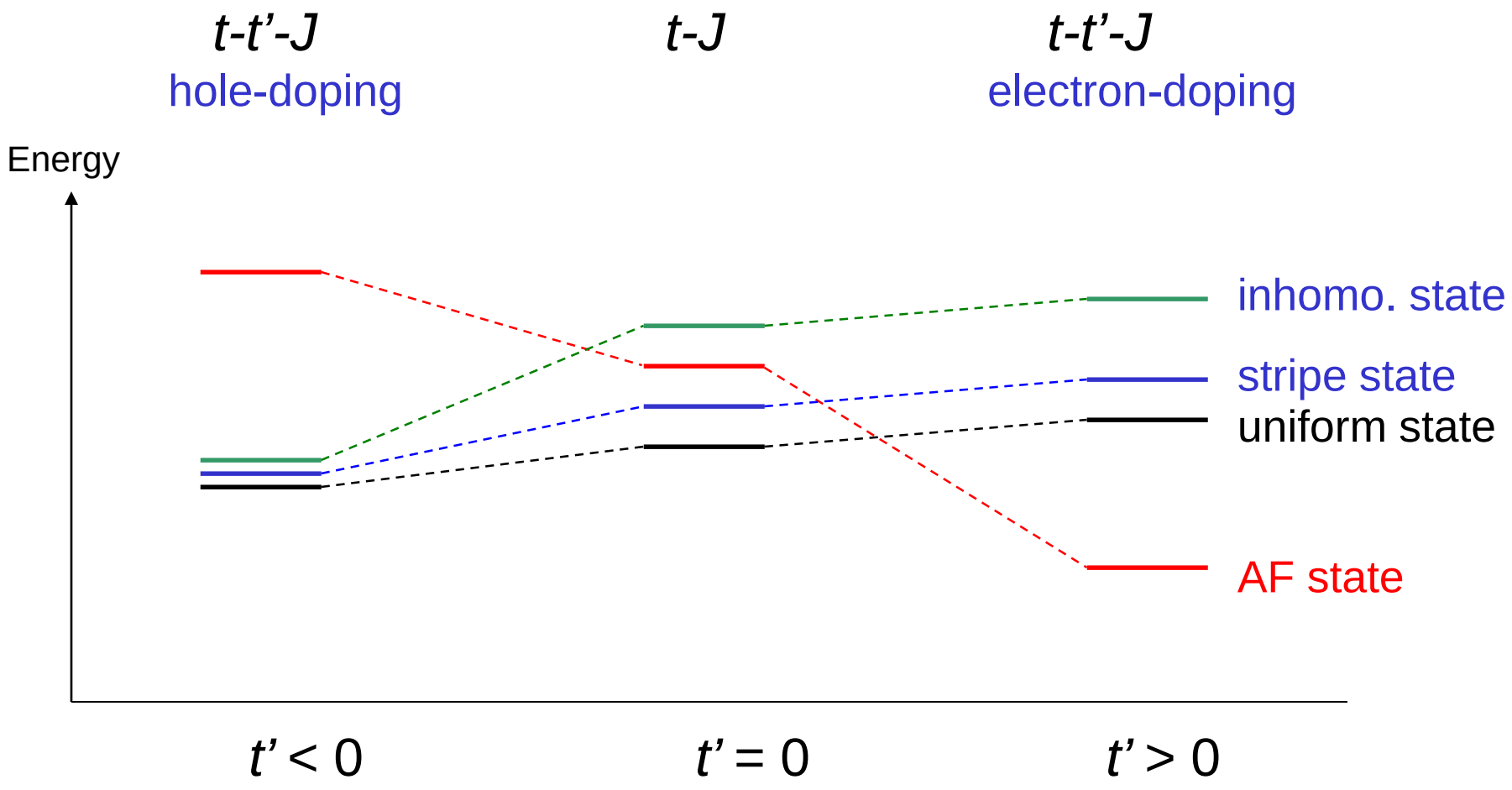
No change of spin configuration

Self-energy gain $-t'$

	hole-doping	electron-doping
$-t'$	> 0	< 0
$|c_1|$	decrease	increase
AF correlation	decrease	increase

Effect of *t'* on many-body electronic states

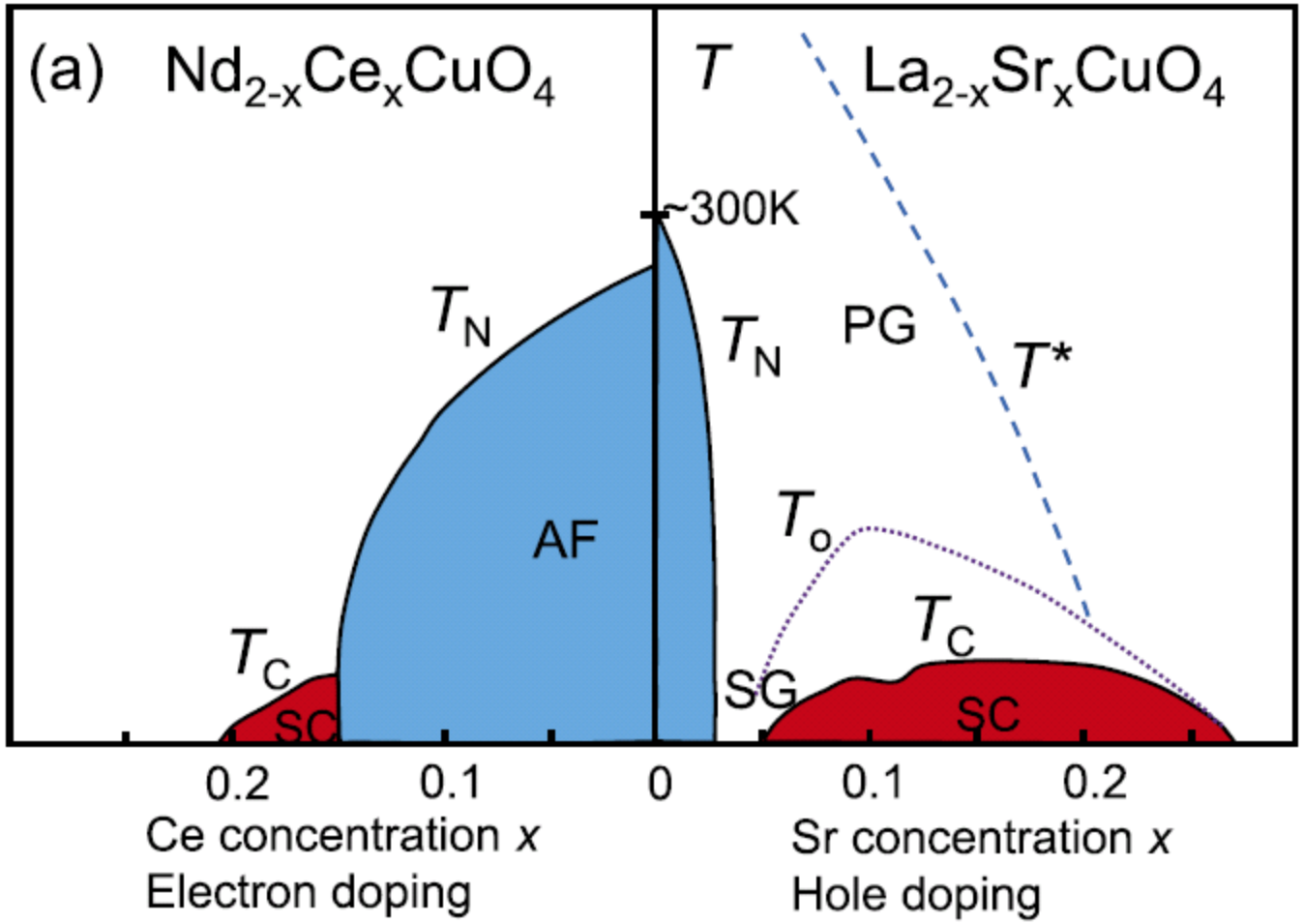
Schematic energy level diagram



T.T., Jpn. J. Appl. Phys. **51**, 010004 (2012)

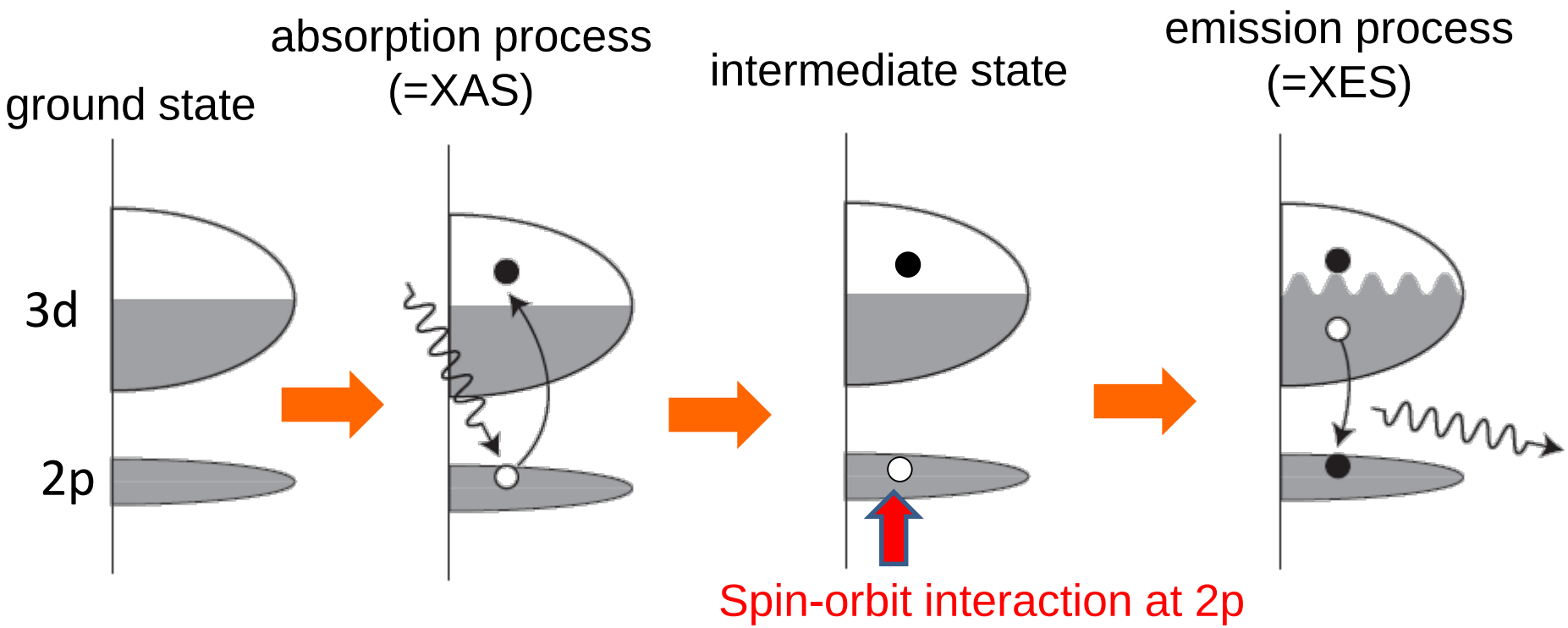
Phase diagrams

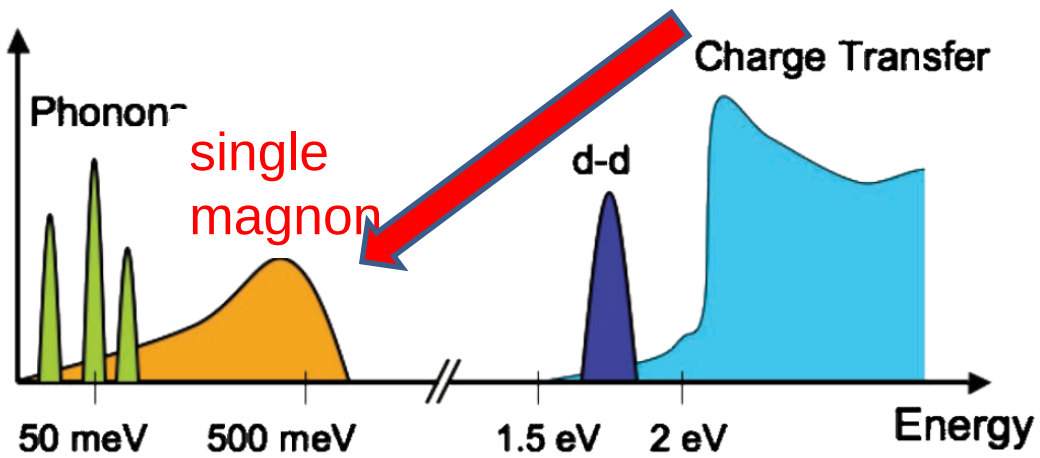
T.T., JJAP **51**, 010004 (2012)



Physics of doped-Mott insulator

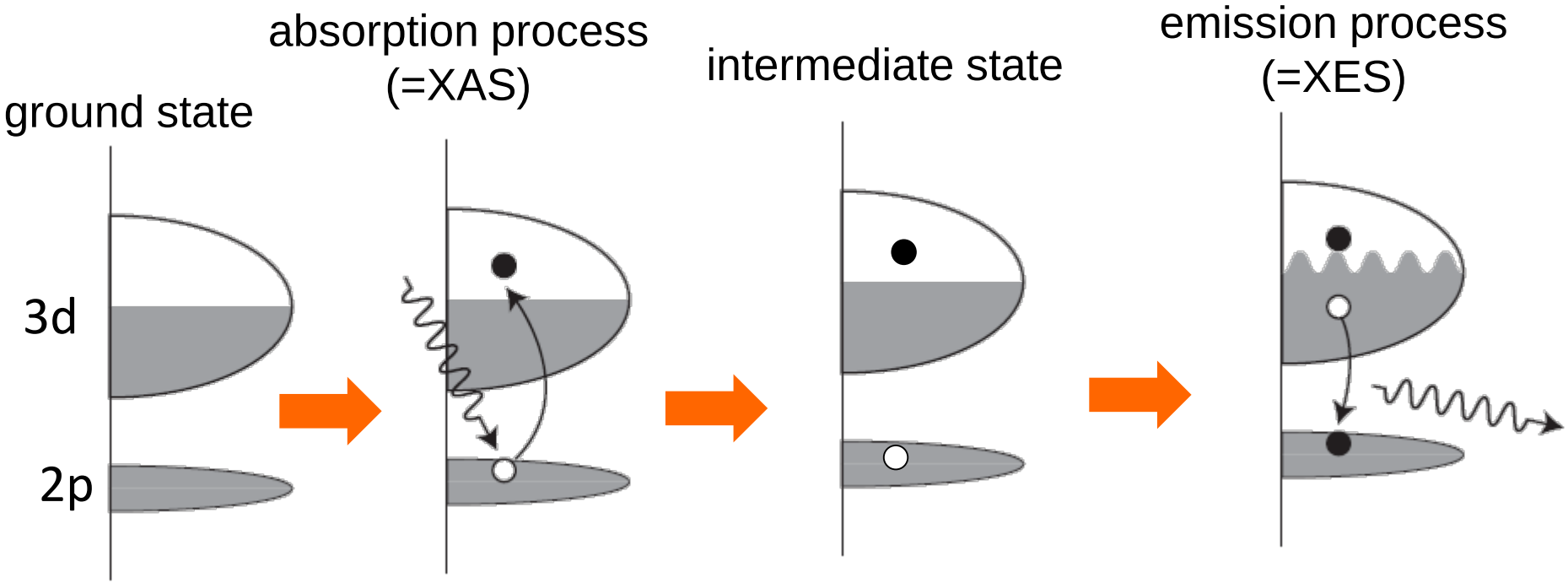
Resonant Inelastic X-ray Scattering (RIXS) for L-edge





L. J. P. Ament *et al.*, Rev. Mod. Phys. **83**, 705 (2011)

RIXS for L-edge



$$I(q,\Delta\omega) \sim \sum_h \left| \langle h | D^{+}_{K_o,\varepsilon_o} \frac{1}{H - E_g - \omega_i - i\Gamma} D_{K_i,\varepsilon_i} | g \rangle \right|^2 \delta\left(\Delta\omega - E_h + E_g\right)$$

$$\Delta\omega = \omega_i - \omega_o$$

$$q = K_i - K_o$$

$|g\rangle$: ground state (no Cu2p hole)

$|h\rangle$: 'final' state with momentum q

$D_{K,\varepsilon}$: dipole transition operator

Two contributions to RIXS spectrum

Non spin-flip excitation → Dynamical charge structure factor

$$\Delta S = 0 \qquad I_{\mathbf{q}}^{\Delta S=0}(\Delta\omega) = \sum_f |\langle f|N_{\mathbf{q}}^j|0\rangle|^2 \delta(\Delta\omega - E_f + E_0)$$

$$N_{\mathbf{q}}^j = B_{\mathbf{q}\uparrow\uparrow}^j + B_{\mathbf{q}\downarrow\downarrow}^j$$

Spin-flip excitation → Dynamical spin structure factor

$$\Delta S = 1 \qquad I_{\mathbf{q}}^{\Delta S=1}(\Delta\omega) = \sum_f |\langle f|S_{\mathbf{q}}^j|0\rangle|^2 \delta(\Delta\omega - E_f + E_0)$$

$$S_{\mathbf{q}}^j = (B_{\mathbf{q}\uparrow\uparrow}^j - B_{\mathbf{q}\downarrow\downarrow}^j)/2$$

$$\boxed{B_{\mathbf{q}\sigma'\sigma}^j = \sum_l e^{-i\mathbf{q}\cdot\mathbf{R}_l} d_{l\sigma'}^\dagger \frac{1}{\omega_{\mathrm{i}} - H_l^j + E_0 + i\Gamma} d_{l\sigma}}$$

Large Γ limit

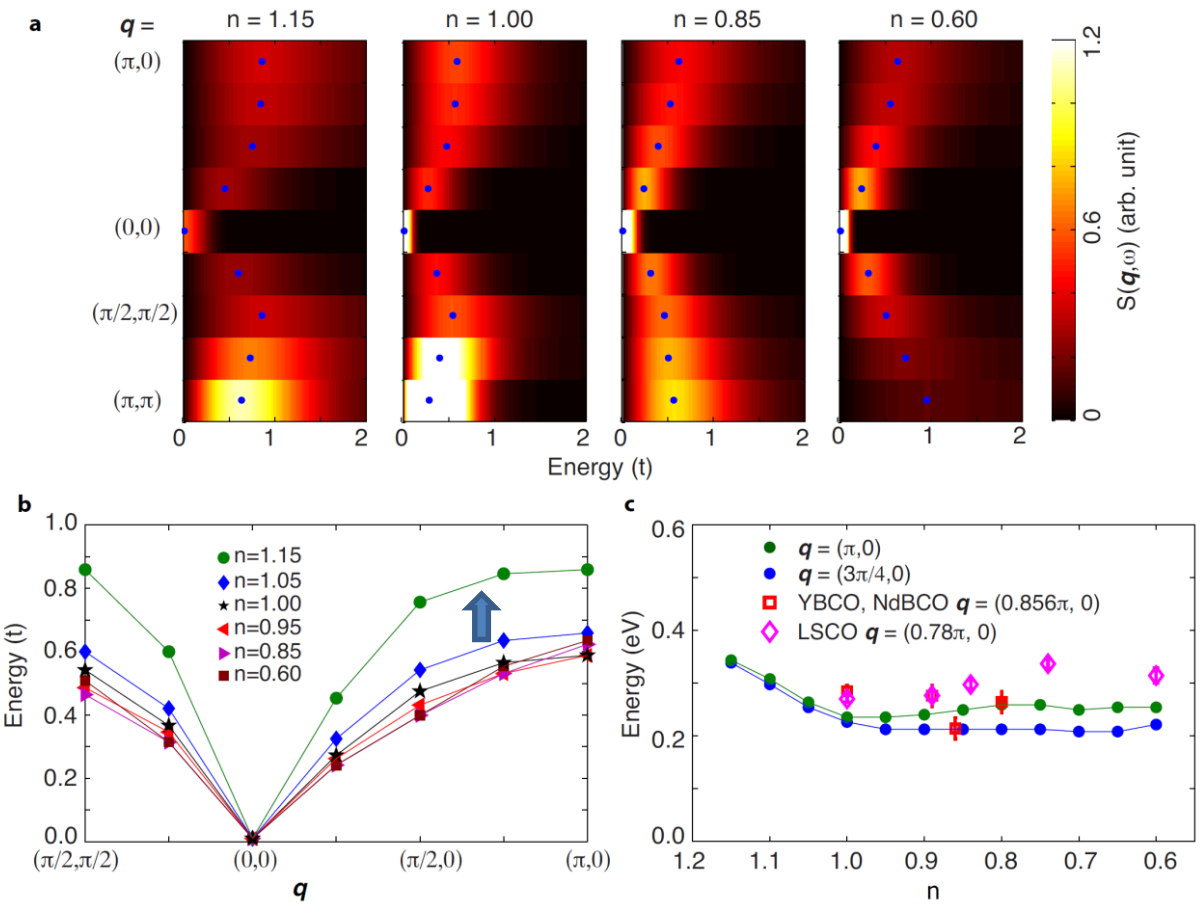
$$H_l^j = H_{3d} + U_{\mathrm{c}} \sum_\sigma n_{l\sigma} + \varepsilon_j$$

Dynamical spin structure factor in 2D t-t' Hubbard model

[C. J. Jia *et al.*, Nature Commun. 5, 3314 (2014)]

U/*t*=8, *t'*/*t*= -0.3
t=400meV

Quantum Monte Carlo, β=3/*t*



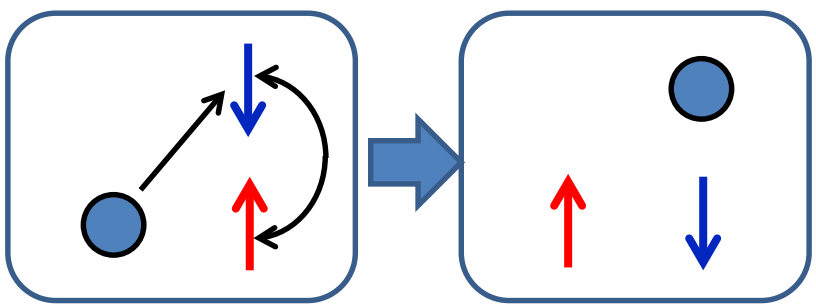
- Increase of spin-excitation energy for *n*>1 (electron doping)

Comparison between t-t'-U model and t-t'-J model

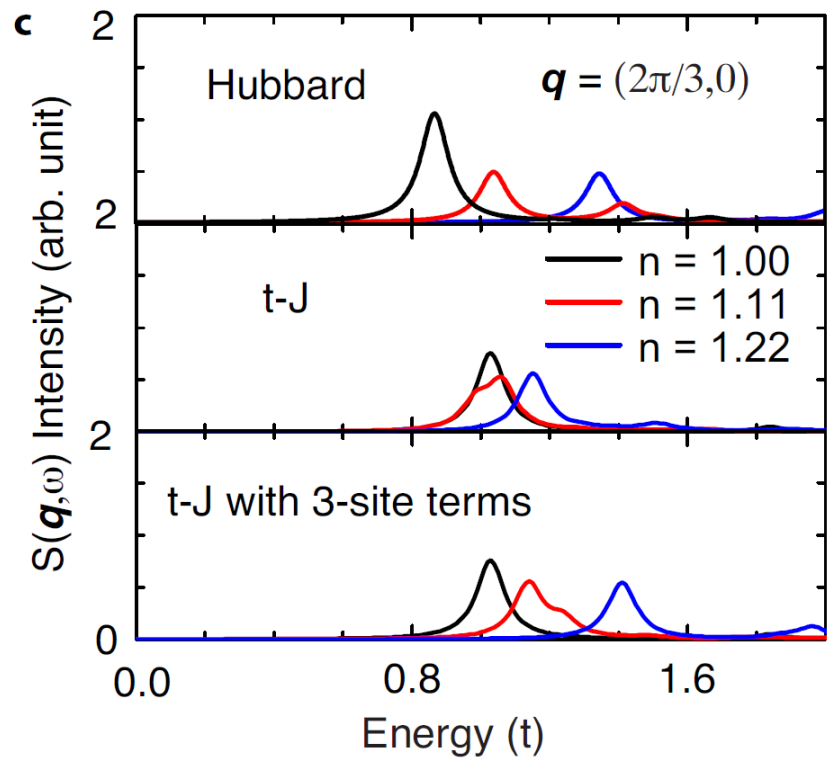
[C. J. Jia *et al*., Nature Commun. 5, 3314 (2014)]

3-site hopping terms (order of J)

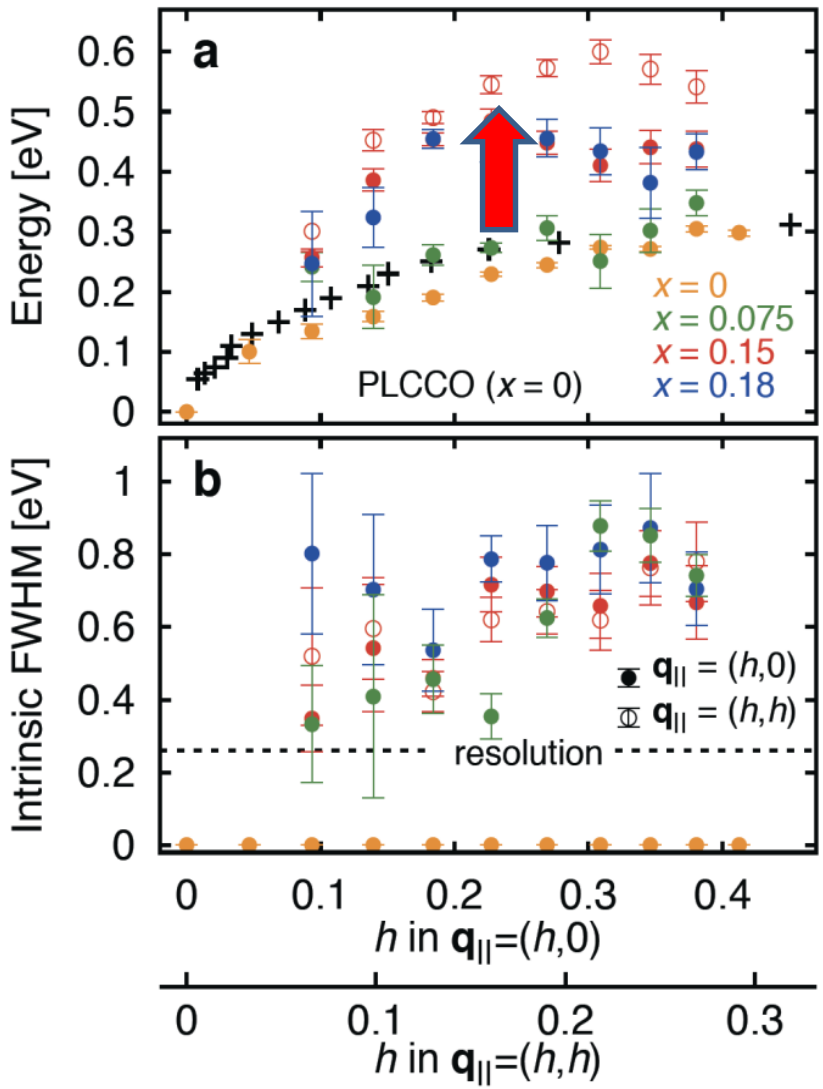
$$H_{3S} = -\frac{J}{4} \sum_{\langle i,j,k\rangle,\sigma,\sigma'} (1 - n_{i,-\sigma}) c^{\dagger}_{i,\sigma} c_{j,\sigma} n_{j,-\sigma} \times n_{j,-\sigma'} c^{\dagger}_{j,\sigma'} c_{k,\sigma'} (1 - n_{k,-\sigma'}),$$



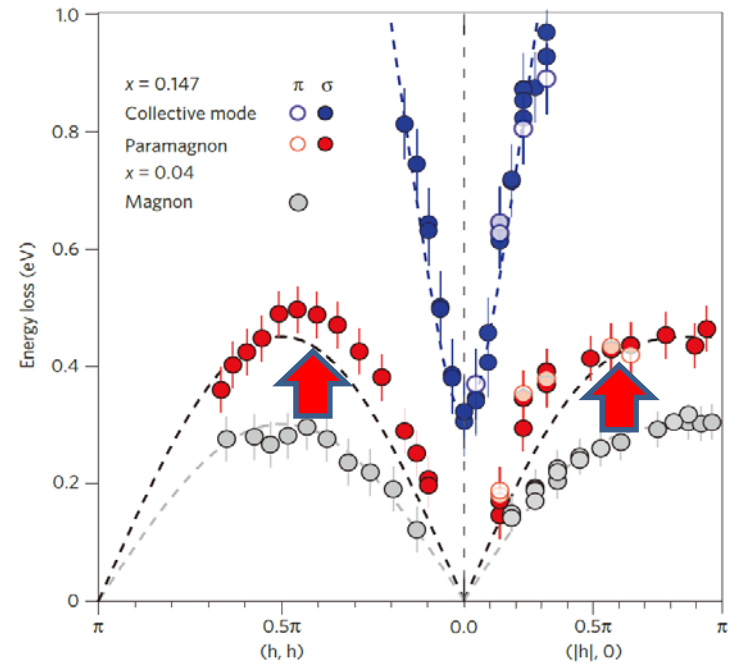
t-t'-J vs. *t-t'-U* Hubbard



Spin-flip excitations observed by RIXS in electron-doped cuprates



[K. Ishii *et al*., Nat. Commun. **5**, 3714 (2014)]



[W. S. Lee *et al*., Nat. Phys. **10**, 883 (2014)]

Increase of spin-flip excitation energy with doping

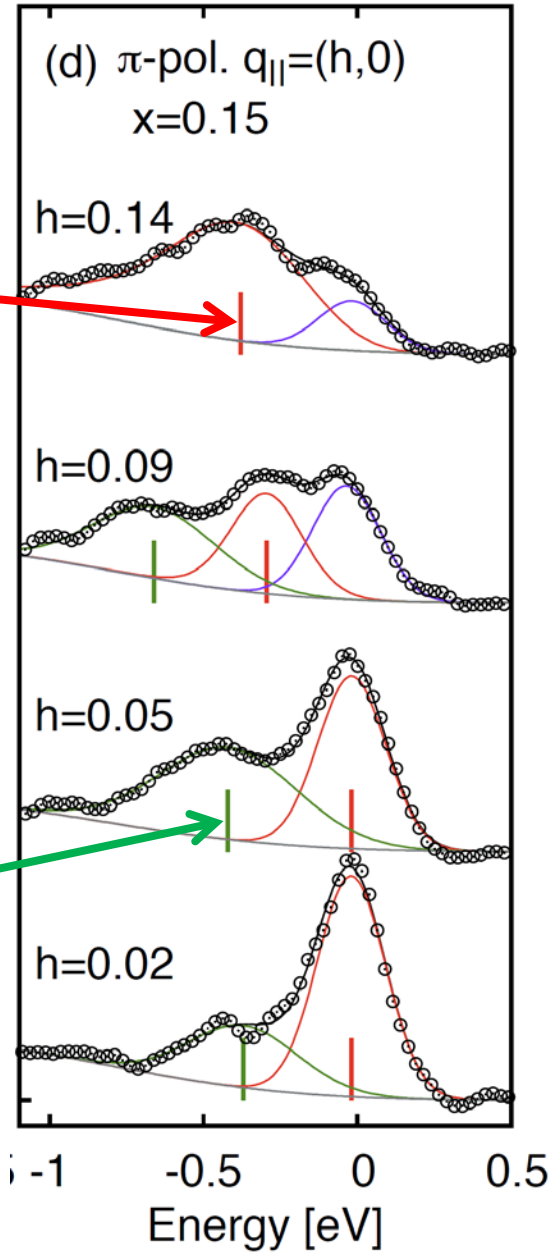
Consistent with theory

Charge dynamics seen in RIXS for $Nd_{2-x}Ce_xCuO_4$

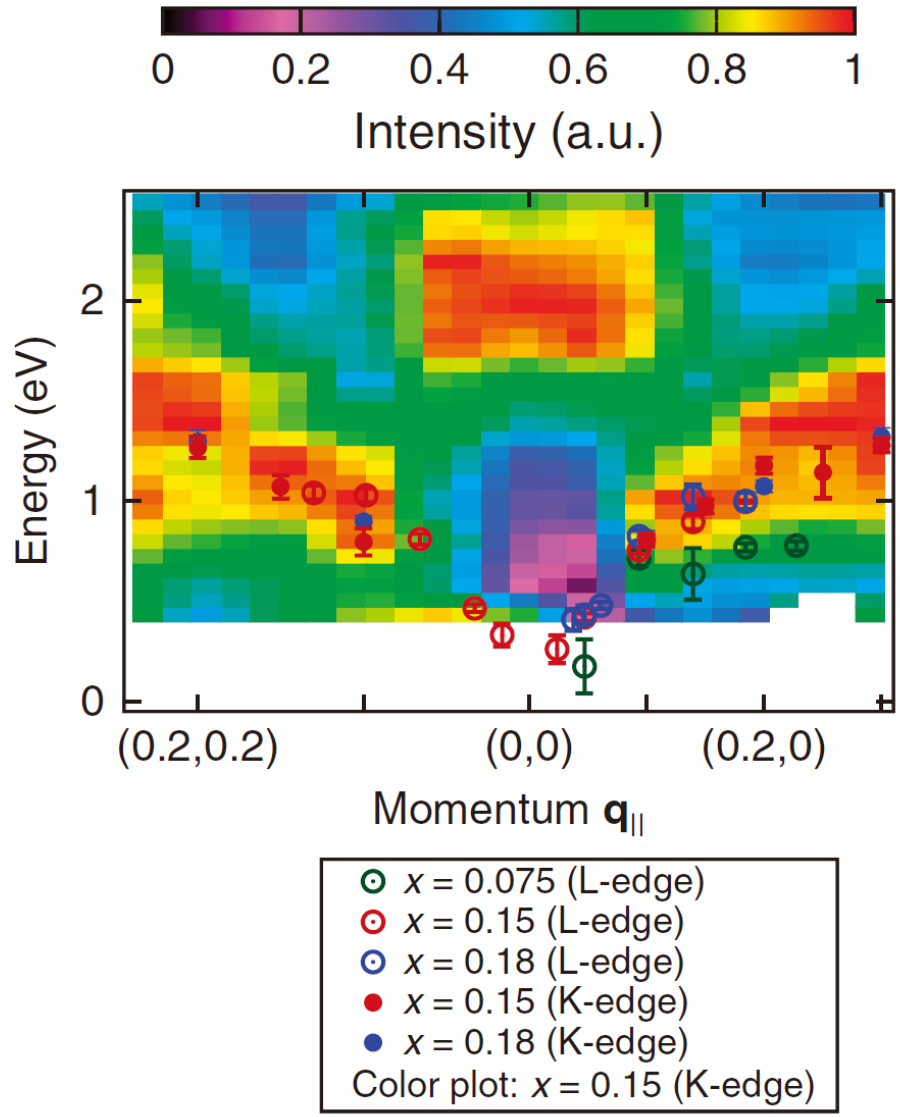
[Ishii *et al.*, Nat. Comm. 5, 3714 (2014)]

spin

charge



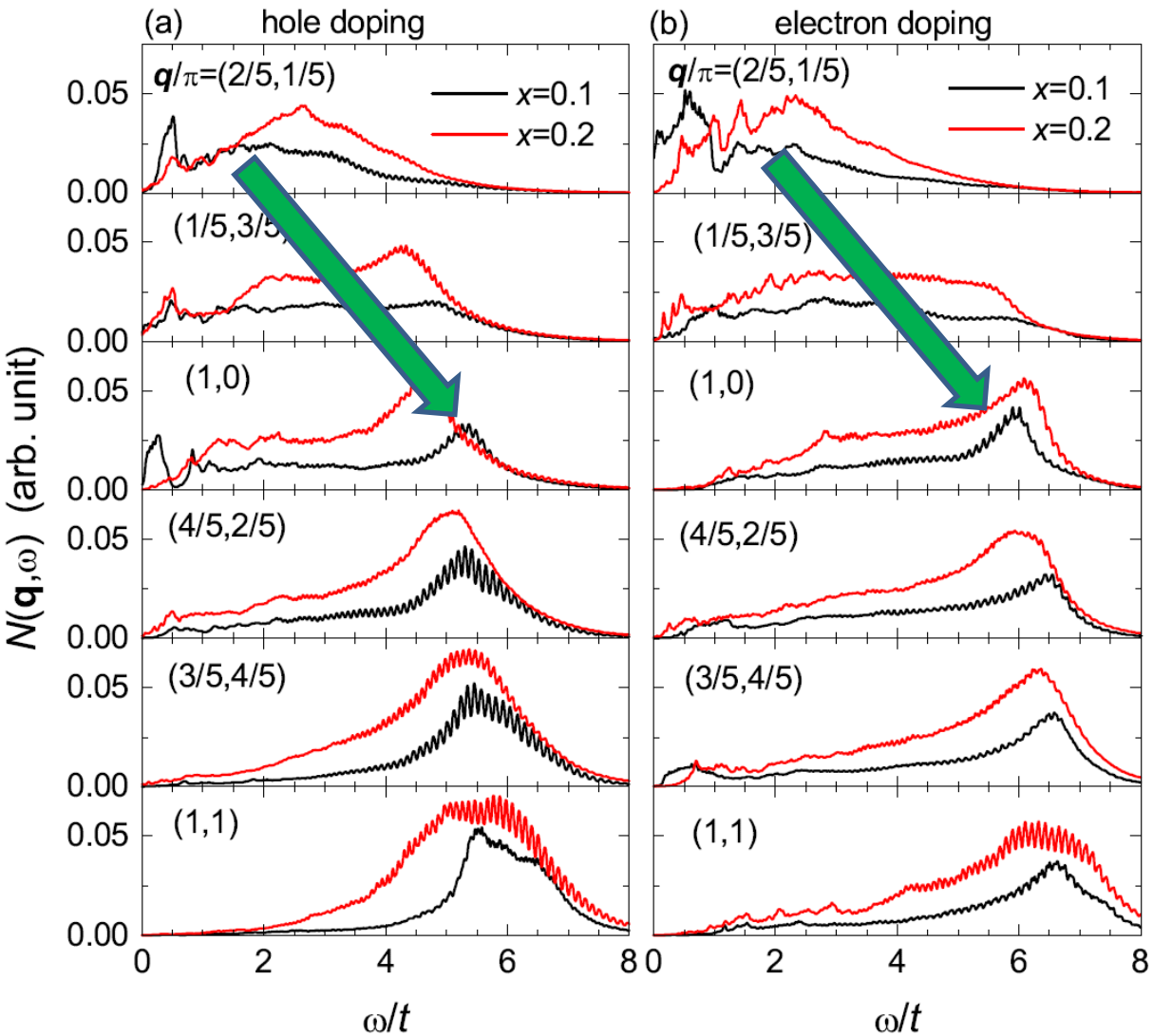
Same as K-edge RIXS [Ishii *et al.*, PRL **91**, 207003 ('05)]



Similar structure in overdoped LSCO [Wakimoto *et al.*, PRB **91**, 184513 (2015)]

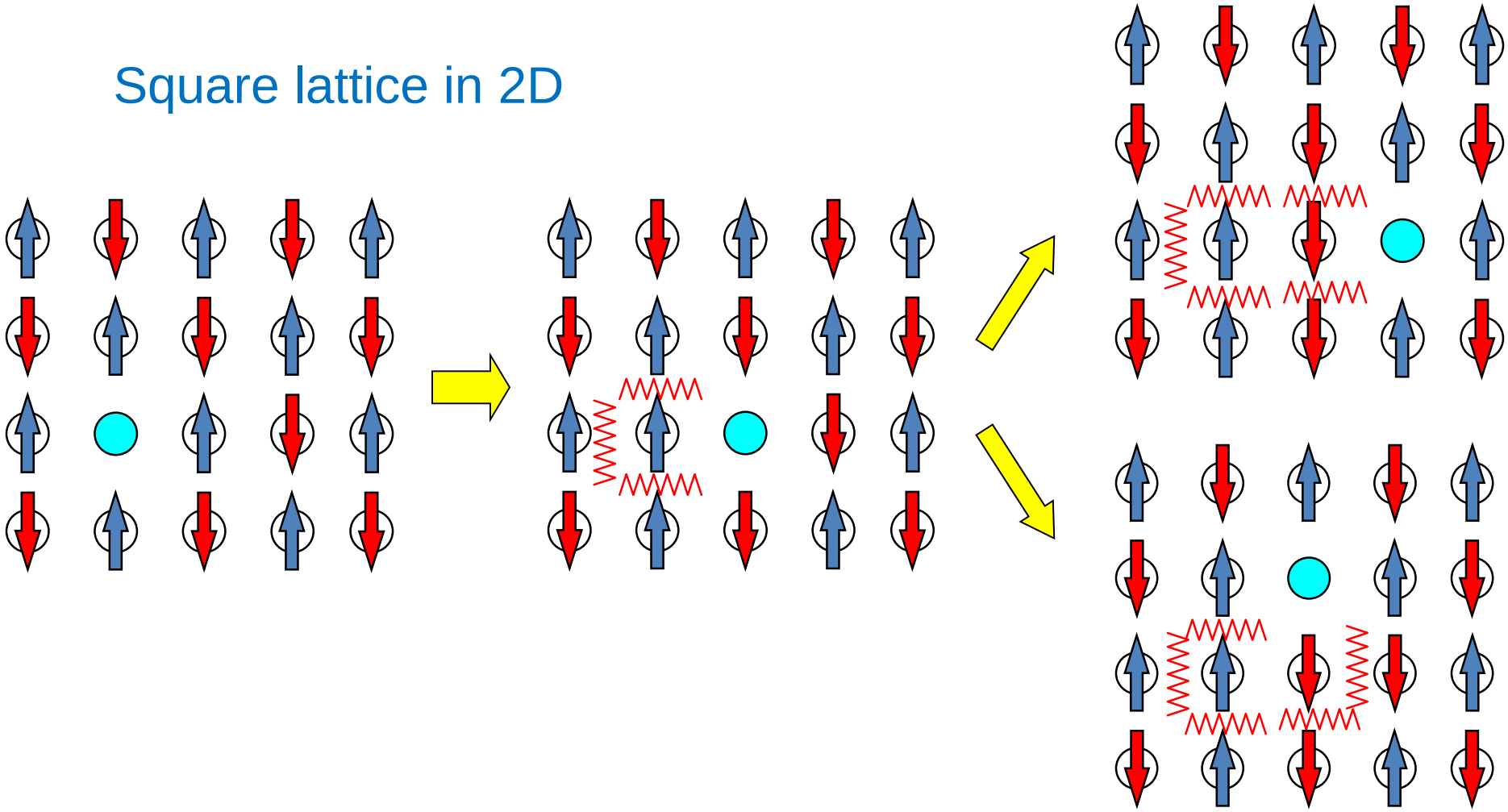
Dynamical charge structure factor in the 20-site *t-t'-J* model

T.T., J. Electro. Spector. and Related Phenome., **200**, 209 (2015)



Doped Mott Insulator

Square lattice in 2D

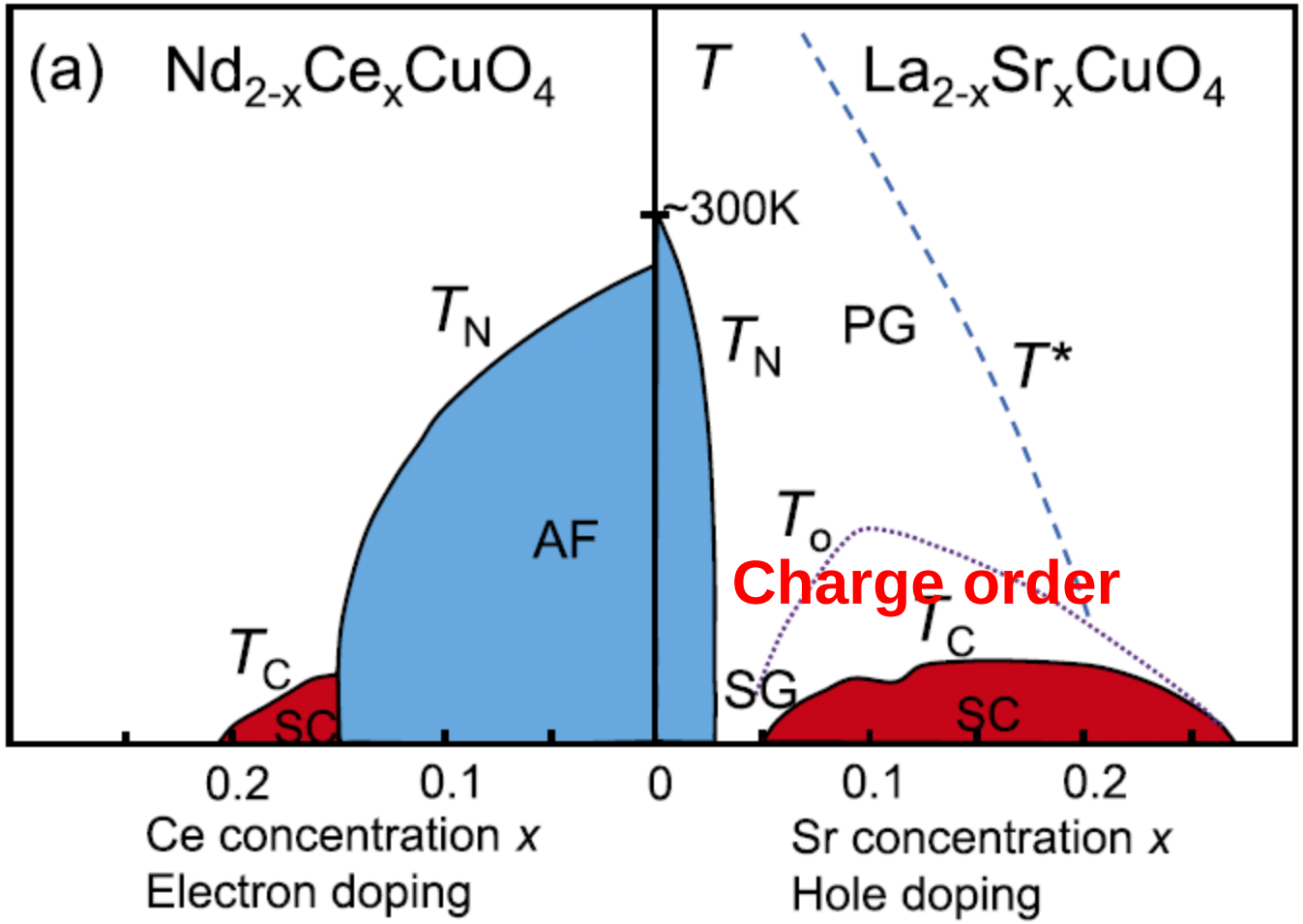


Hole motion is affected by spin background: spin-charge coupling

Incoherent charge excitation

Phase diagrams

T.T., JJAP **51**, 010004 (2012)

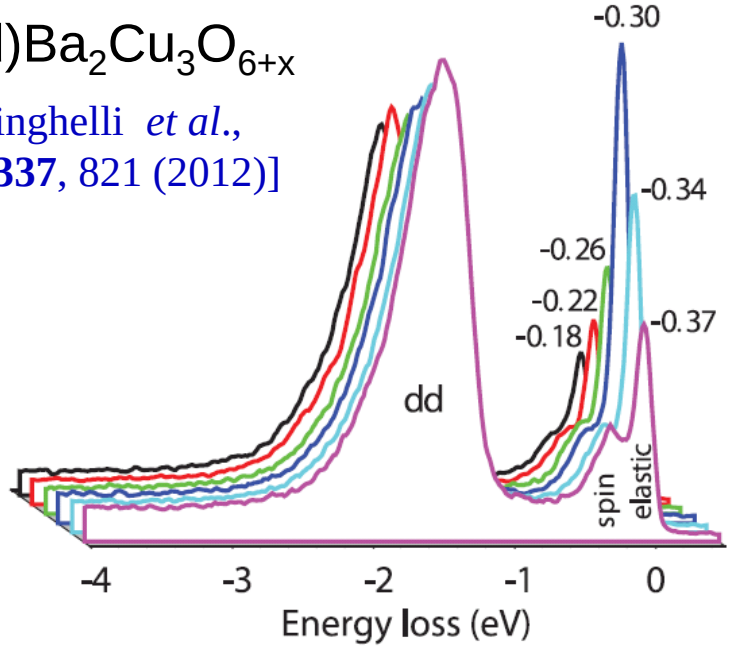


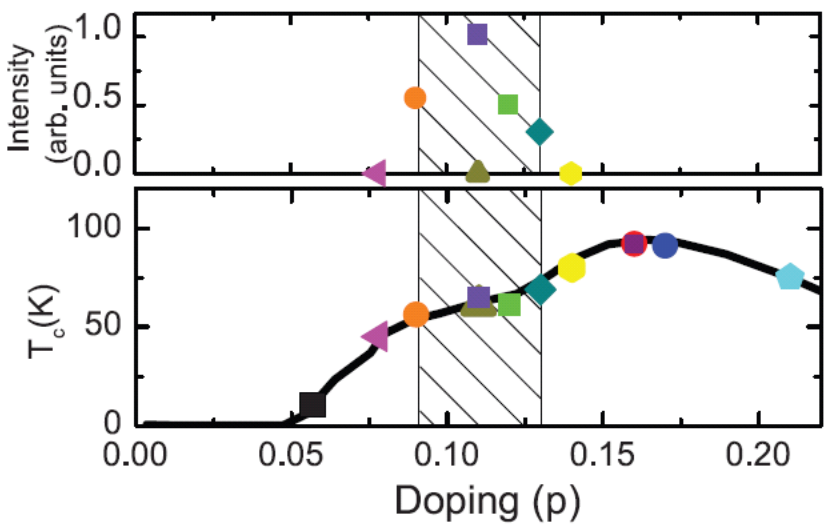
Physics of doped-Mott insulator

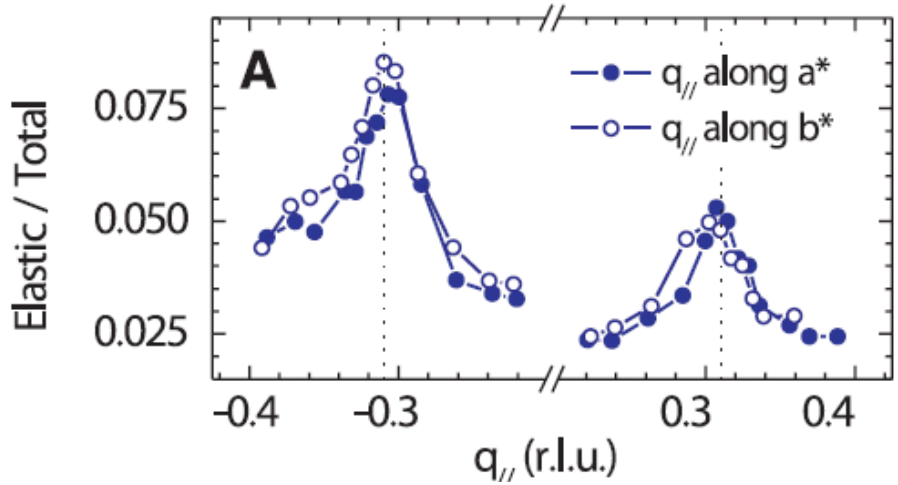
Charge order of hole-doped cuprates seen by RIXS

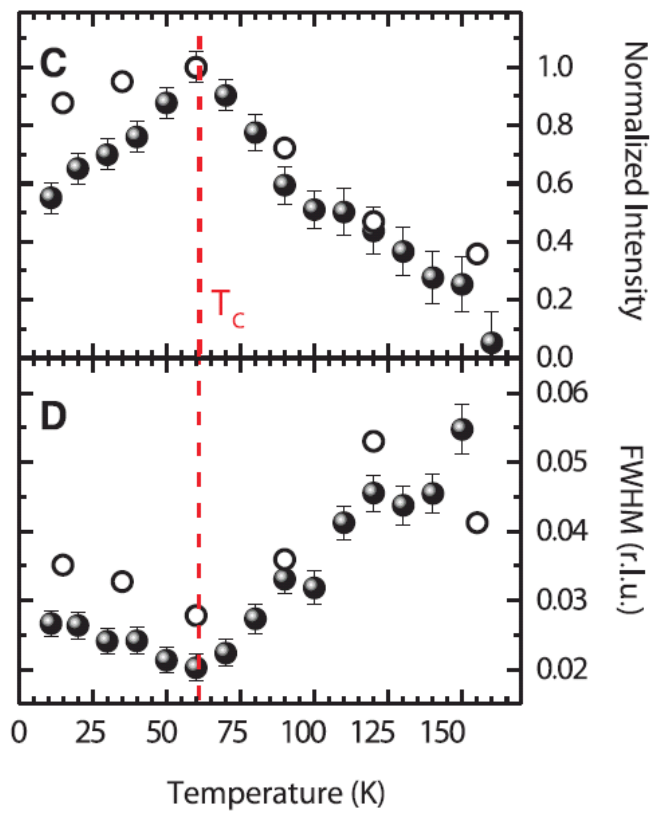
$(Y,Nd)Ba_2Cu_3O_{6+x}$

[G. Ghiringhelli *et al.*, Science **337**, 821 (2012)]









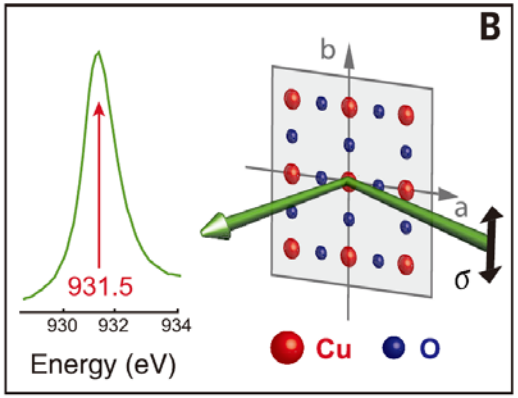
Long-range incommensurate charge order

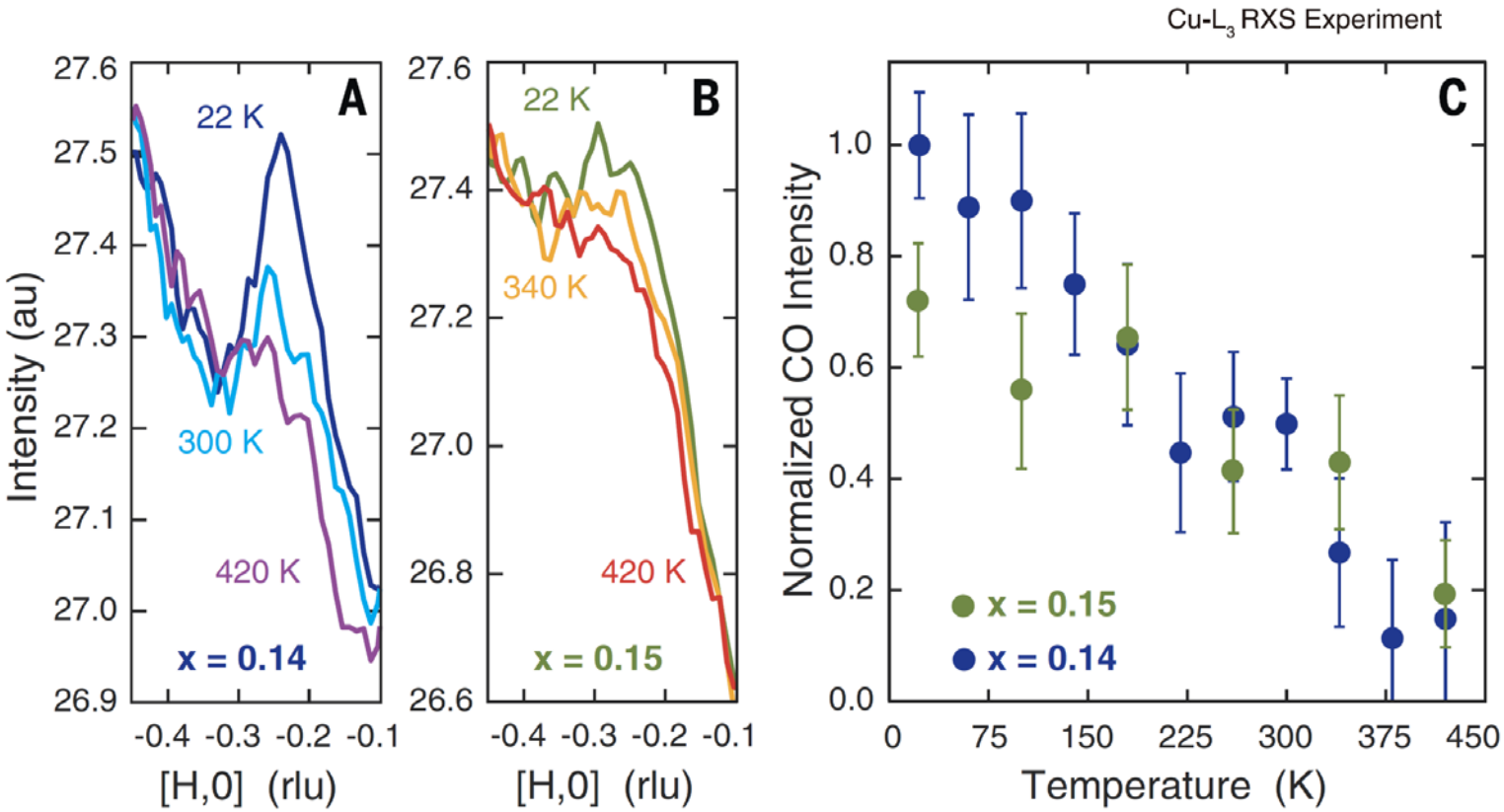
Charge order of electron-doped cuprates

$Nd_{2-x}Ce_xCuO_4$

[E. H. da Silva Neto *et al*., Science **347**, 282 (2015)]

Resonant x-ray scattering

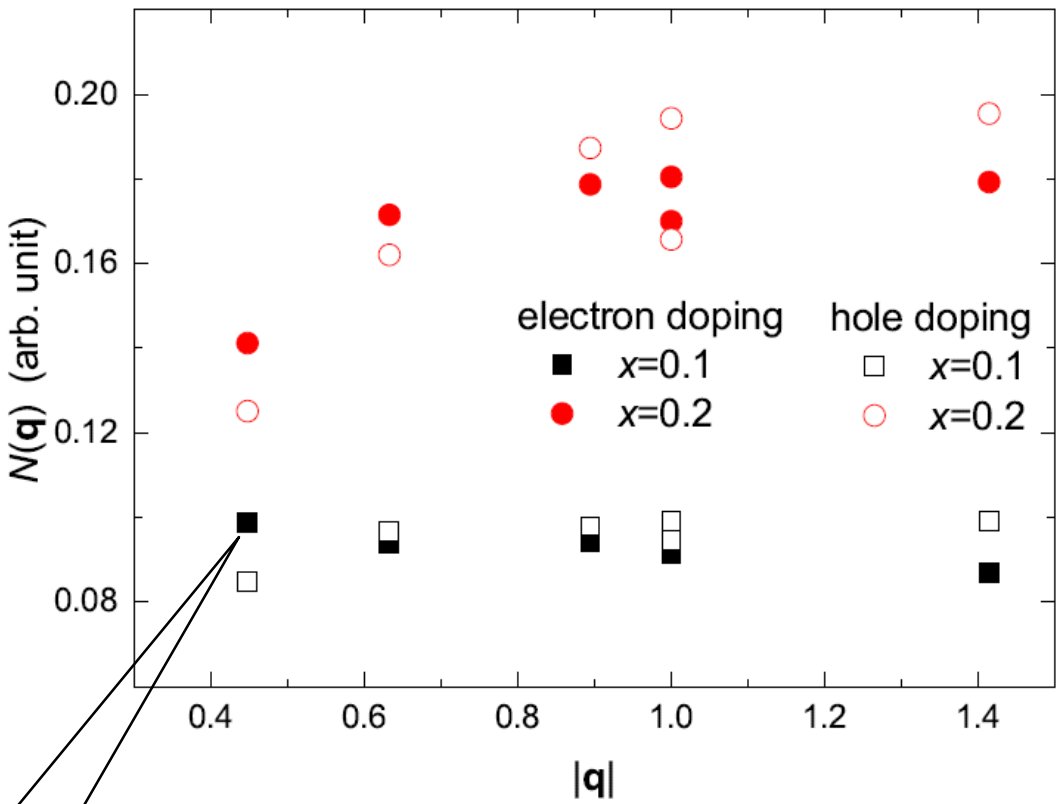




Static charge structure factor in the 20-site *t-t'-J* model

T.T., J. Electro. Spector. and Related Phenome., **200**, 209 (2015)

$$N(\mathbf{q}) = \int N(\mathbf{q},\omega)d\omega$$



Enhancement of $N(\mathbf{q})$ at small $\mathbf{q}$ in underdopded electron system

c.f. Enhancement of charge fluctuations near the phase separation

t-J model (many groups), Hubbard model (e.g., T. Misawa, M. Imada, PRB 95,115137 (2014))

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3. Q-dependent "coherent" charge excitations

Theoretical prediction t-t'-U Hubbard model

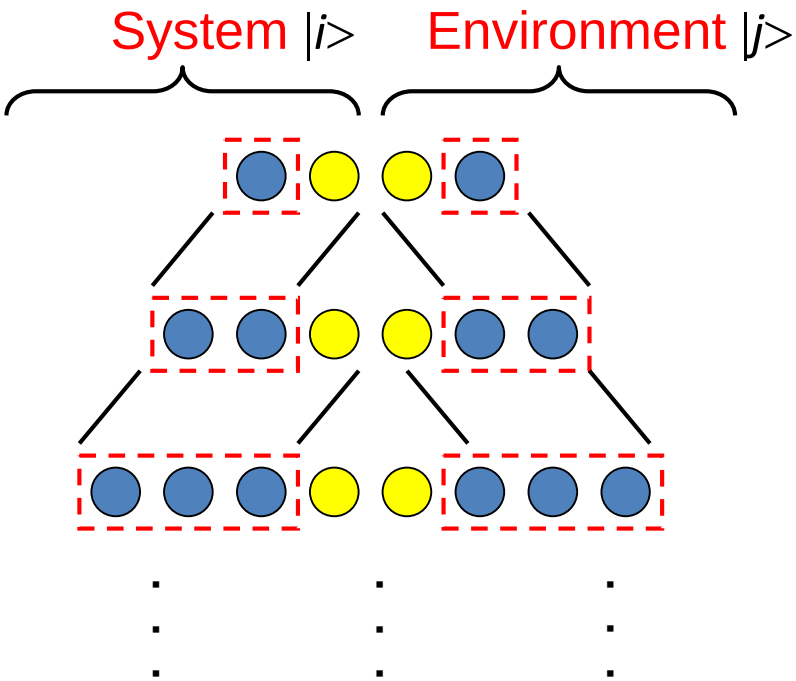
Density-matrix renormalization group (DMRG)

Low-energy charge mode, detectable by RIXS

K. Ishii, M. Fujita, T. T. *et al*., Nat. Commun. **5**, 3714 (2014)
T. T., K. Tsutsui, M. Mori, S. Sota, S. Yunoki, Phys. Rev. B **92**, 014515 (2015)
T.T., J. Electro. Spector. and Related Phenome., **200**, 209 (2015)

Density-matrix renormalization group（DMRG)

[S. R. White, PRL **69**, 2863 (1992)]



Renormalize the states of the Environment into those of the System for each step, by using the density-matrix given by the ground-state wave function.

ground-state wave function

$$|\psi\rangle = \sum_{i,j} \psi_{ij} |i\rangle |j\rangle$$

density matrix

$$\rho_{ii'} = \sum_j \psi_{ij} \psi_{i'j}$$

$$\langle A \rangle = \mathrm{Tr}(\rho A) = \sum_\alpha \omega_\alpha \langle u^\alpha | A | u^\alpha \rangle \approx \sum_{\alpha=1}^{m} \omega_\alpha \langle u^\alpha | A | u^\alpha \rangle$$

$|u^\alpha\rangle$: eigenstate of ρ

$\omega_\alpha (\geq 0)$: eigenvalue of ρ

m: truncation number

discard unimportant states: $\omega_\alpha \approx 0$

Dynamical density-matrix renormalization group (DMRG)

ψ_{ij} system environment

i j

[E.Jeckelmann, PRB**66**, 045114 (2002)]

Density matrix $\rho_{ii'} = \sum_{\alpha} p_{\alpha} \sum_{j} \psi_{\alpha,ij}\, \psi_{\alpha,i'j}$, $\sum_{\alpha} p_{\alpha} = 1$

Multi target: α

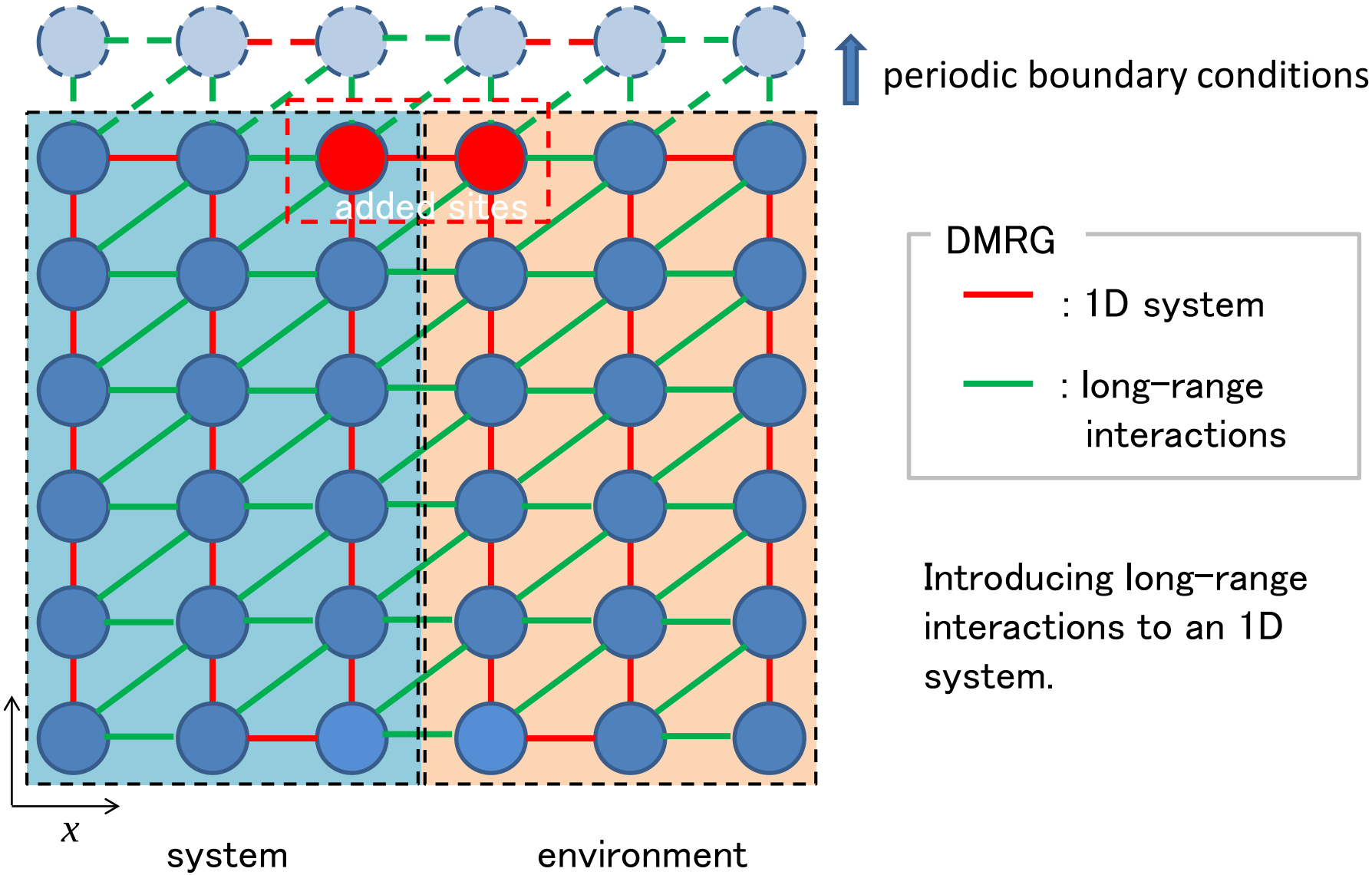
$$S(q,\omega) = \frac{1}{\pi} \mathrm{Im} \langle 0 | S_{-q} \frac{1}{\omega - E_0 + H - i\gamma} S_q | 0 \rangle$$

$$\psi_{\alpha}(\omega) = \begin{cases} |0\rangle \\ S_q |0\rangle \\ (\omega - E_0 + H - i\gamma)^{-1} S_q |0\rangle \end{cases} \Rightarrow \rho(\omega)$$

The last target depends on the energy ω.

DMRG in 2 Dimensions

DMRG method can be applied to 2-D systems



Introducing long-range interactions to an 1D system.

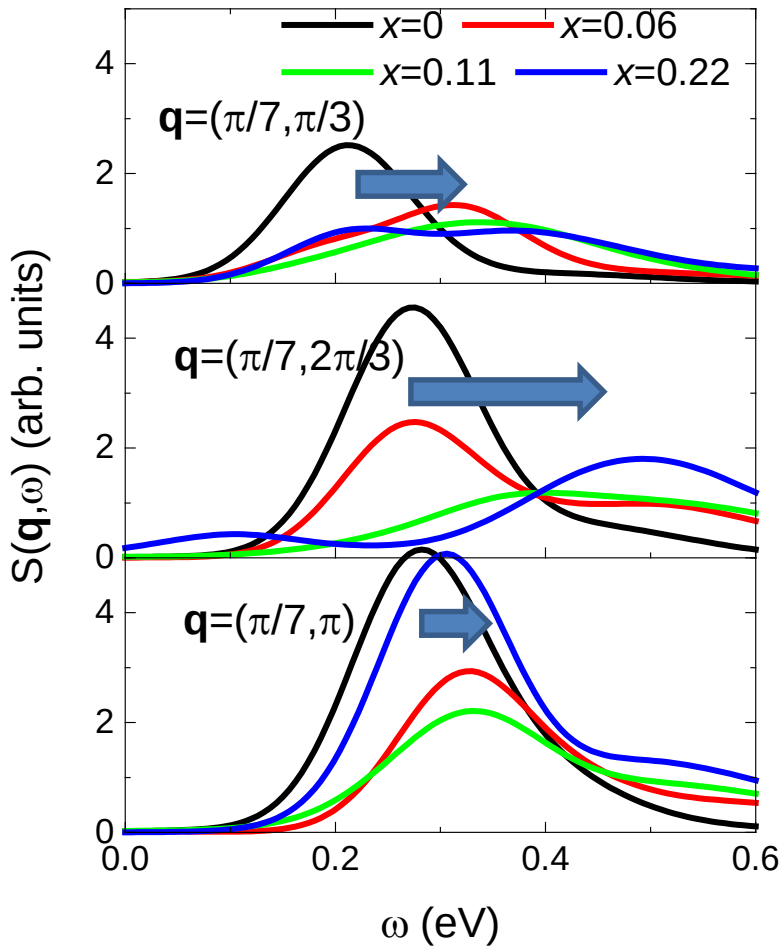
Dynamical spin structure factor S(**q**,ω) by DMRG

Doping dependence of S(**q**,ω) in electron-doped 2D Hubbard model

6x6 sites

Parameters:
t=0.3eV
U/*t*=8, *t'*/*t*=-0.3

x: carrier concentration



☑ Shift of peak toward high energy with x

Consistent with experiment

[K. Ishii *et al*., Nat. Comm. **5**, 3714 (2014)]

[W. S. Lee *et al*., Nat. Phys. **10**, 883 (2014)]

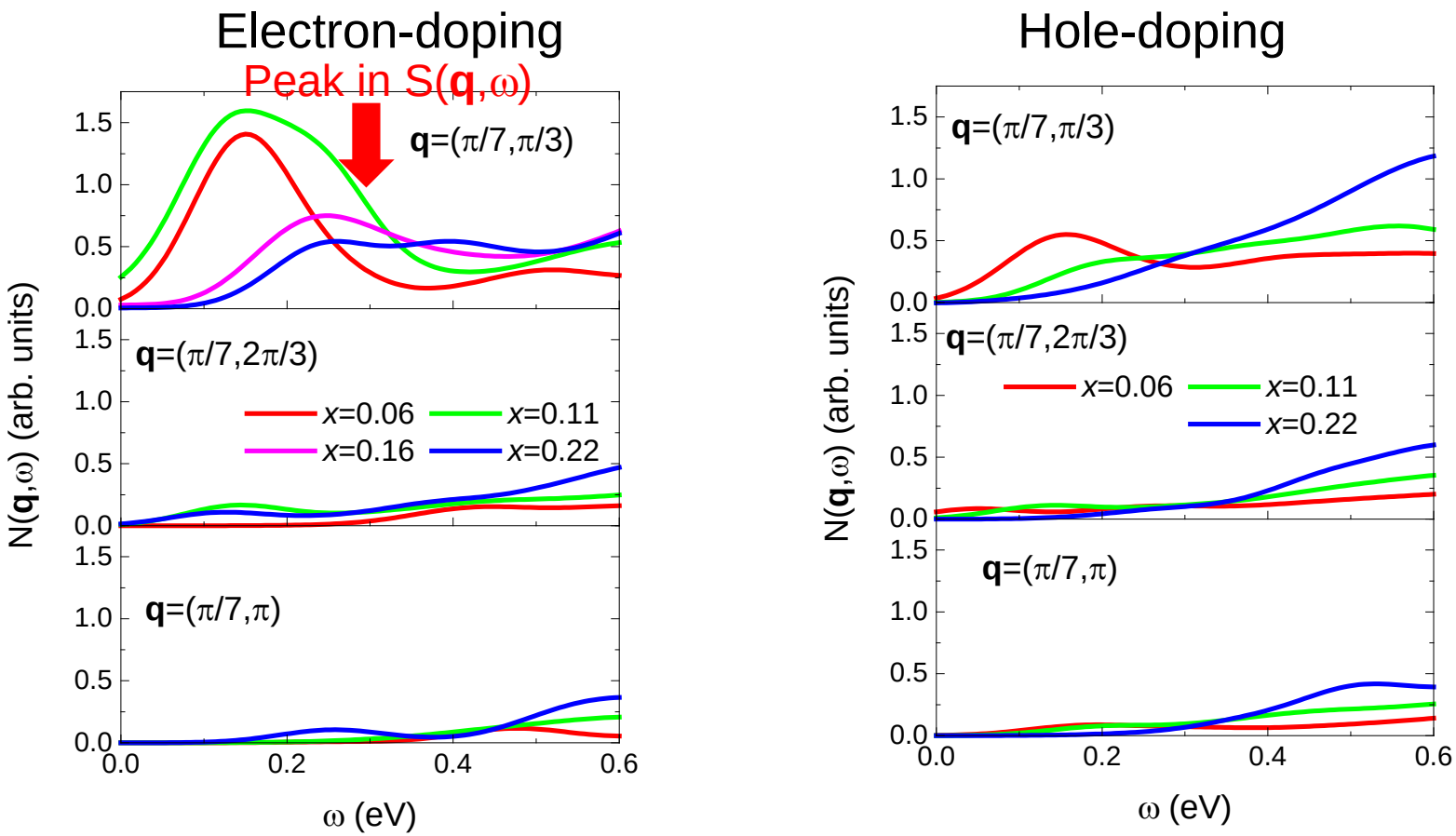
☑ Consistent with quantum Monte Carlo calculations

[C. J. Jia *et al*., Nat. Commun. **5**, 3314 (2014)]

Dynamical charge structure factor N(**q**,ω) by DMRG

Doping dependence of N(**q**,ω) in electron-doped *t-t'-U* Hubbard model

T. T., K. Tsutsui, M. Mori, S. Sota, S. Yunoki, Phys. Rev. B **92**, 014515 (2015)

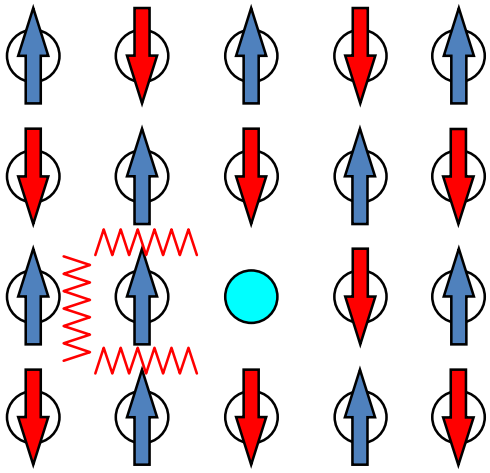


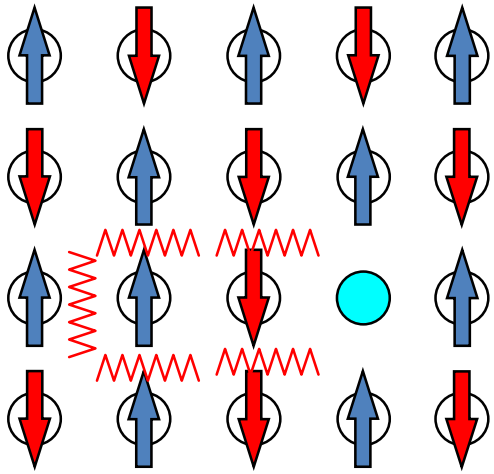
☑ Strong intensity at low-q, low-energy

☑ Lower in energy than spin excitations

Prediction for experiments

Charge motion in doped Mott insulator





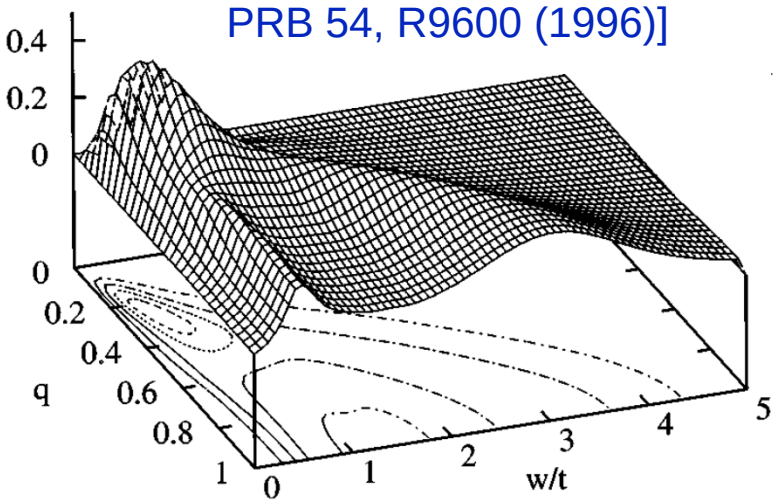
Incoherent motion

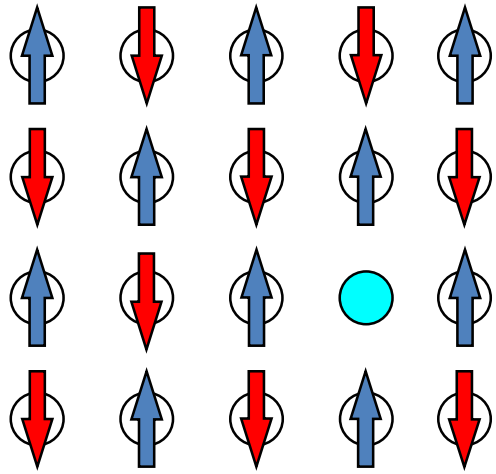
energy scale:
hopping t

already observed by RIXS (Ishii et al.)

$N(\mathbf{q},\omega)$ of t-J model

[G. Khaliullin, P. Horsch, PRB 54, R9600 (1996)]





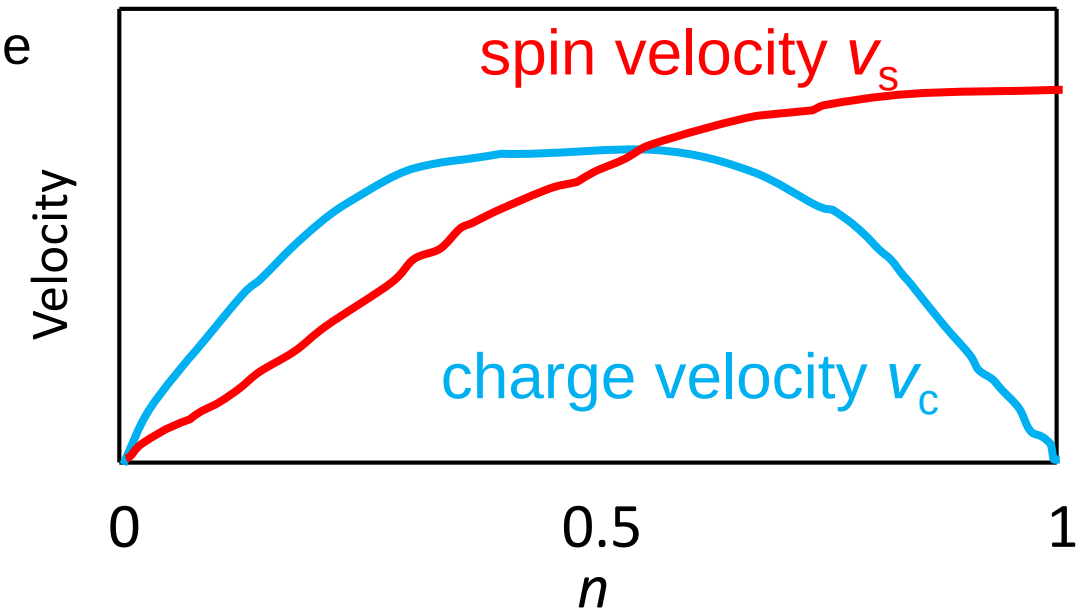
Coherent motion

energy scale:
magnetic J

Prediction to RIXS (T. T. et al.)

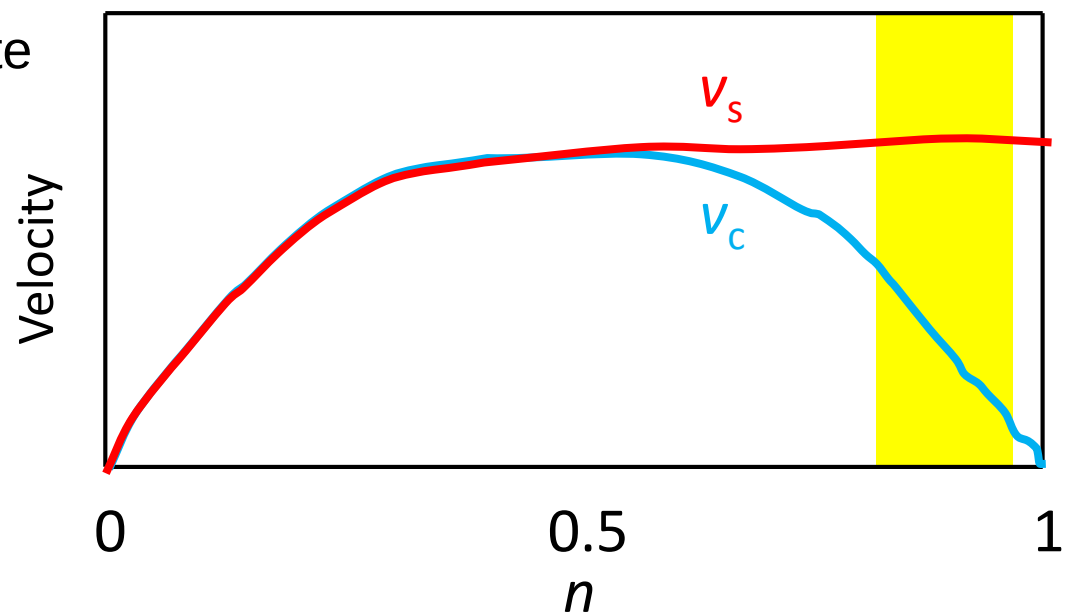
Spin and charge velocities in the Hubbard-type model

1D: spin-charge separation



2D: approximate spin-charge separation

[T. T. and S. Maekawa, JPSJ **65**, 1902 (1996)]



Summary

1. Electron-hole asymmetry in cuprates

2. Q-dependent “incoherent” charge excitations

Resonant inelastic x-ray scattering (RIXS)

K. Ishii, M. Fujita, T. T. *et al*., Nat. Commun. **5**, 3714 (2014)
T.T., J. Electro. Spector. and Related Phenome., **200**, 209 (2015)

3. Q-dependent “coherent” charge excitations

Theoretical prediction

Density-matrix renormalization group (DMRG)

Low-energy charge mode, detectable by RIXS

T. T., K. Tsutsui, M. Mori, S. Sota, S. Yunoki, Phys. Rev. B **92**, 014515 (2015)

Perspectives

O K-edge RIXS for charge mode