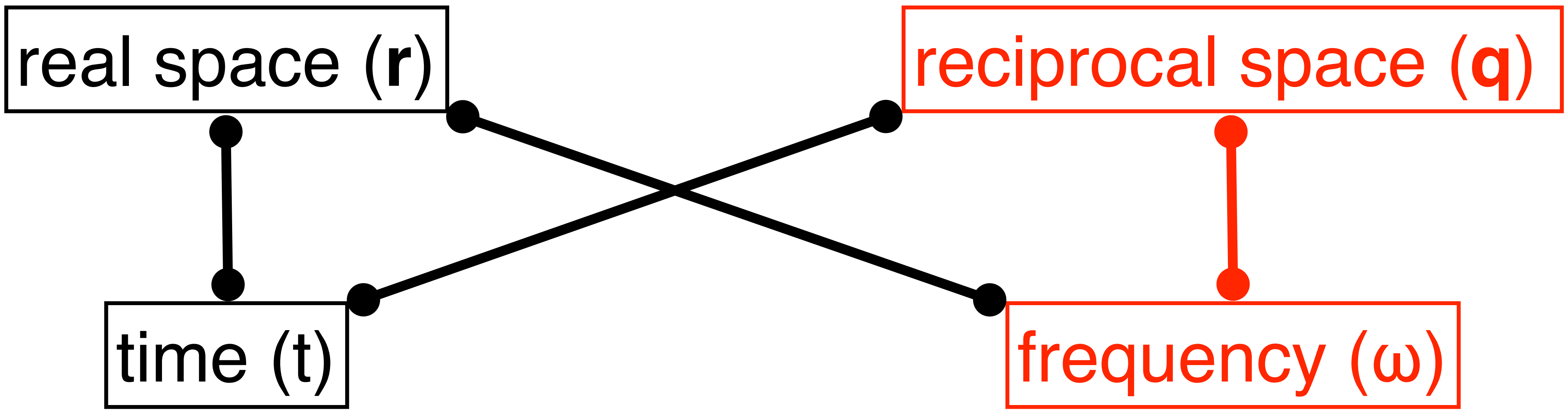
X線非弾性散乱による
電子の動的構造の研究

石井賢司
原子力機構

Study of electron dynamics

Choice of 3+1 dimensions



Conventional inelastic scattering

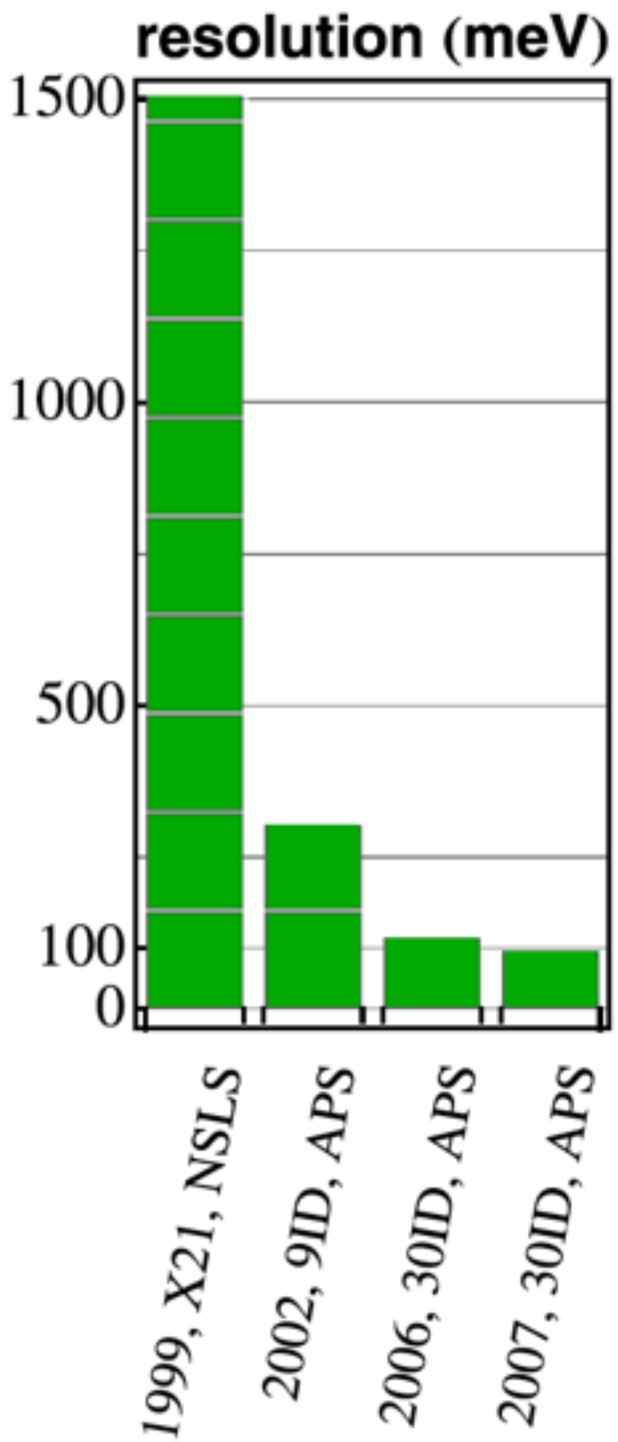
$$S(\mathbf{Q},\omega) \sim [V(\mathbf{Q})]^2[1-e^{\beta\omega}]^{-1}\mathrm{Im}\chi(\mathbf{Q},\omega)$$

- $S(\mathbf{Q},\omega)$: dynamical structure factor
- $V(\mathbf{Q})$: interaction of probe
- $\chi(\mathbf{Q},\omega)$: response function (susceptibility)

At present, resonant enhancement is necessary for electronic excitations → **RIXS**

Progress of energy resolution

hard x-ray (Cu K-edge)

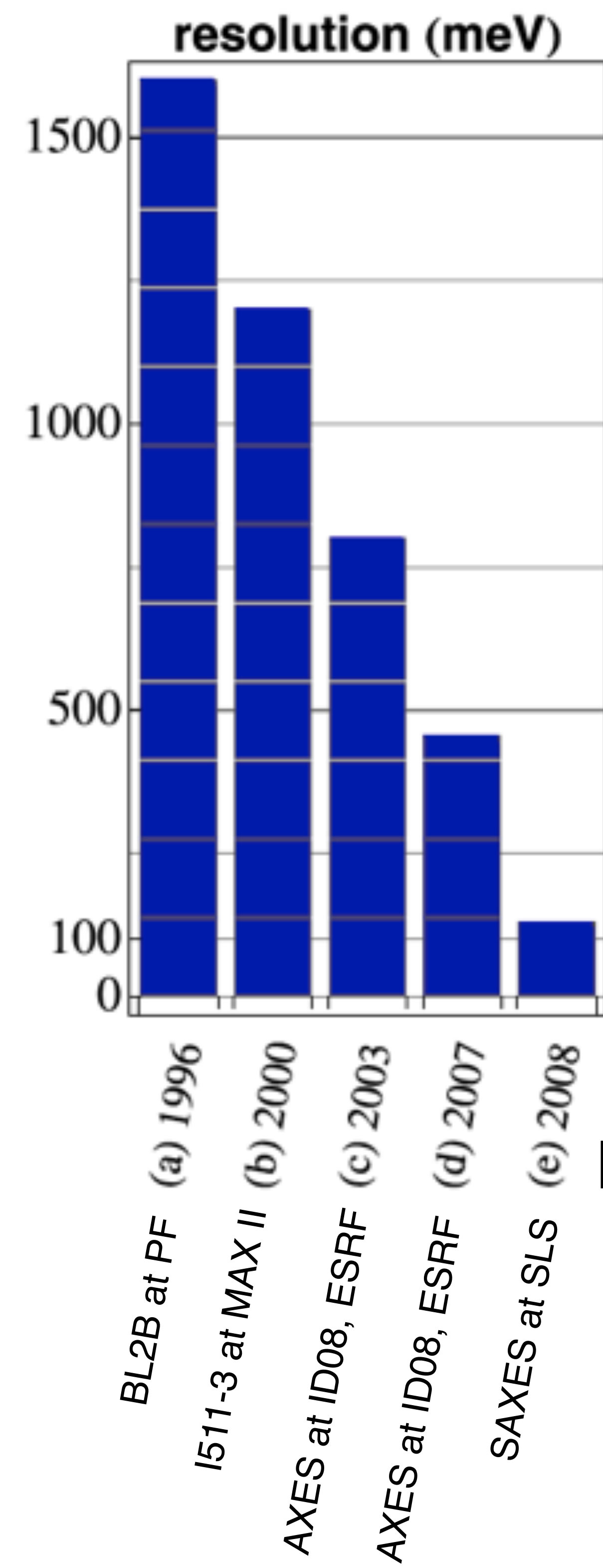


P01, PETRA III
BL12XU, SPring-8

25 meV !!

(30 meV at Ir L_3-edge)

soft x-ray (Cu L_3-edge)



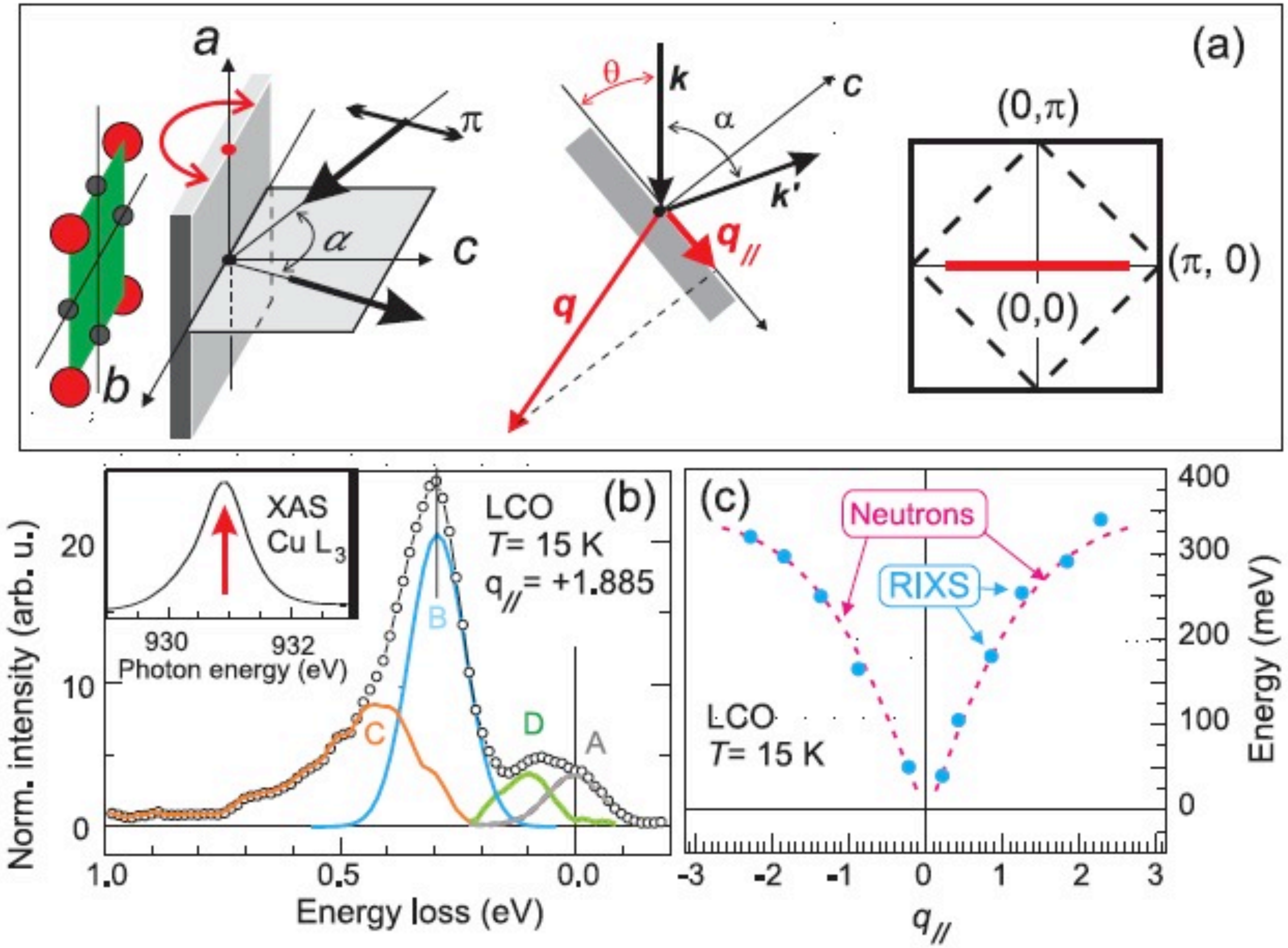
ID32, ESRF

22 meV !!

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

Biggest breakthrough – Spin excitations in RIXS –

Single magnon in La_2CuO_4

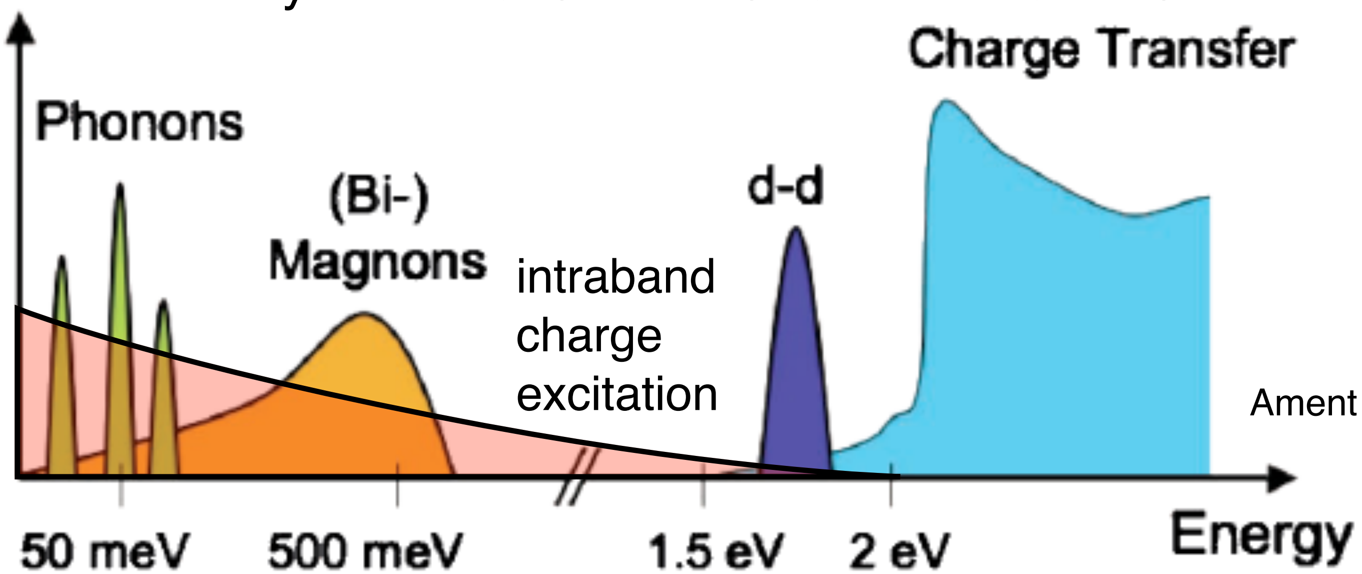


A: elastic
D: optical phonon
B: single magnon ←
C: multiple magnon

L.Braicovich et al., PRL **104**, 077002 (2010).

Electronic excitations observed by RIXS

Elementary excitations in transition metal oxides



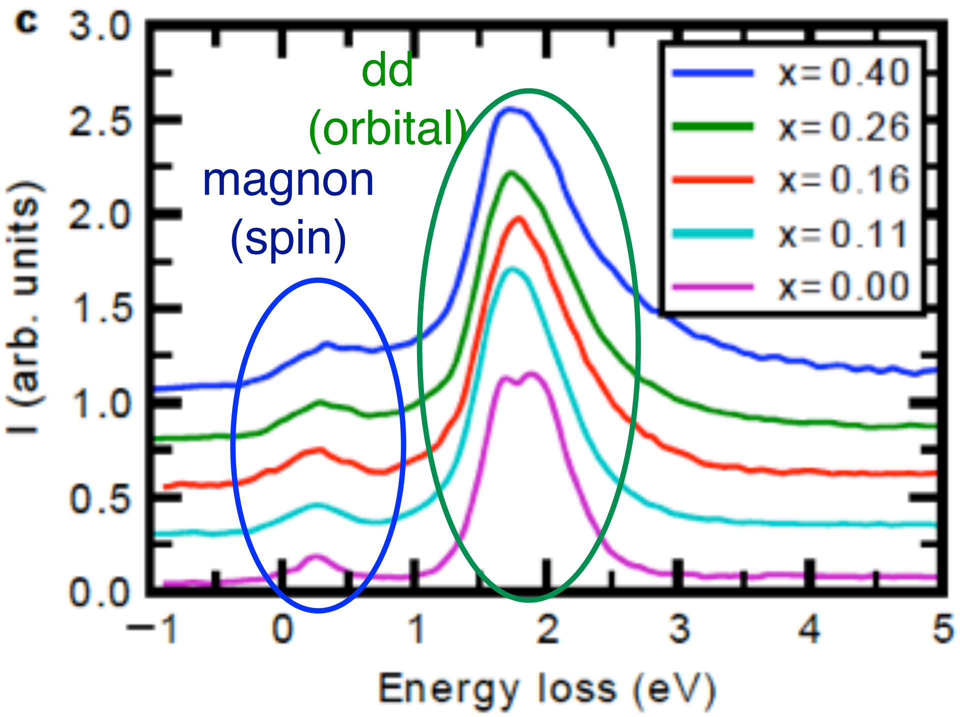
Ament et al., Rev. Mod. Phys. **83**, 705 (2011)

energy scale in cuprates: J, t, 10Dq, Δ, U

sensitive to **charge**, **spin**, and **orbital**

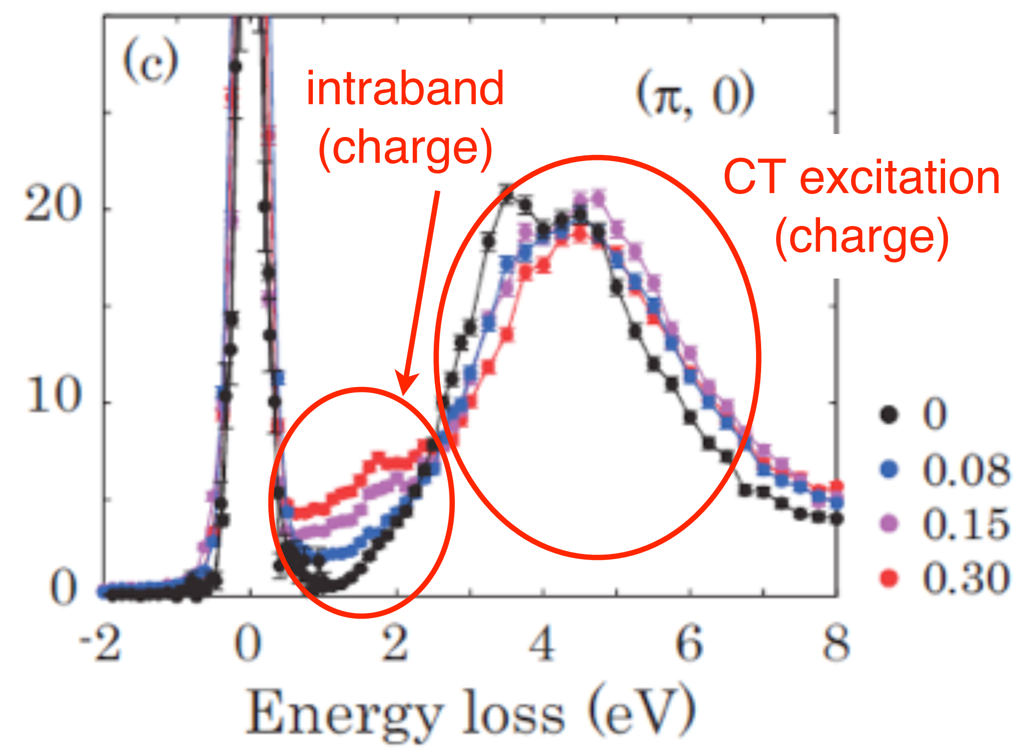
RIXS spectra of $La_{2-x}(Ba,Sr)_xCuO_4$

L-edge



Dean et al., Nat. Mater. **12**, 1019 (2013)

K-edge



Wakimoto et al., Phys. Rev. B **87**, 104511 (2013)

Recent topics (presented in this talk)

1. Layered high-T_c cuprates
   spin excitations + **charge** excitations
   共同研究者: 藤田全基, 佐々木隆了, M. Minola, G. Dellea, C. Mazzoli,
   K. Kummer, G. Ghiringhelli, L. Braicovich, 遠山貴己,
   山田和芳, 吉田雅洋, 黒岡雅仁, 清水裕友, 水木純一郎
   佐藤研太朗, 宮脇淳, 原田慈久, J. Pelliciari, Y. Huang, T. Schmitt

2. Layered iridates
   (pseudo-)**spin** excitations coupled to **charge**
   共同研究者: I. Jarrige, 吉田雅洋, 水木純一郎, 松本章代, 加藤晃彦,
   高山知弘, 高木英典

3. Layered iridate
   exciton in **spin**-**orbit** entangled states (d-d excitation)

4. 1-dimensional cuprate
   spin-**orbital** separation (d-d excitation)

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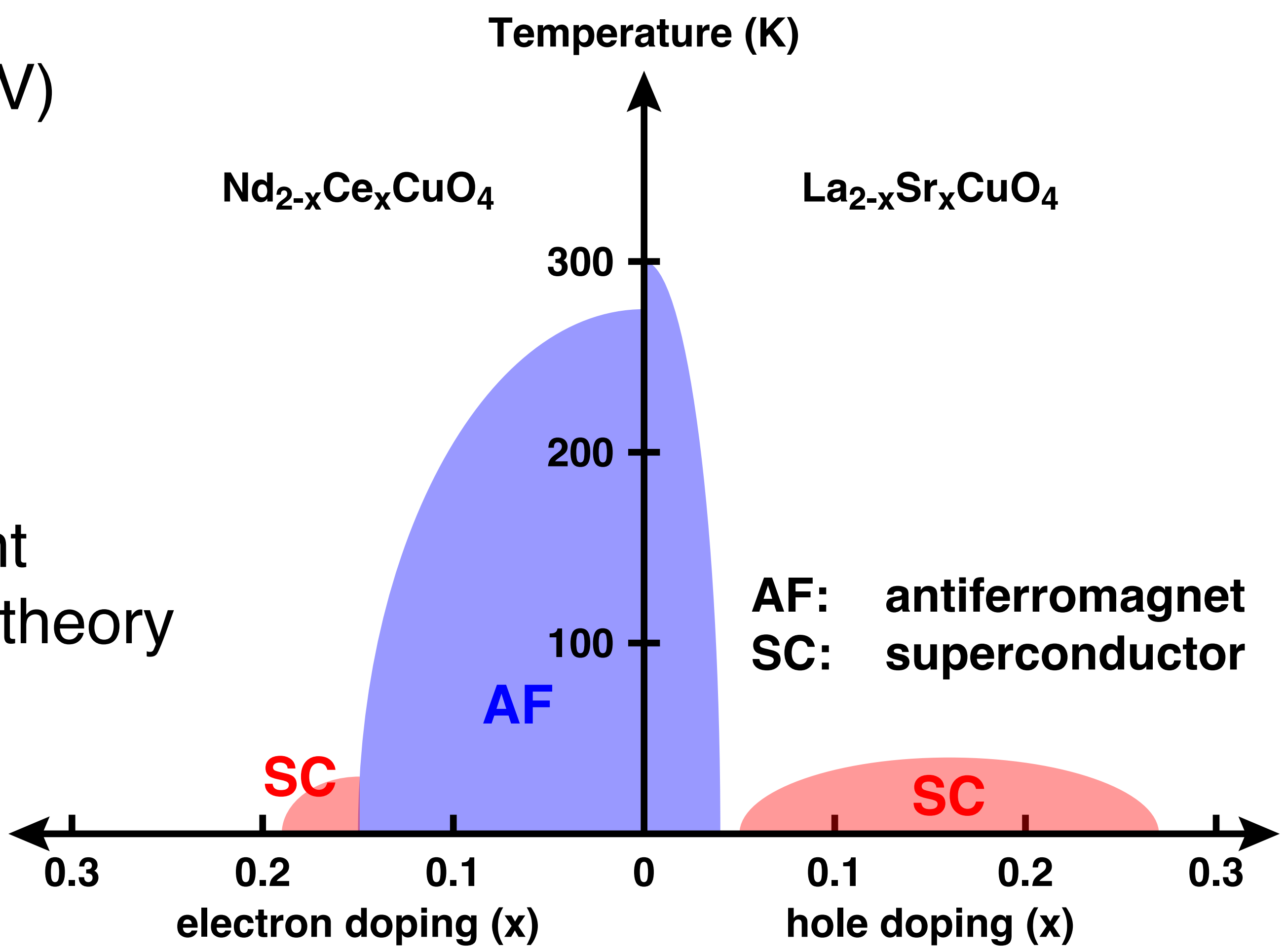
Characteristics of copper oxides

- high-Tc superconductivity
  large energy scale (T_c ~ 100 K, J ~ 100 meV)
  → Excitations are detectable
  with inelastic x-ray scattering

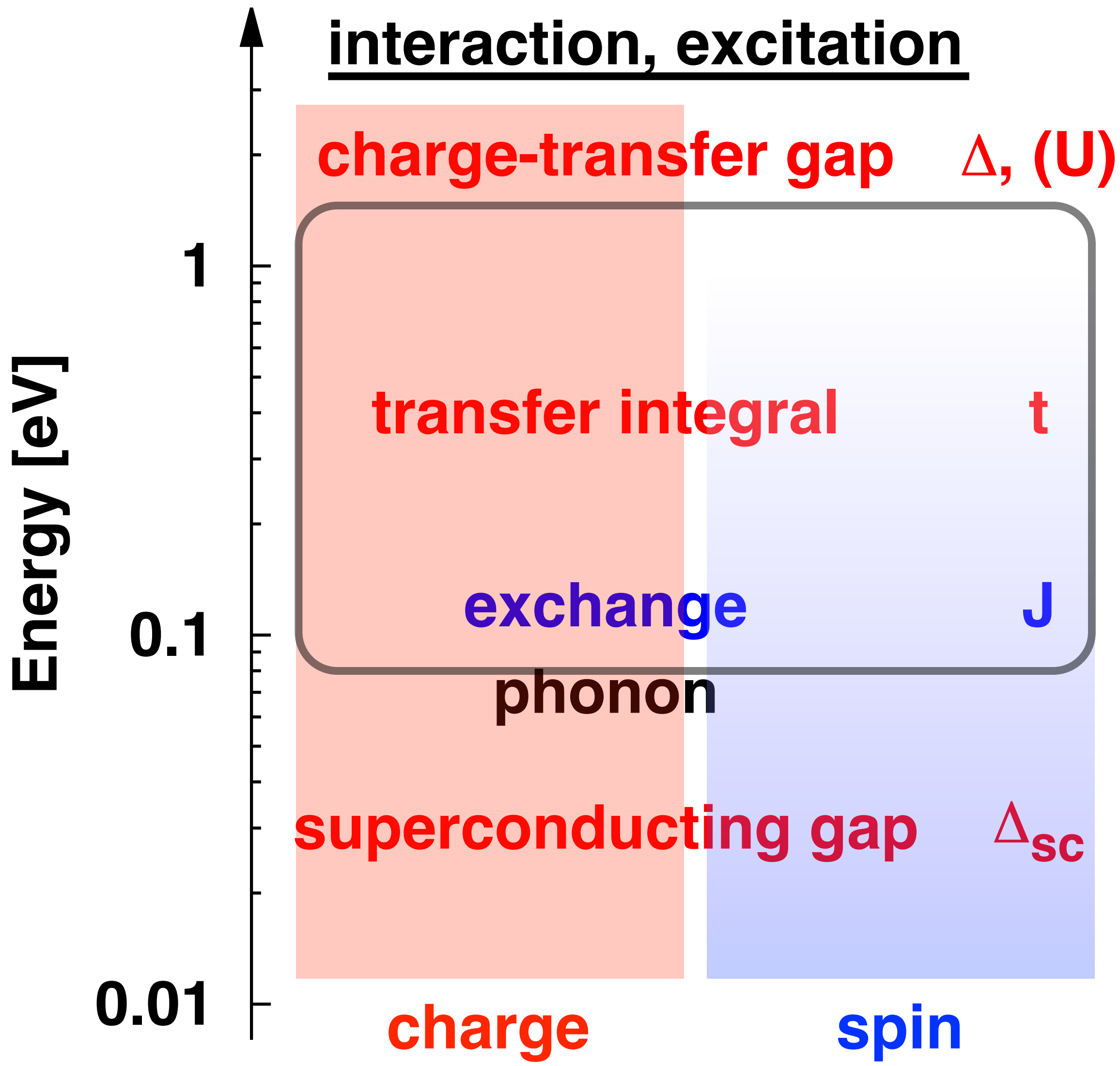
- simple electronic structure
  CuO_2 square lattice
  only a Cu $3dx^2$-y^2 (+ O $2p_x$, $2p_y$) are relevant
  → exact treatment of many-body effect in theory

- both electron and hole can be doped
  well-understood undoped limit

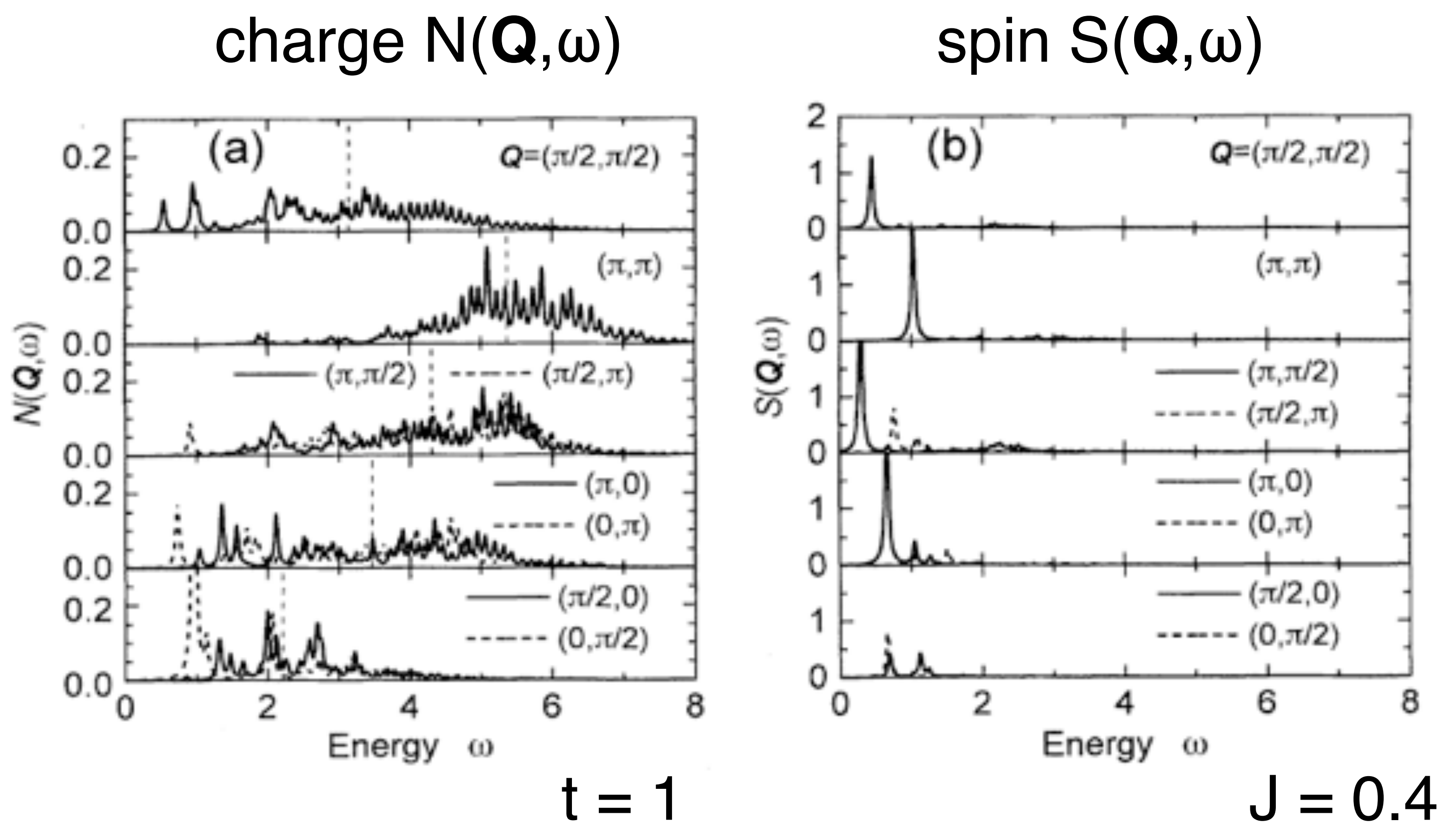


A suitable system for the study of spin and charge excitations
in correlated electron systems

Spin and charge excitations in cuprates



dynamical correlation in t-J model



T. Tohyama et al., Phys. Rev. Lett. **74**, 980 (1995)

Electronic excitations at sub-eV

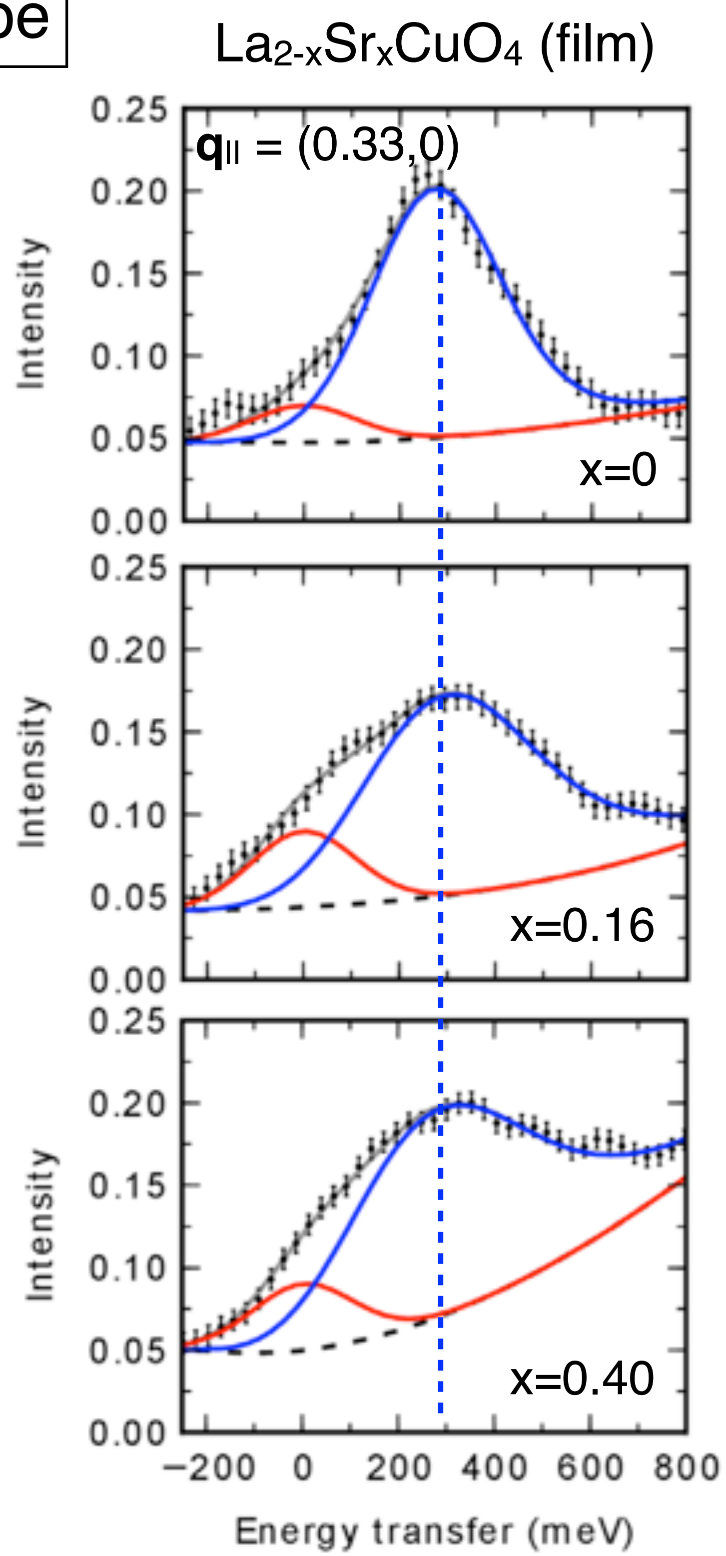
important for comprehensive understanding of electron dynamics in copper oxides

charge dynamics (~ t), spin dynamics (~ J)

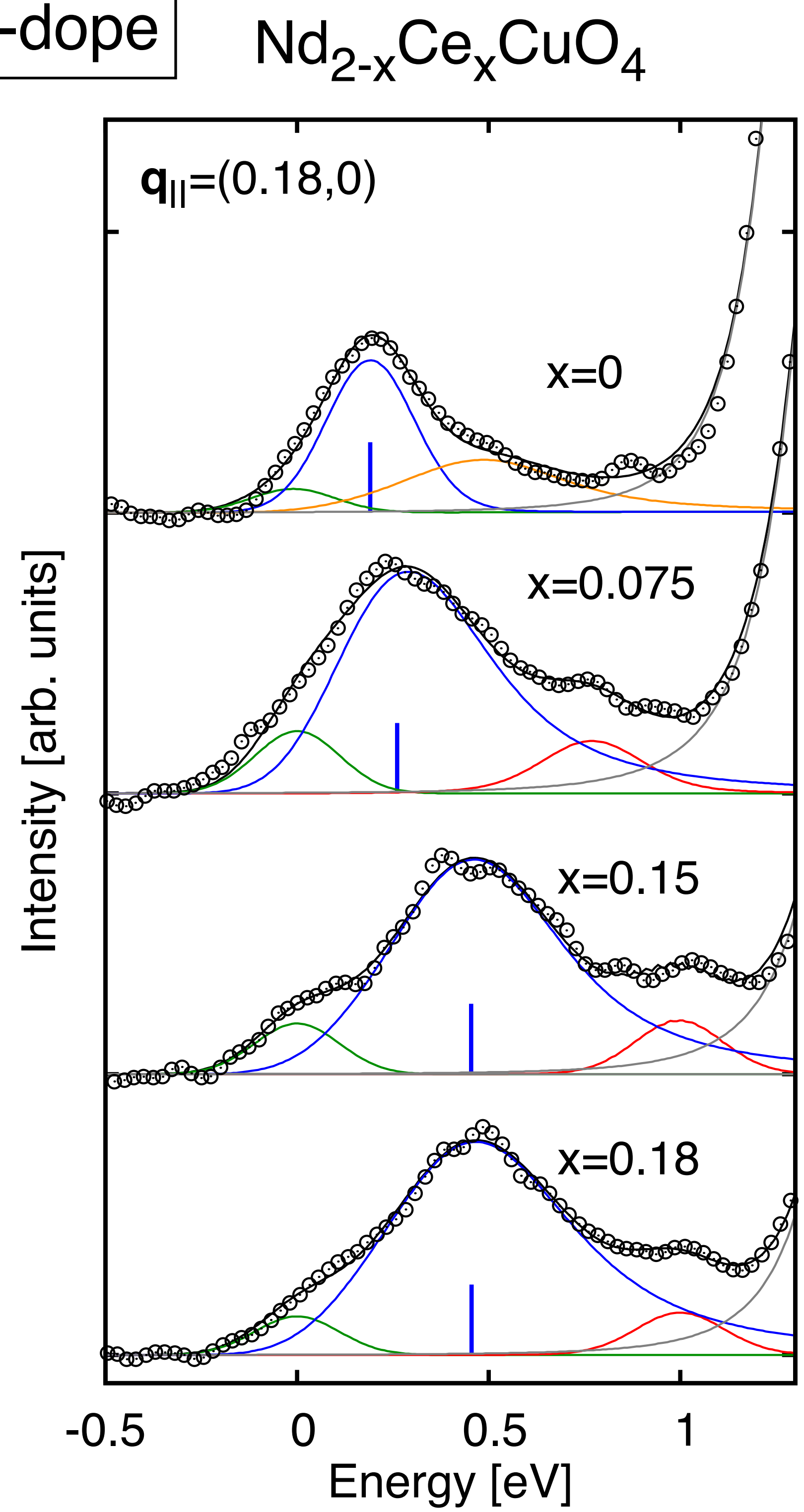
- evolution of spin dynamics upon carrier doping
- charge dynamics of doped carrier

Spin excitations (Cu L_3-edge RIXS)

h-dope



e-dope



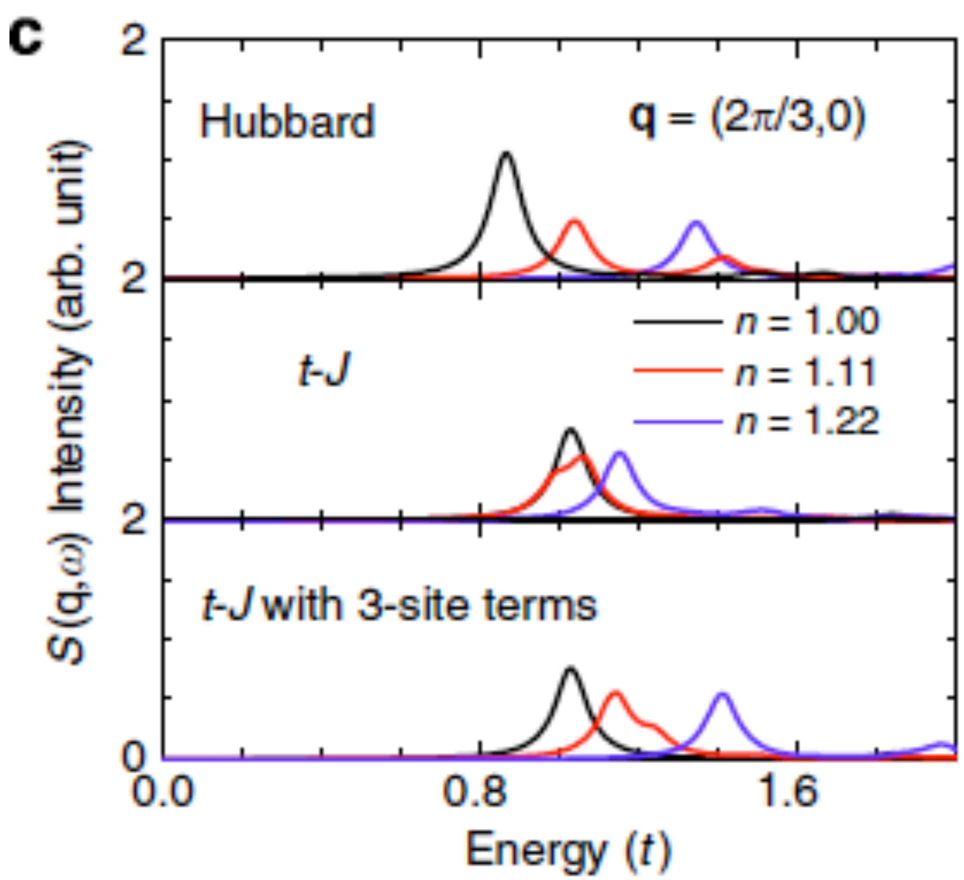
unchaged peak position

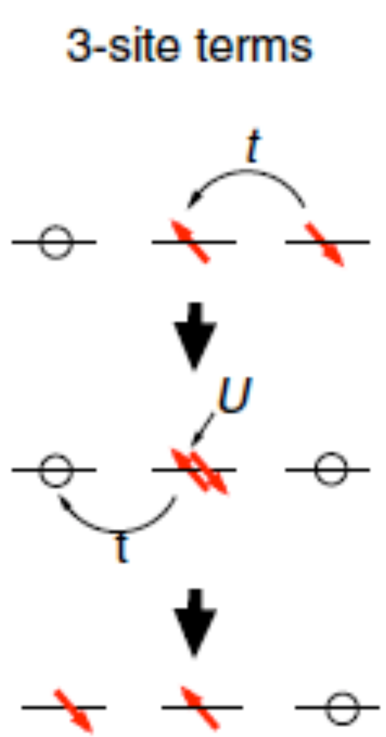
shift to higher energy

Dean et al., Nat. Mater. **12**, 1019 (2013)

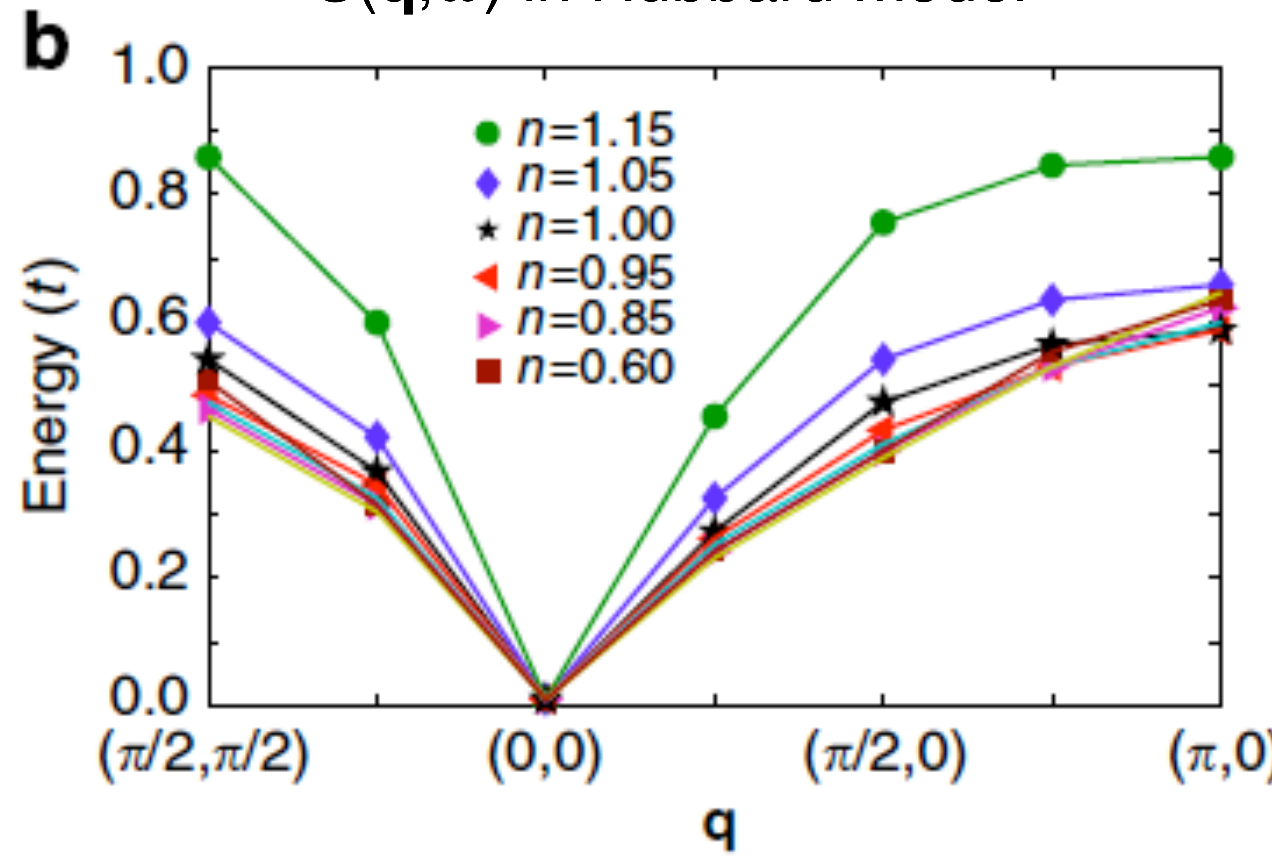
Ishii et al., Nat. Commun. **5**, 3714 (2014)

Recent theory





S(**q**,ω) in Hubbard model

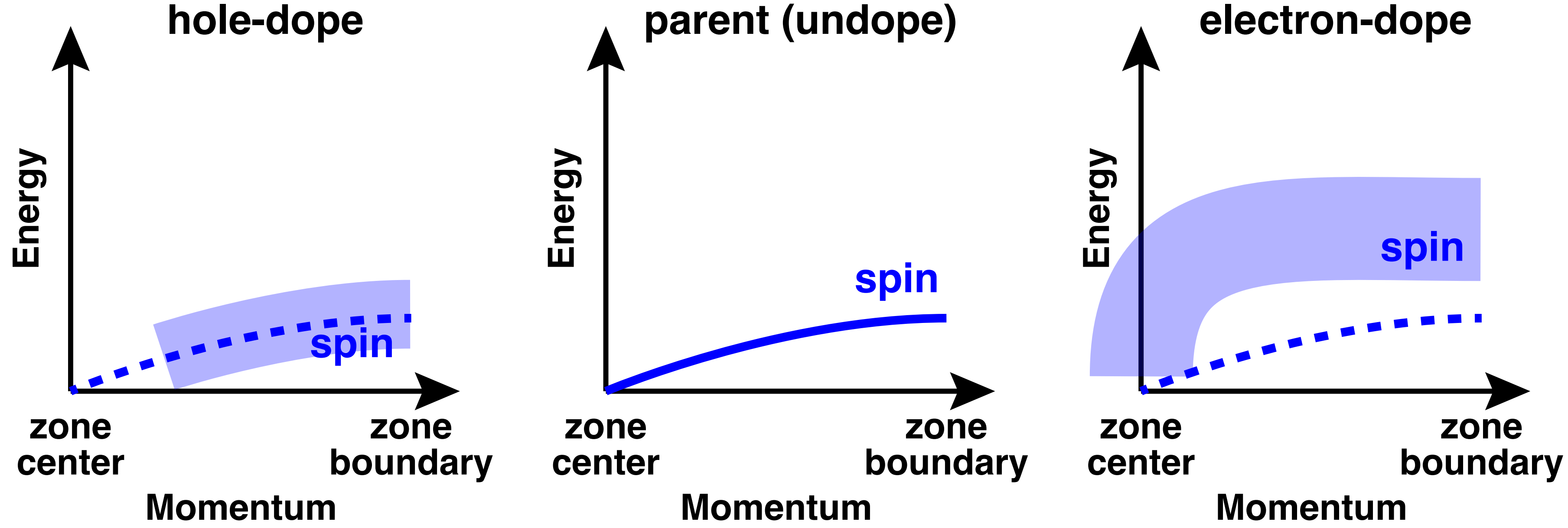


To explain high-energy shift upon electron doping, 3-site term is needed in the t-J model.

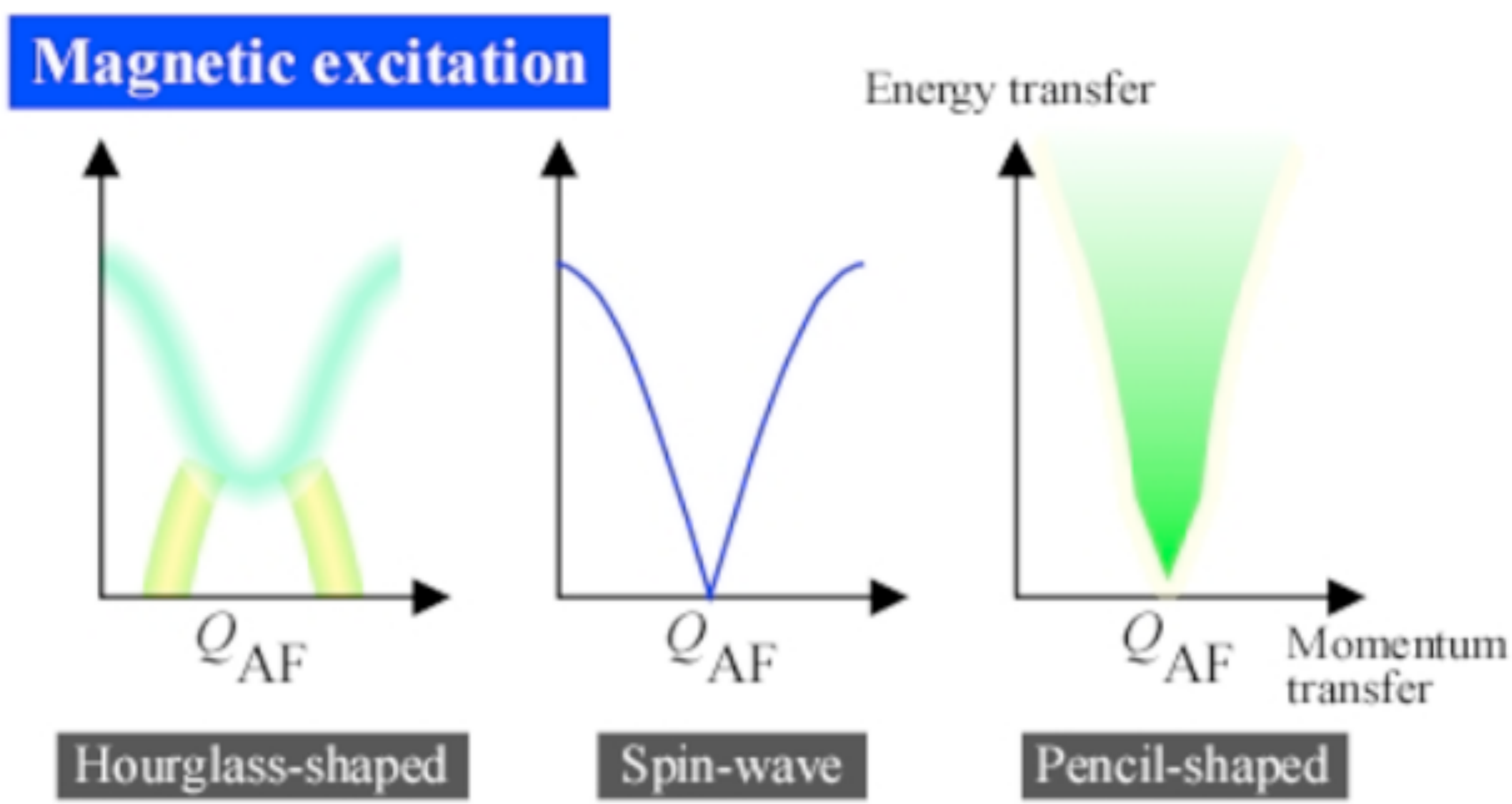
Jia et al., Nat. Commun. **5**, 3314 (2014)

Summary of spin excitations in cuprates

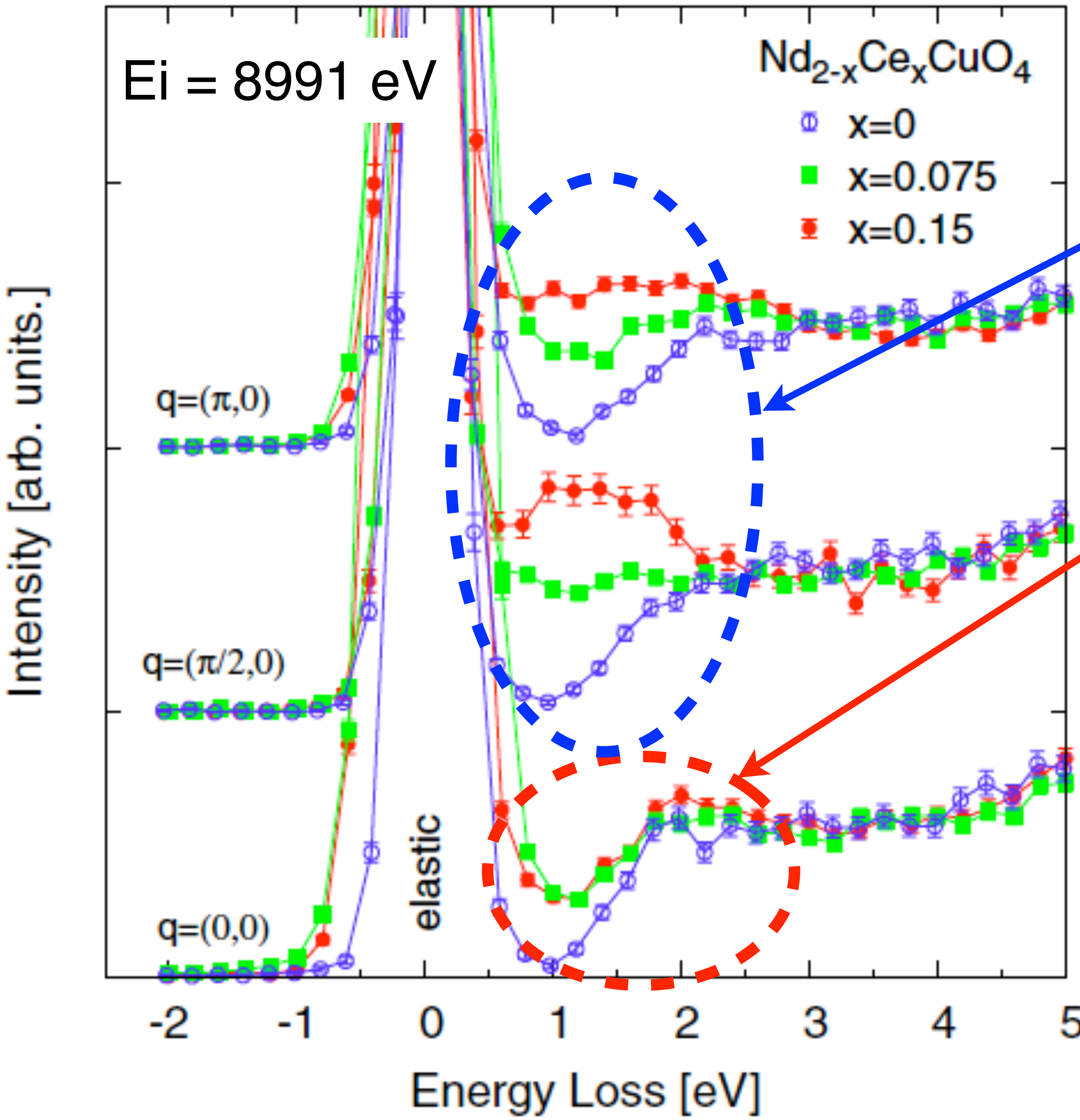
high-ω
(RIXS)



low-ω
(INS)



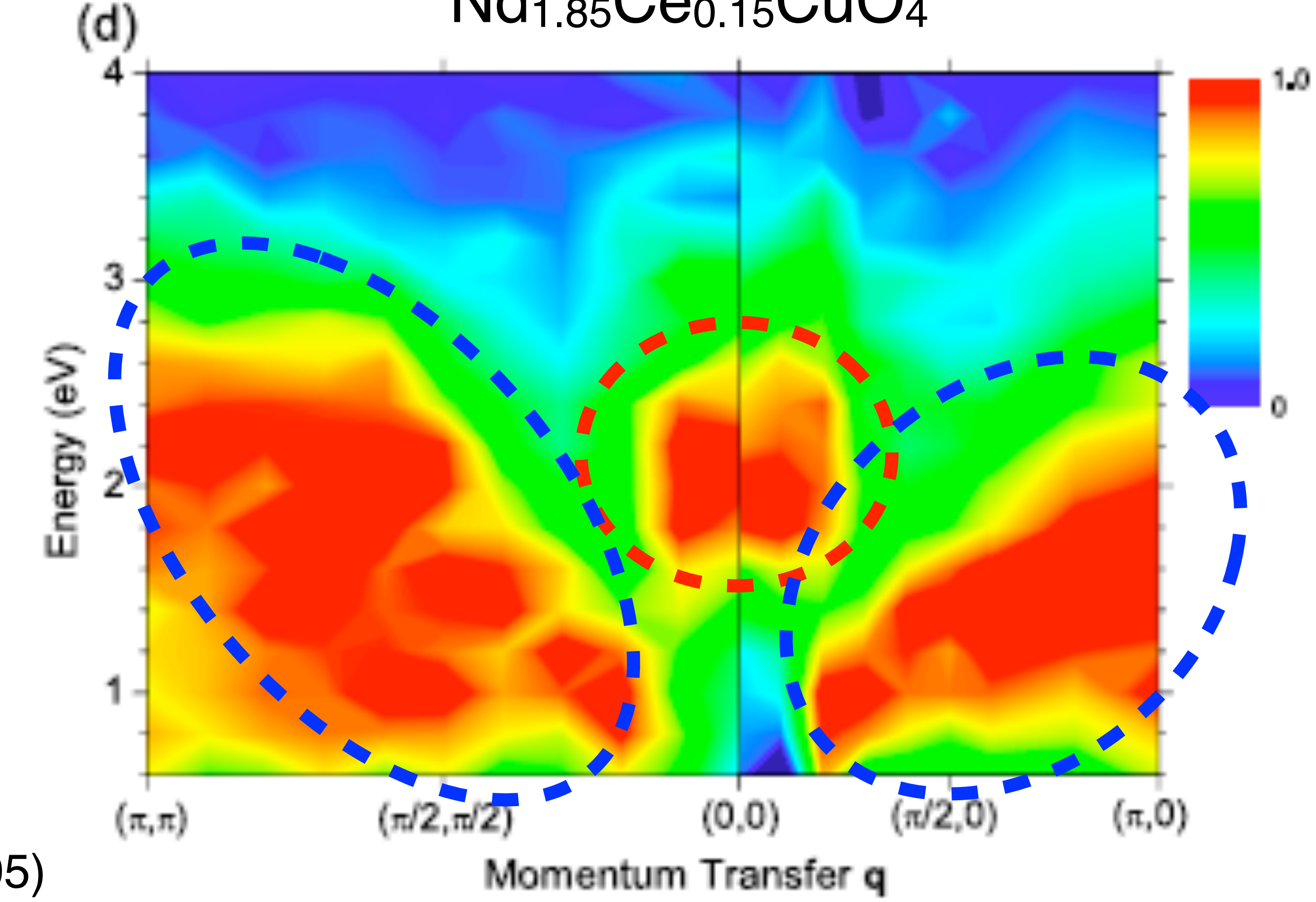
Charge excitations in e-doped NCCO (Cu K-edge RIXS)



Excitations at finite **q**
intensity proportional to x
→ intraband charge excitation

Excitation at **q** = (0,0)
doping independent
→ interband charge excitation
across the Mott gap

$Nd_{1.85}Ce_{0.15}CuO_4$

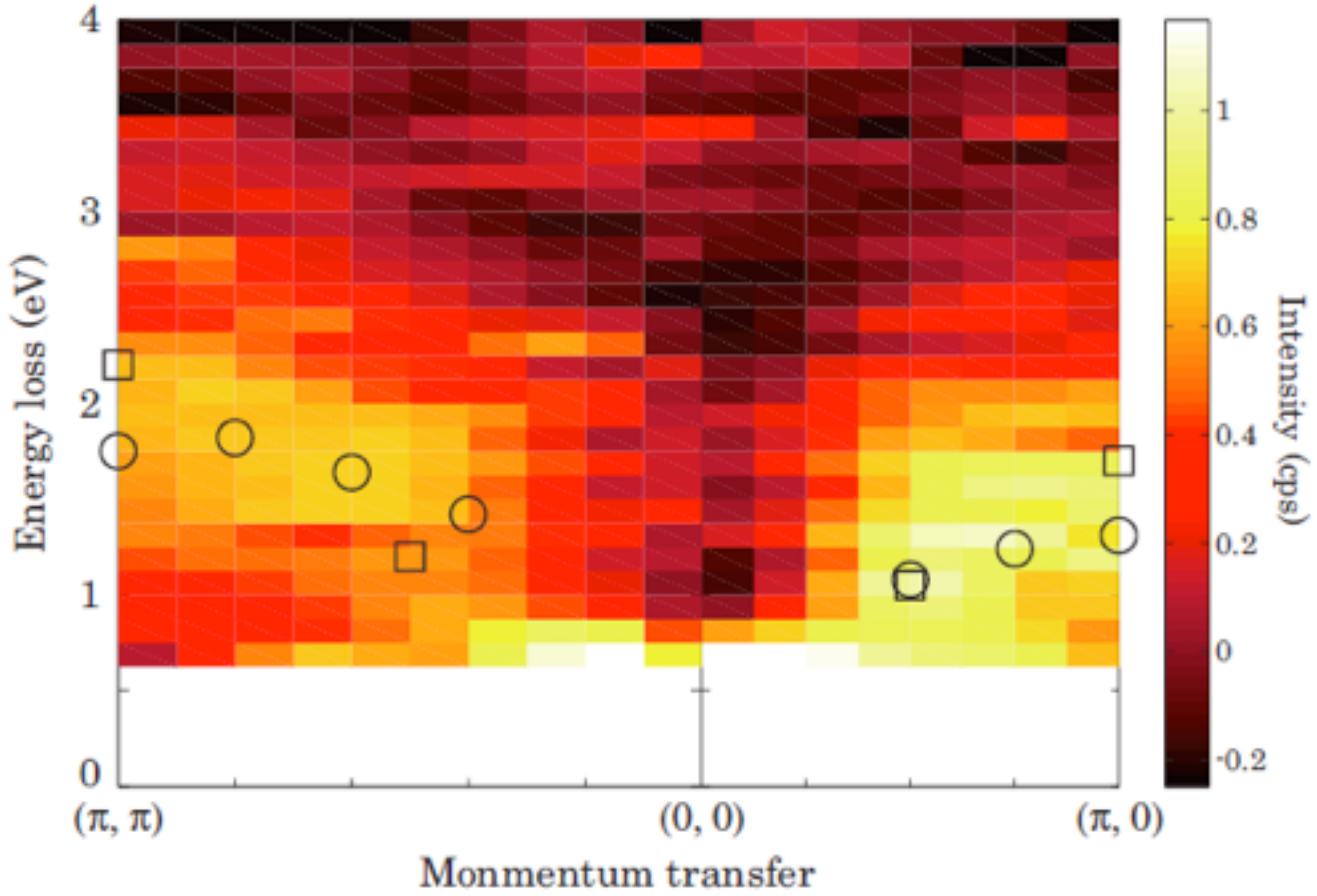


Dispersive charged excitations appear when electrons are doped.

K. Ishii et al.,PRL **94,** 207003 (2005)

Charge excitations in h-doped LSCO (Cu K-edge RIXS)

$La_{2-x}Sr_xCuO_4$ (x=0.3) E_i = 9003 eV



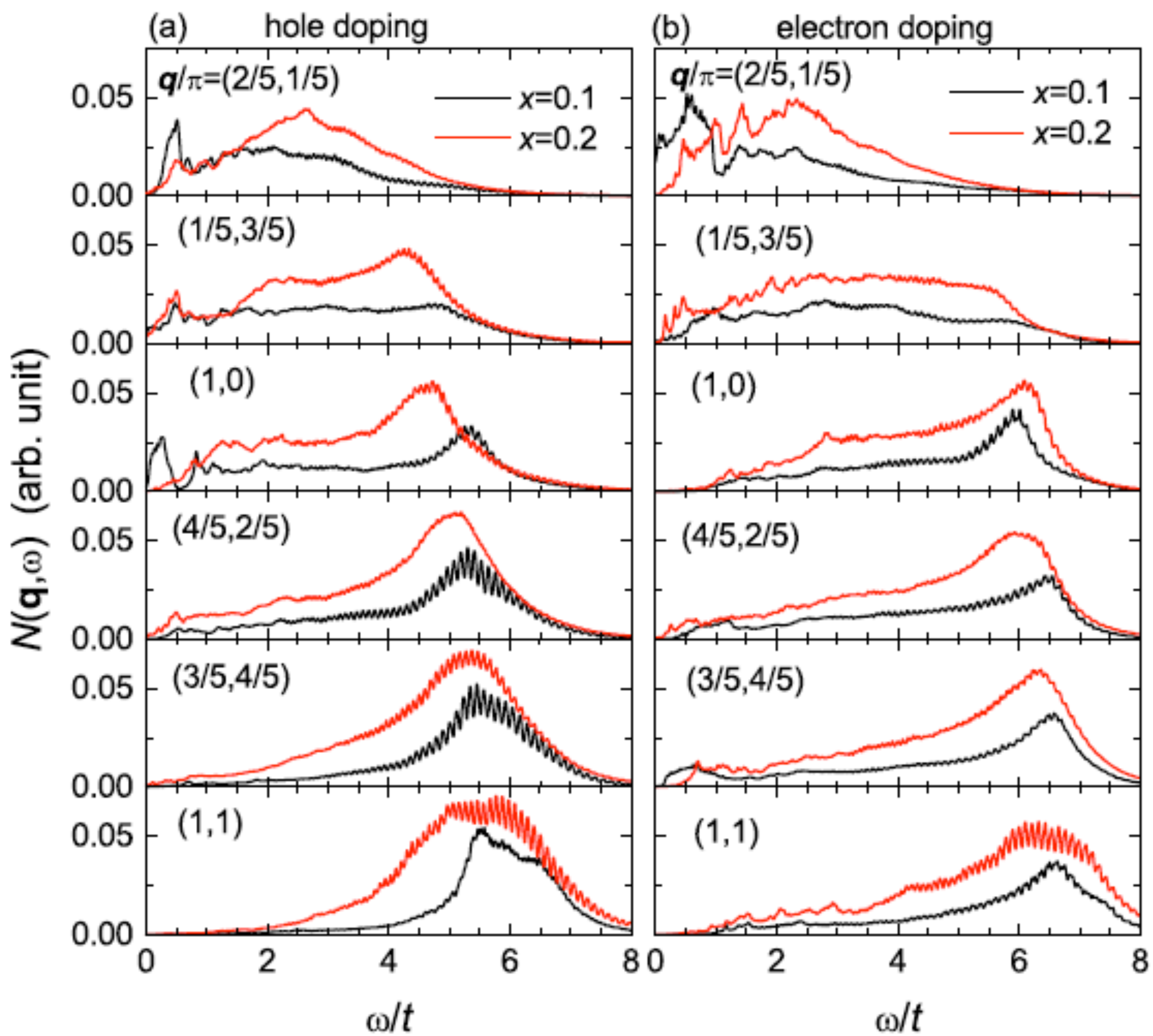
circle: LSCO x=0.30 (hole dope)
square: NCCO x=0.15 (electron dope)

Momentum dependence is similar to electron-doped NCCO.

S. Wakimoto & KI et al., PRB **87**, 104511 (2013)

Dynamical charge structure factor N(q,ω) in a theory

20 site t-t'-t"-J cluster

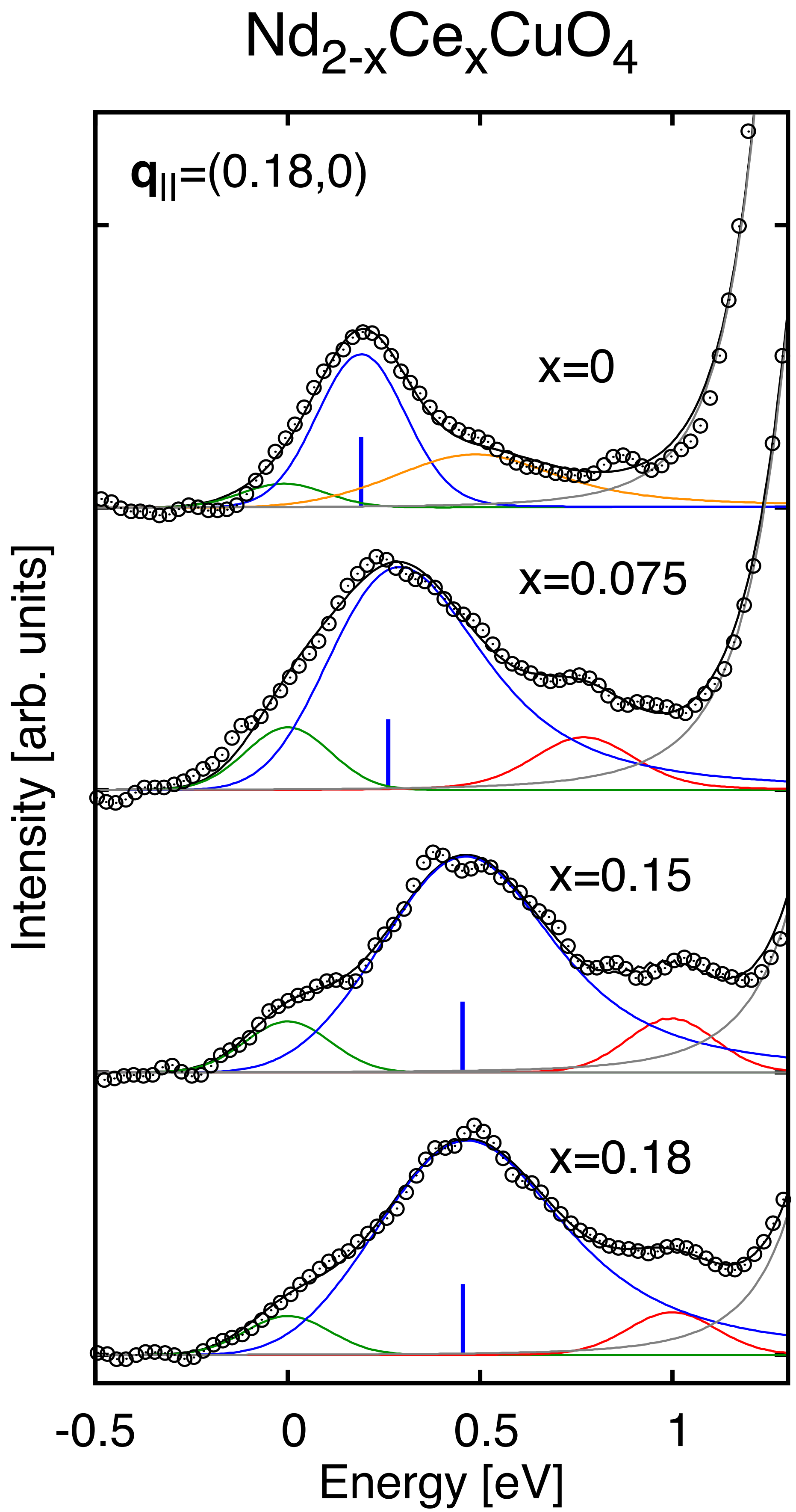


t ~ 0.4 eV

Above 1 eV,
N(q,ω) of h-dope and e-dope are similar.

Tohyama, J. Electron Spectrosc. Relat. Phenom., to be pubished

Charge excitations of e-doped NCCO (Cu L3-edge RIXS)



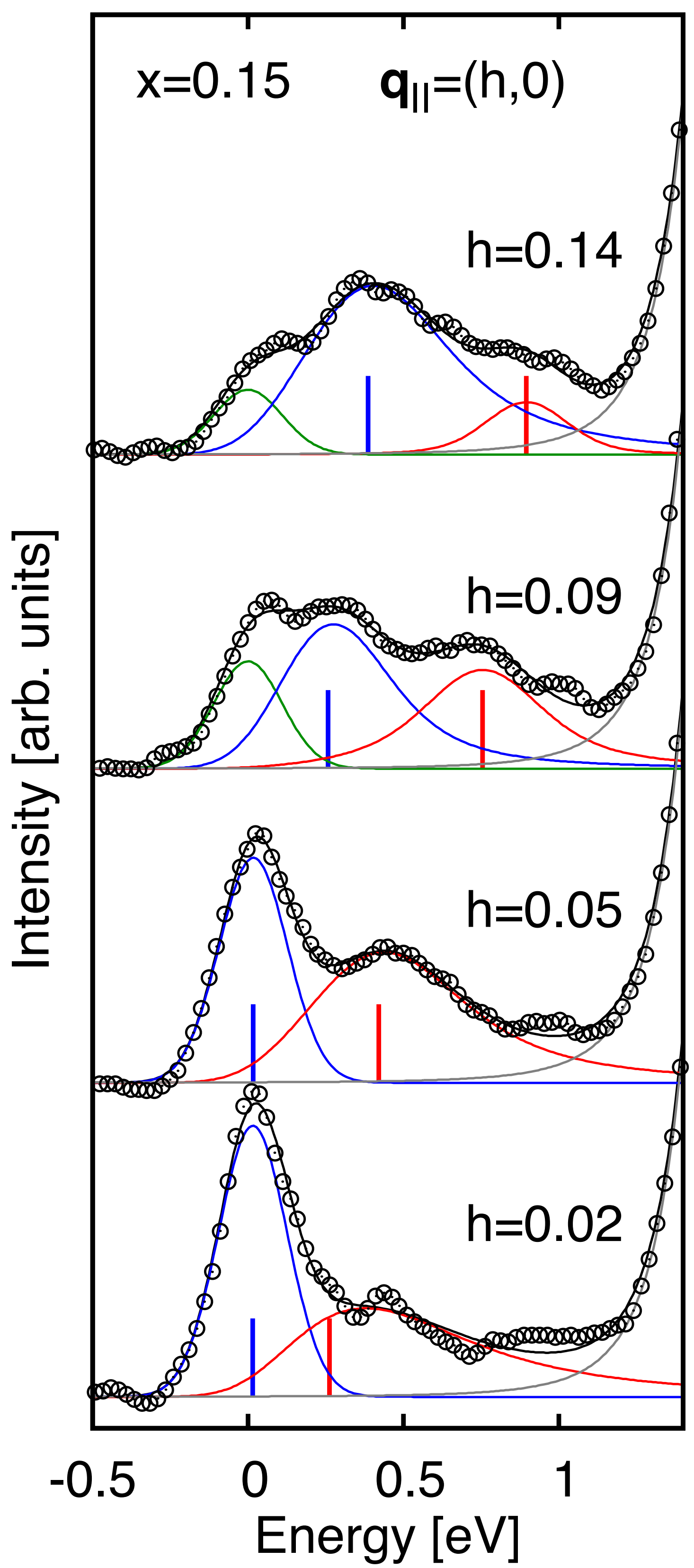
<u>parent (x = 0)</u>

elastic scattering (resolution-limited)
+ single magnon (resolution-limited)
+ multi-magnon
+ dd excitations

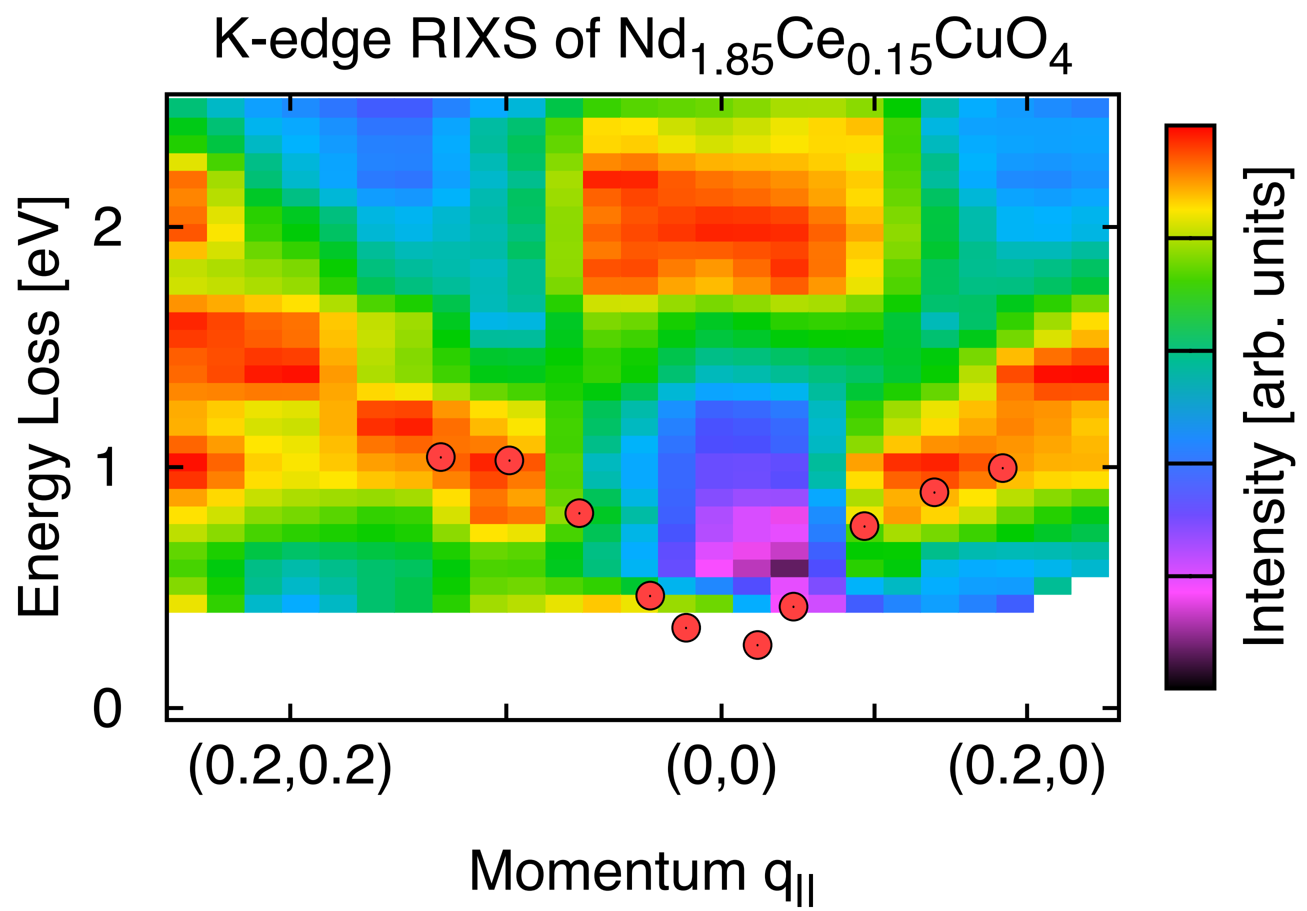
<u>electron-doped (x ≠ 0)</u>

elastic scattering (resolution-limited)
+ single magnon
+ additional mode (at low q)
+ dd excitations

Charge excitations in e-doped NCCO

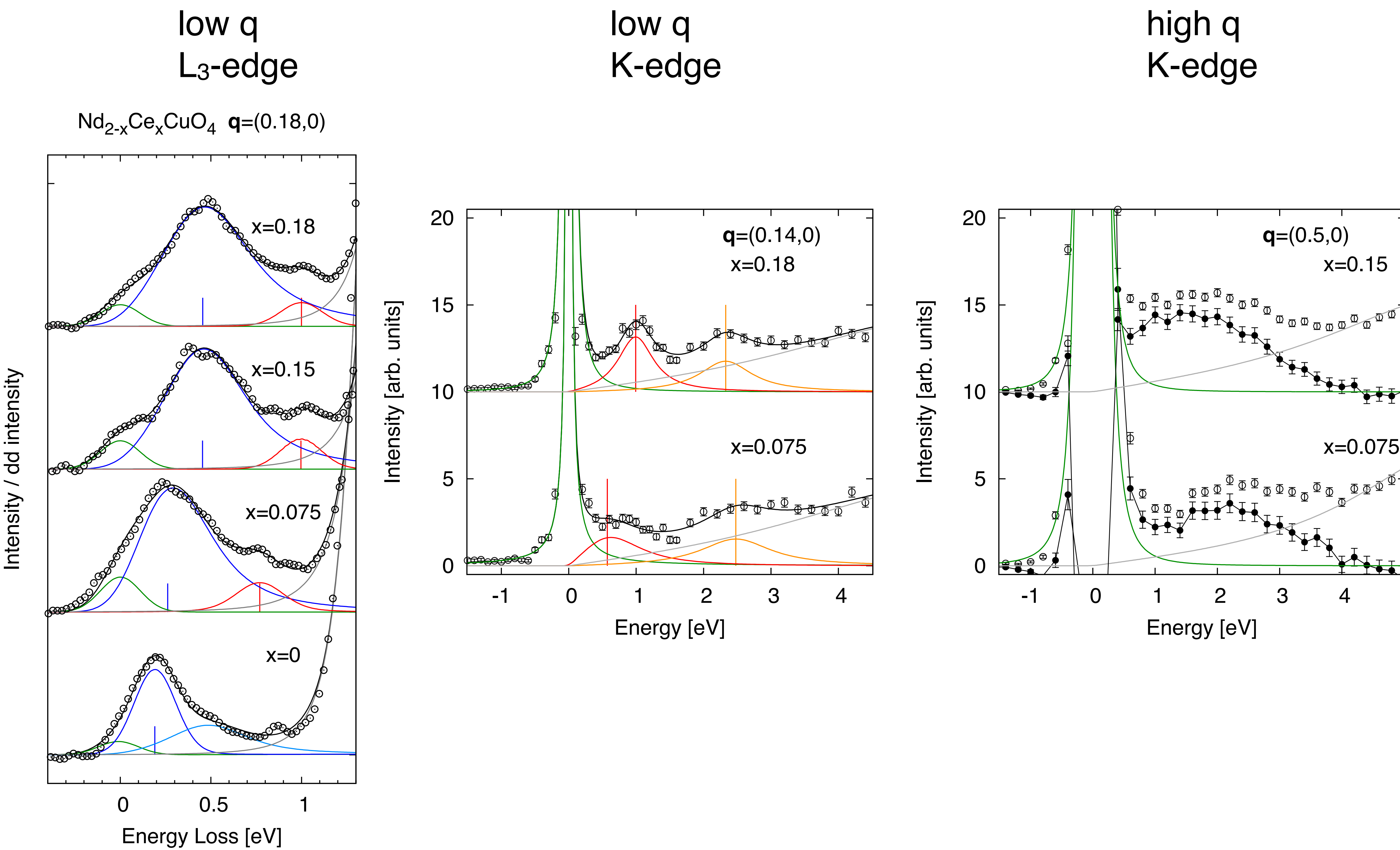


peak position
of
additional mode



Probably, additional mode has the same charge origin.

Doping dependence

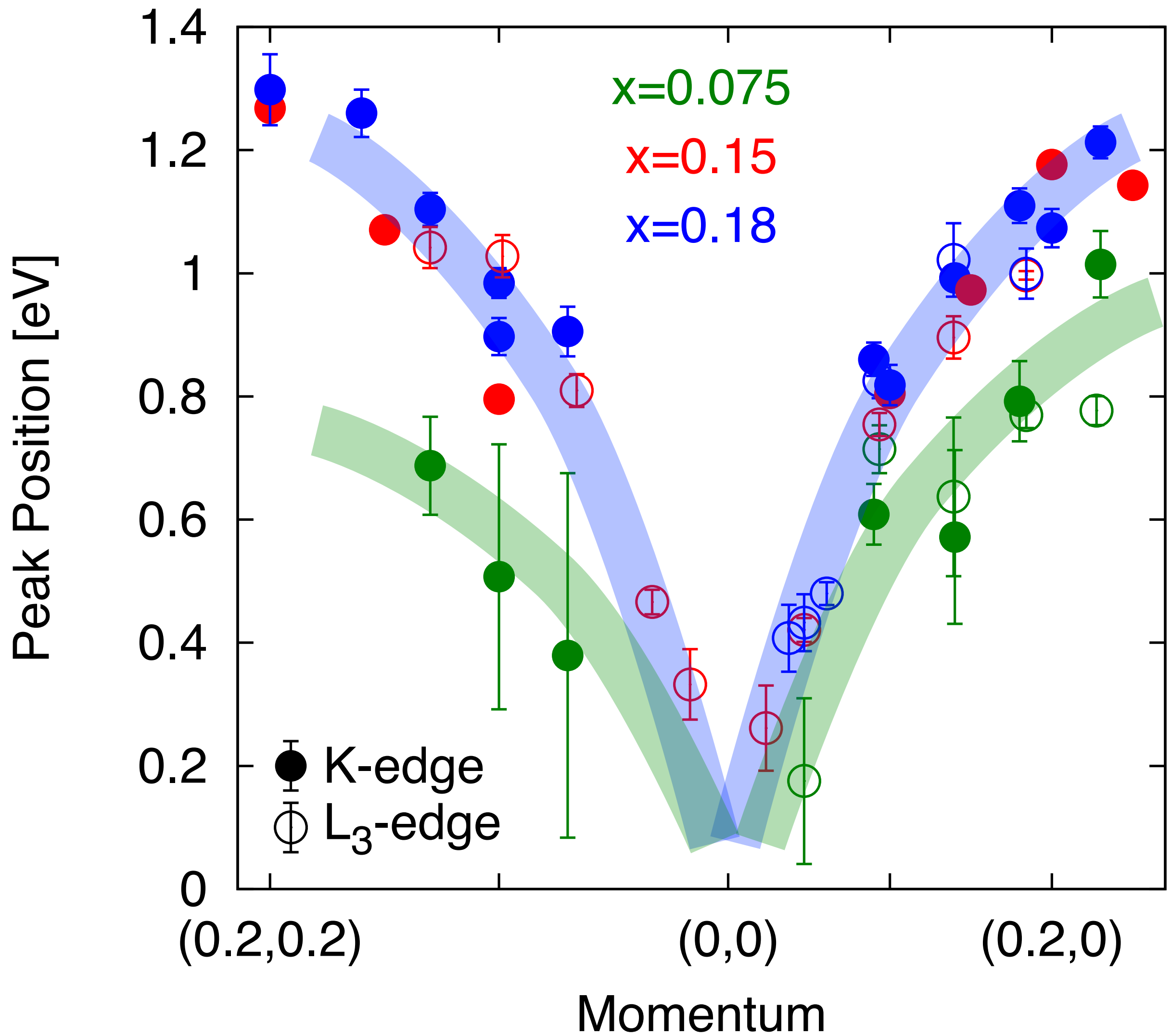


shift to high energy

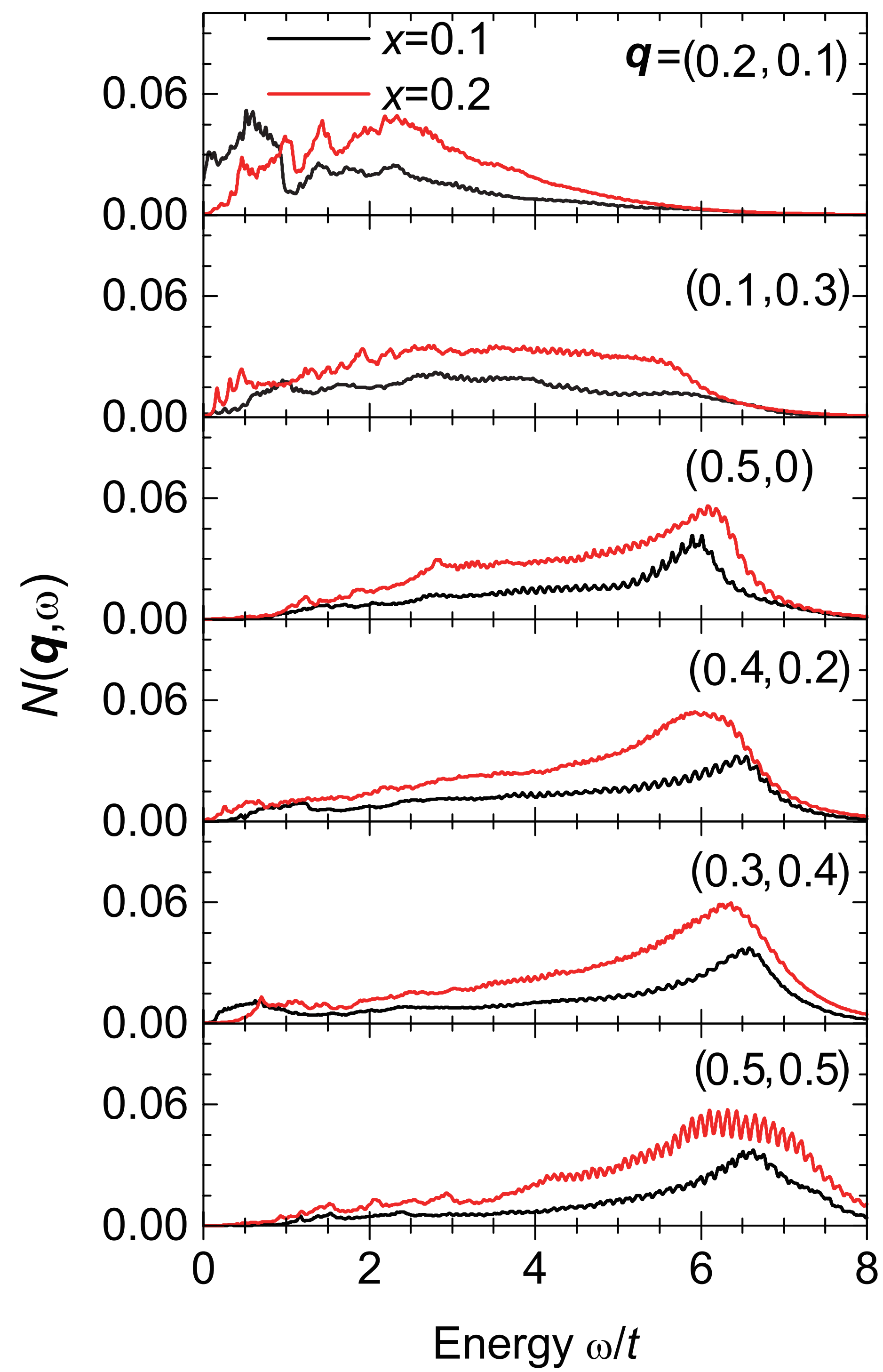
increase of intensity

Dispersion of charge excitations

low-**q**



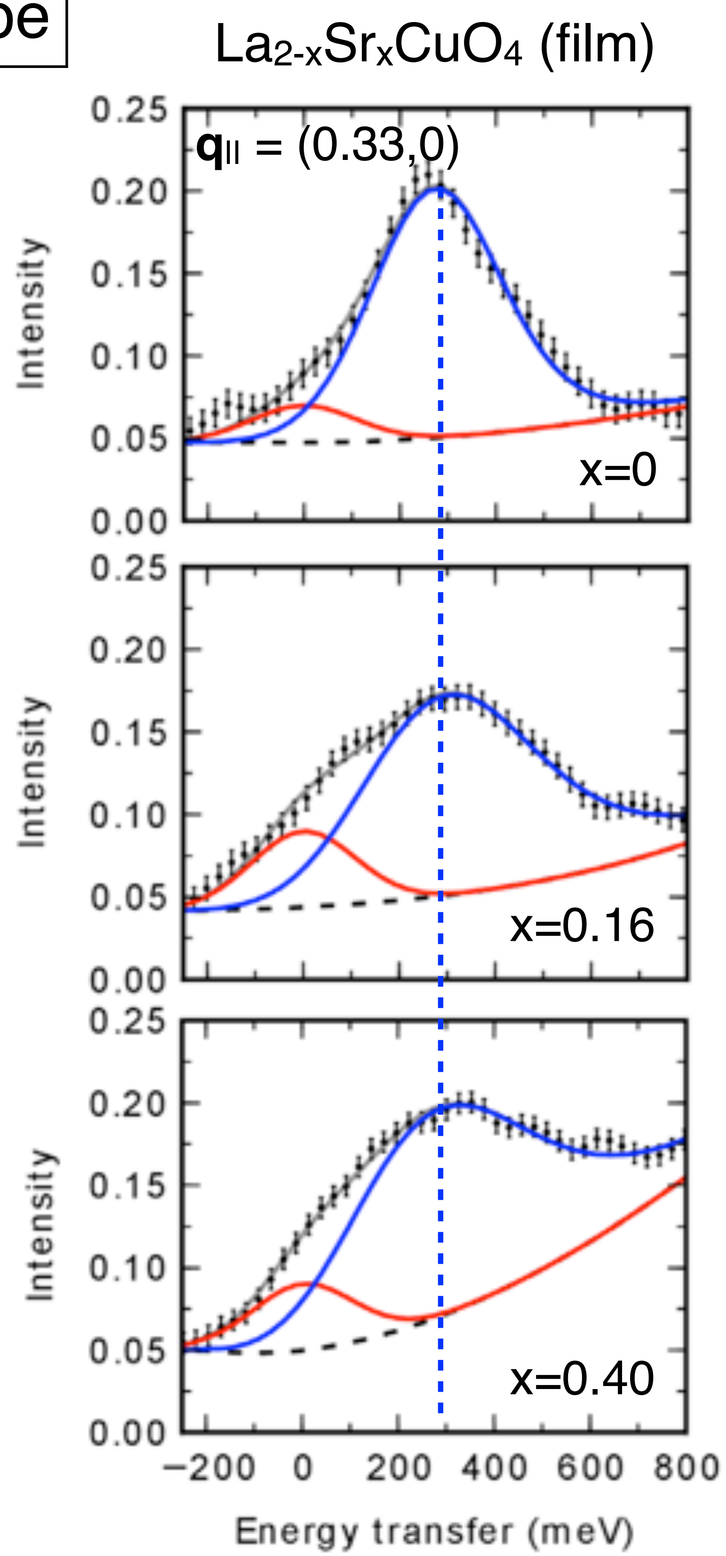
Dynamical charge correlation function N(**q**,ω)
Calculation t-t'-t"-J model



electron doping
→ low-q: high-energy shift of peak position
high-q: increase of spectral weight
(unchanged peak position)

Cu L_3-edge RIXS

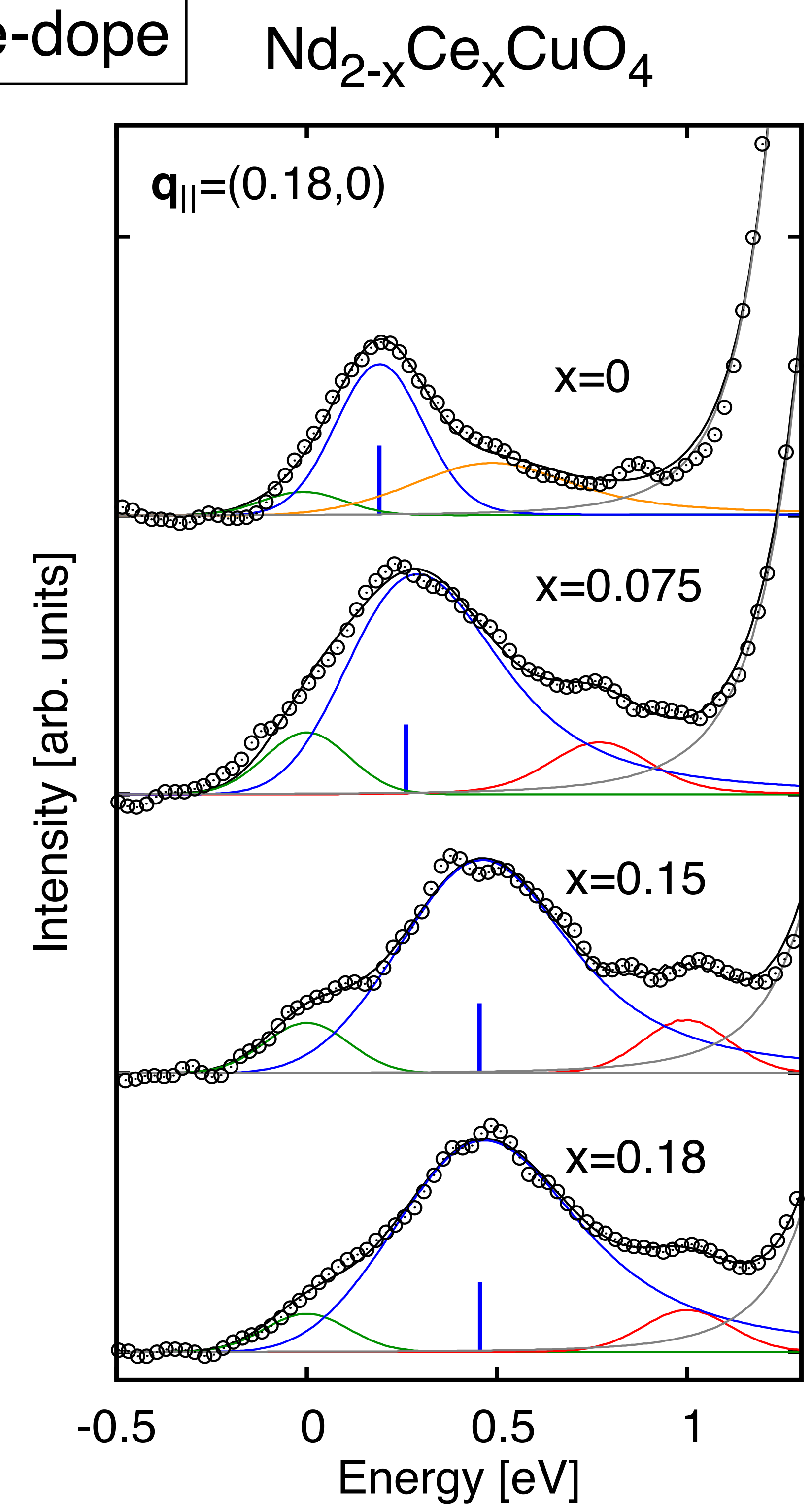
h-dope



magnetic excitations only

Dean et al., Nat. Mater. **12**, 1019 (2013)

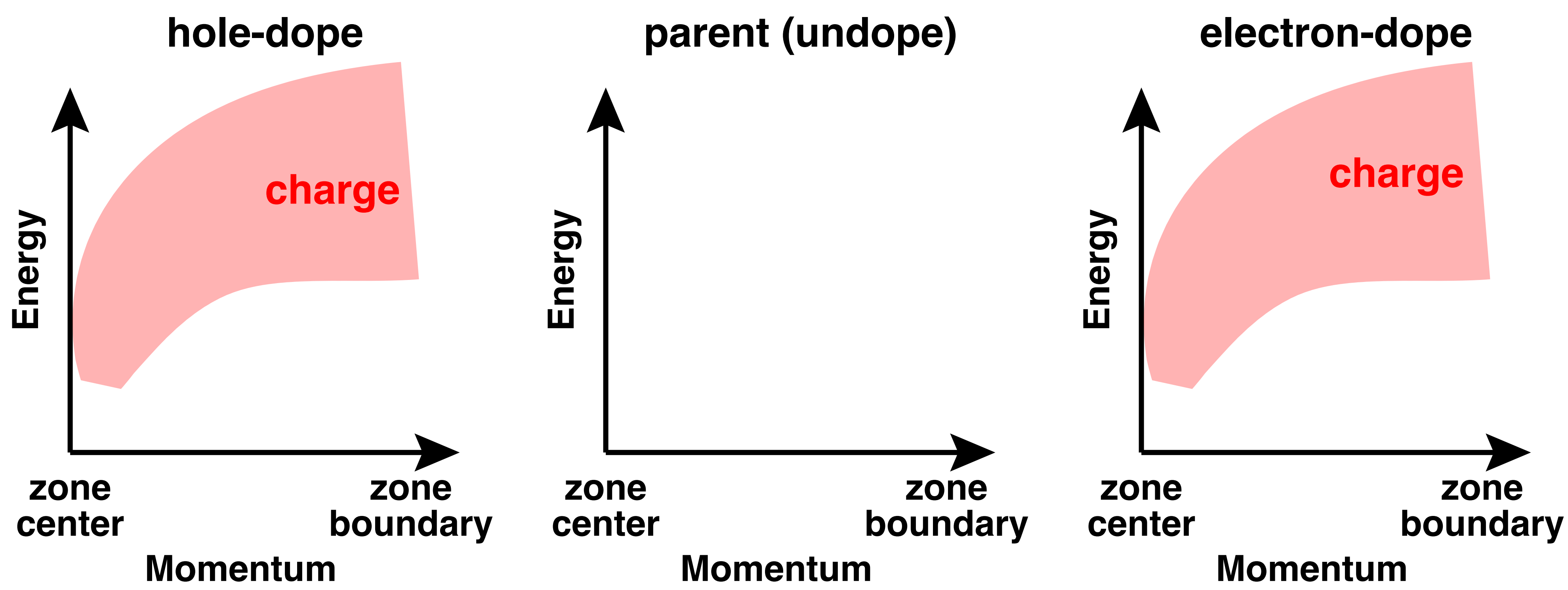
e-dope



additional mode
above magnetic excitations

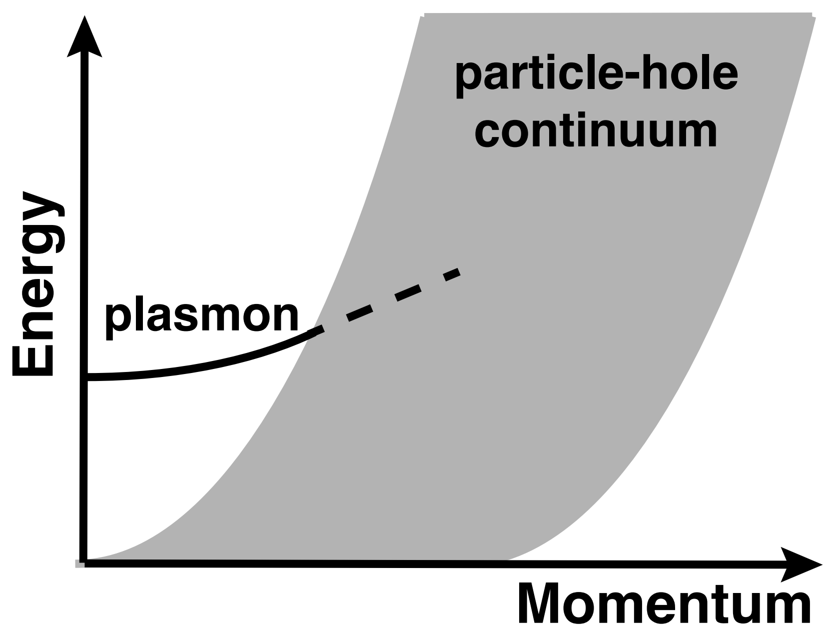
Ishii et al., Nat. Commun. **5**, 3714 (2014)

Summary of charge excitations in cuprates



Now we have a tool to measure charge excitations !

charge excitations of free-electron model

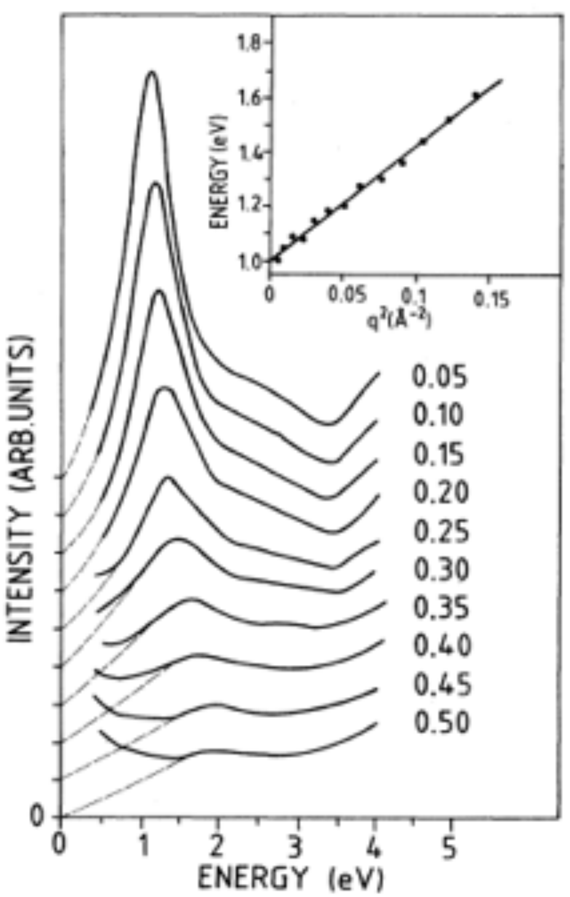


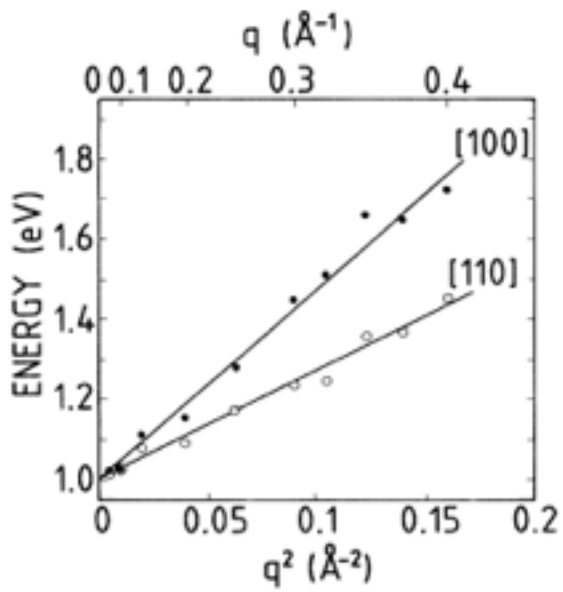
Next issues

- effect of strong electron correlation
- relation to charge order, pseudo-gap, superconductivity, ...

EELS studies of hole-doped cuprates

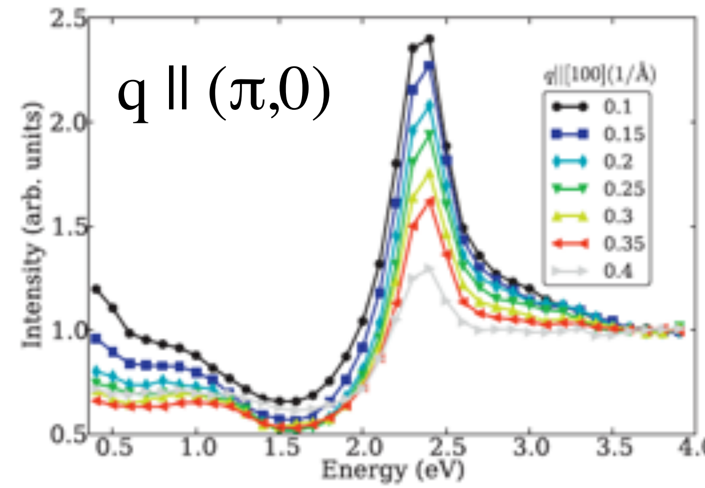
$Bi_2Sr_2CaCu_2O_8$

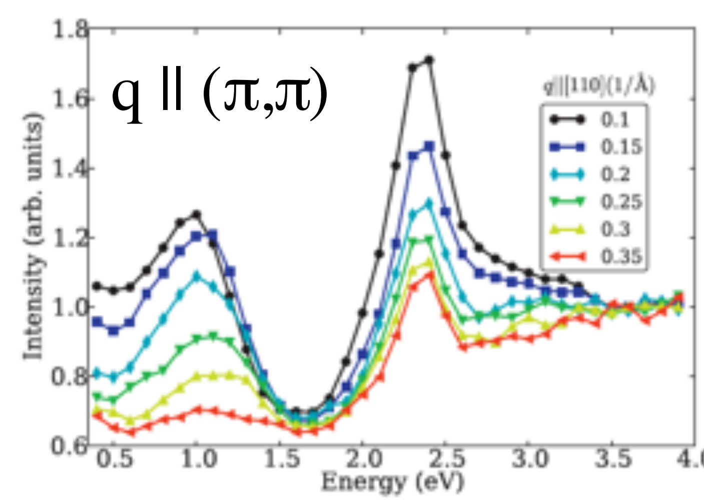


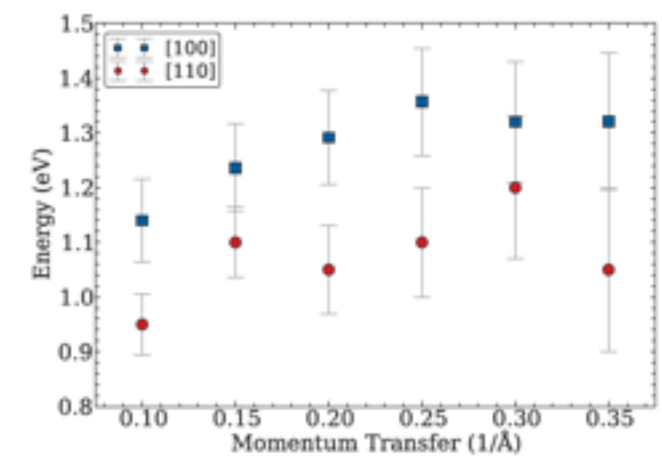


q = 0.1 $Å^{-1}$
→(0.060,0) or
(0.043,0.043)
in r.l.u.

$Ca_{1.9}Na_{0.1}CuO_2Cl_2$







- plasmon
- q^2 dispersion
- ~ 1 eV gap at Γ-point

- plasmon (?)
- q dependent intensity
- ~ 1 eV gap at Γ-point

N. Nücker et al., PRB **39**, 12379 (1989)
N. Nücker et al., PRB **44**, 7155 (1991)

R. Schuster et al., PRB **86**, 245112 (2012)

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1. Layered high-T_c cuprates
   spin excitations + **charge** excitations

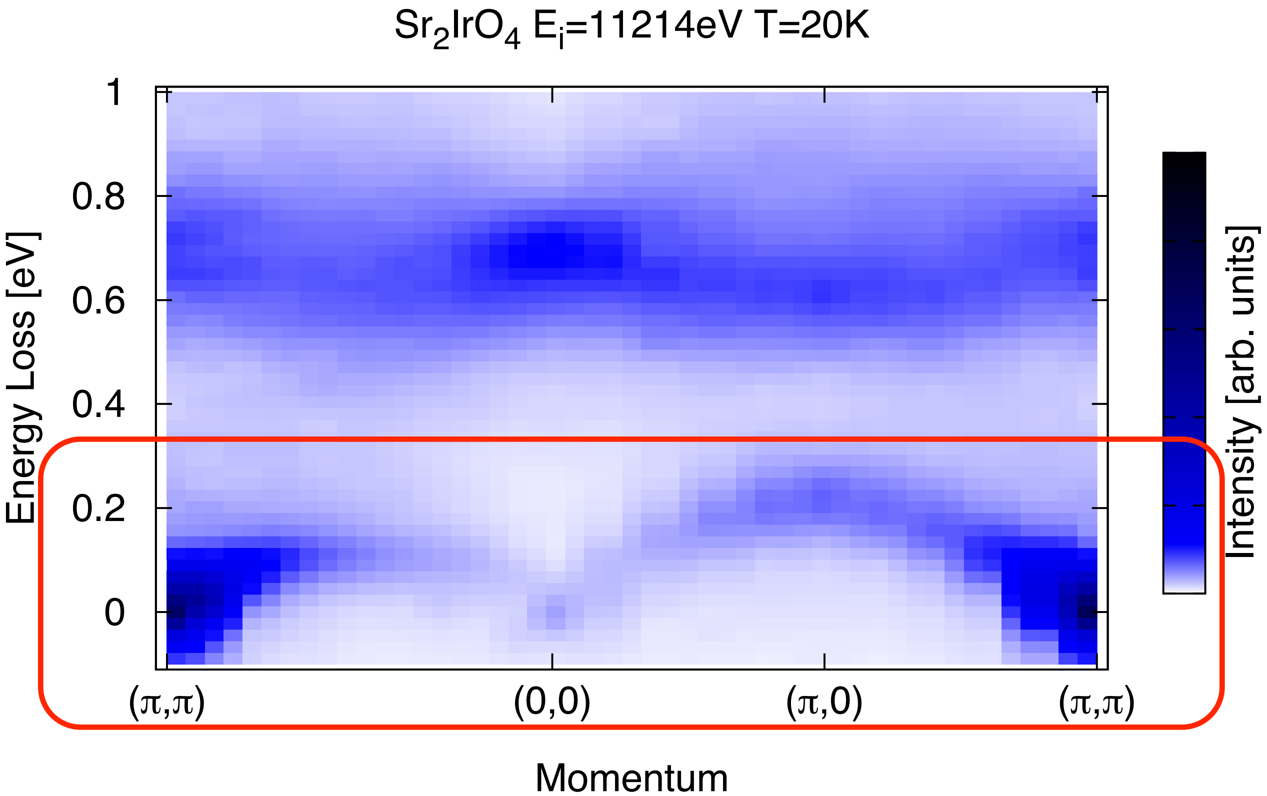
2. Layered iridates
   (pseudo-)**spin** excitations coupled to **charge**

3. Layered iridate
   exciton in **spin**-**orbit** entangled states (d-d excitation)

4. 1-dimensional cuprate
   spin-**orbital** separation (d-d excitation)

Magnetic excitations in Sr_2IrO_4

BL11XU at SPring-8

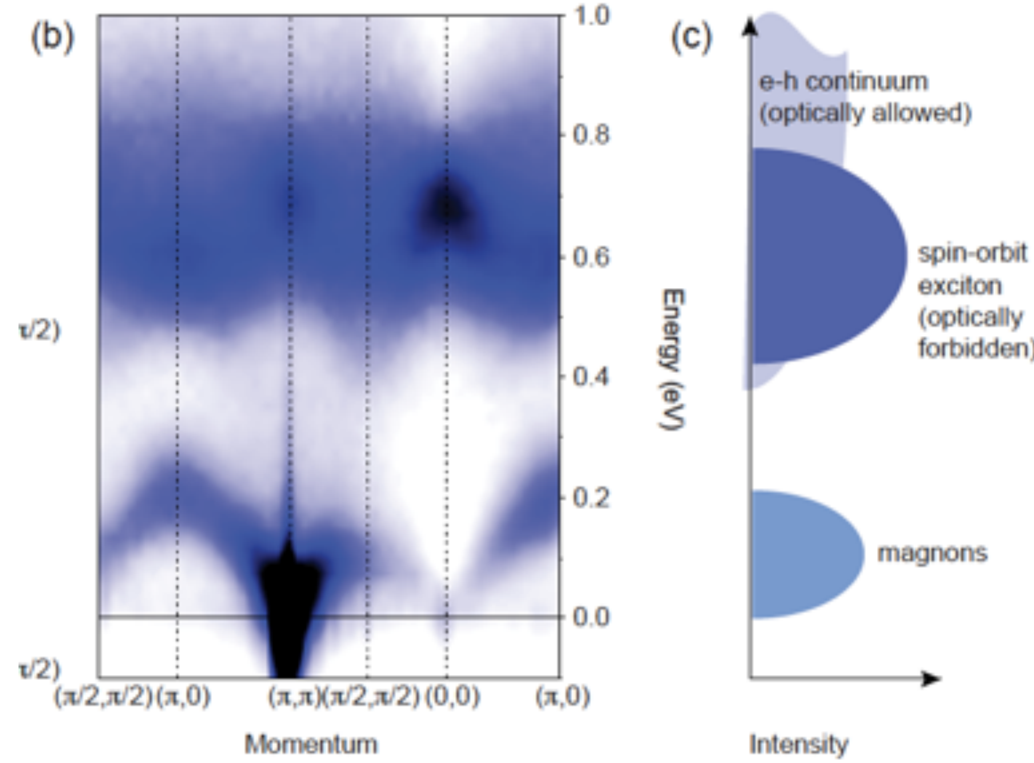


Dispersive magnetic excitation
(magnon of J_{eff} = 1/2)

J-J'-J" Heisenberg model
J = 60 meV, J' = –20 meV, J" = 15 meV

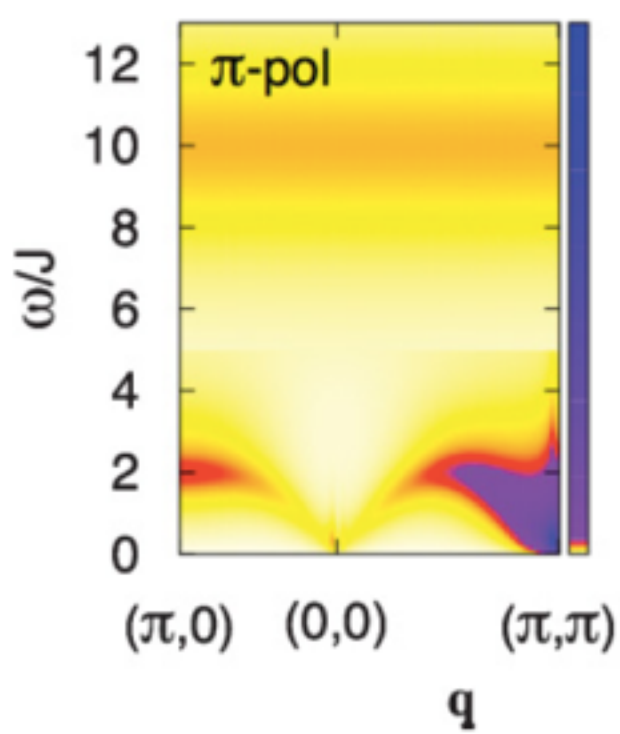
30ID at APS

J. Kim PRL **108**, 177003 (2012)

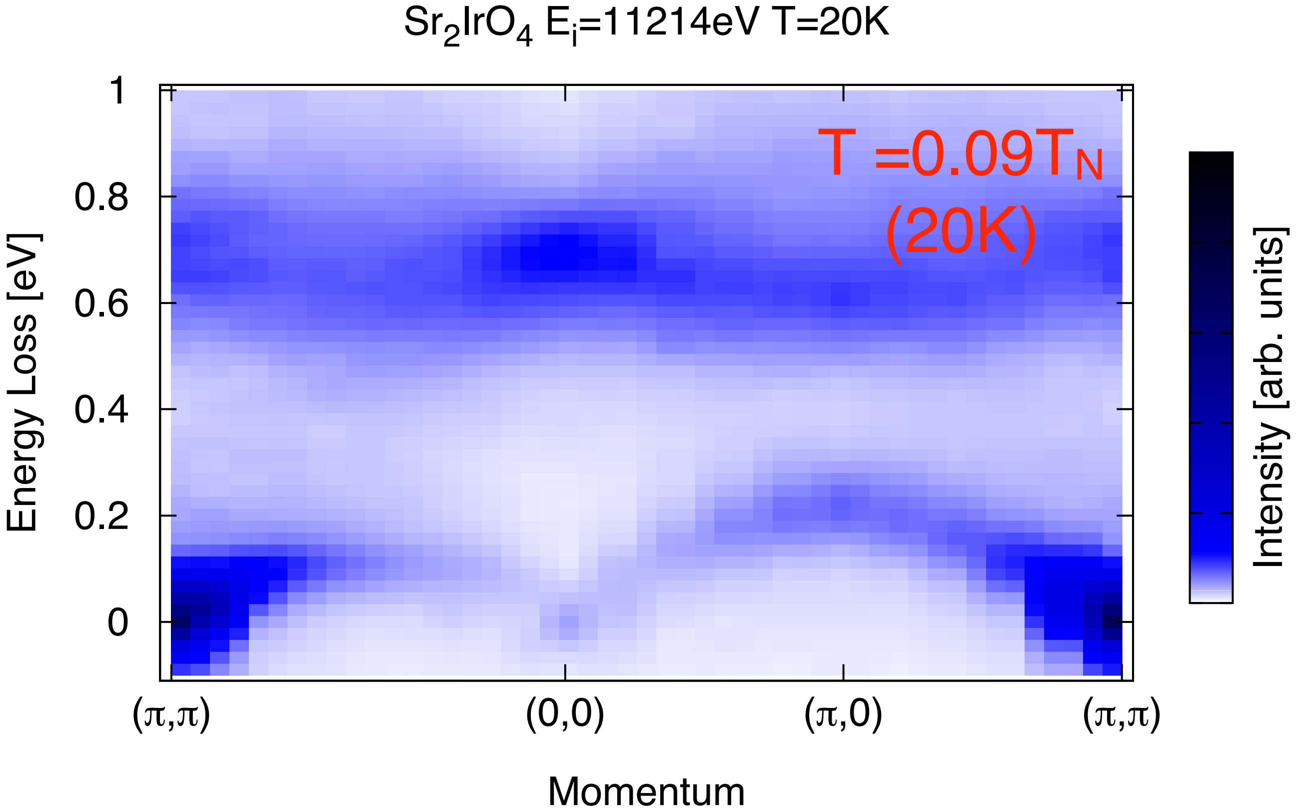


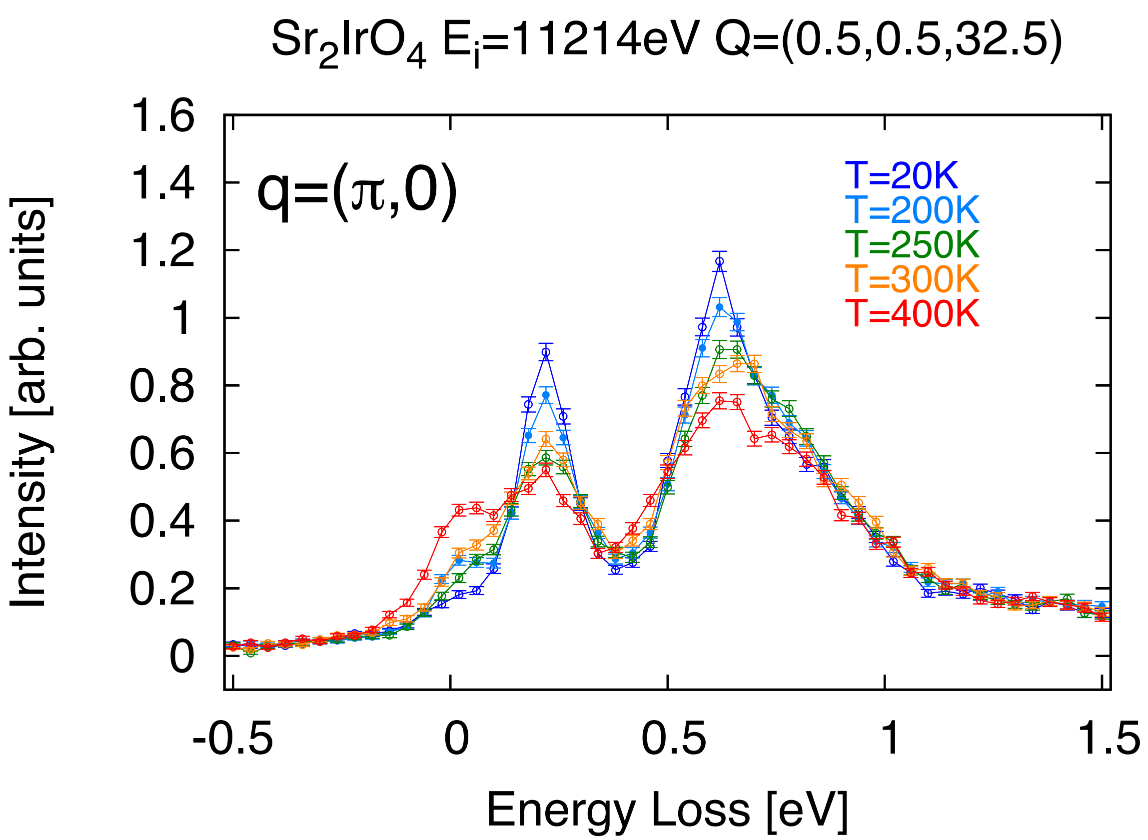
Theory

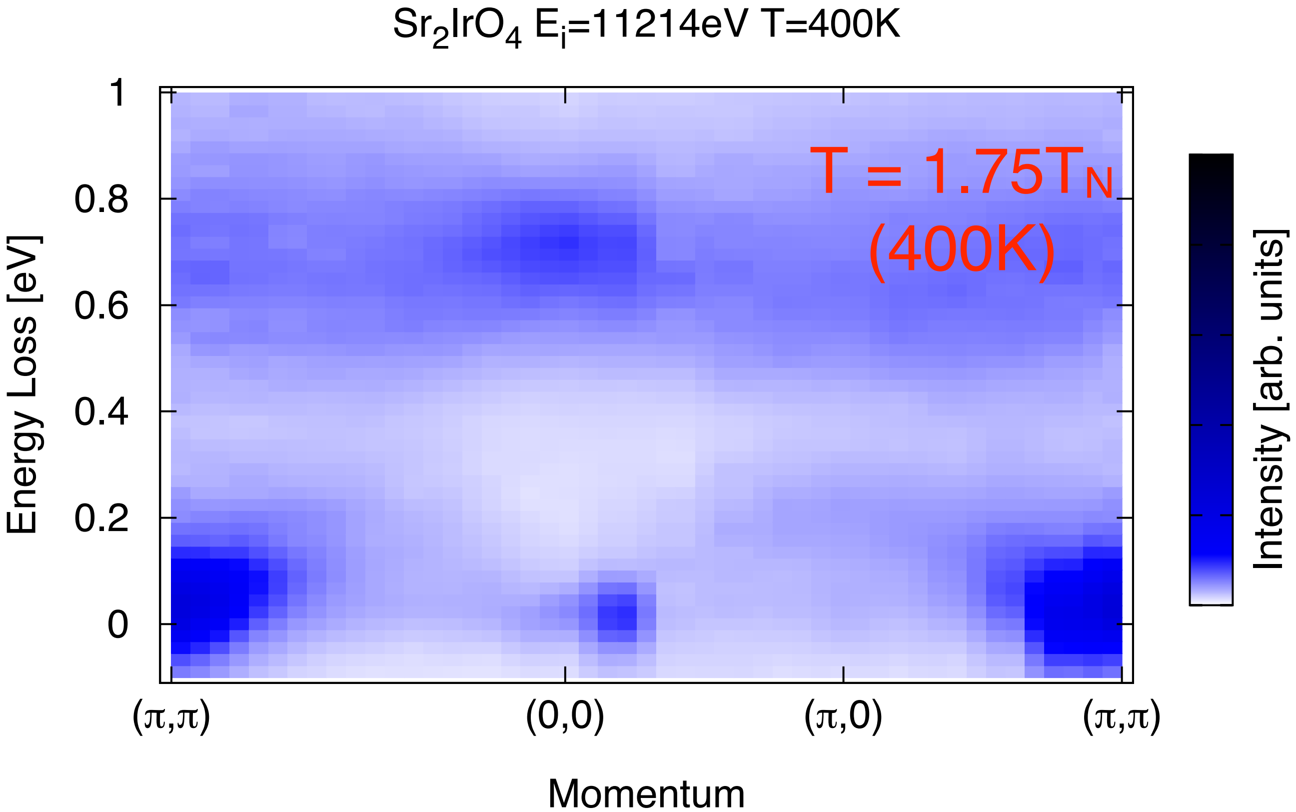
L. J. P. Ament et al., PRB **84**, 020403 (2011)



Temperature dependence





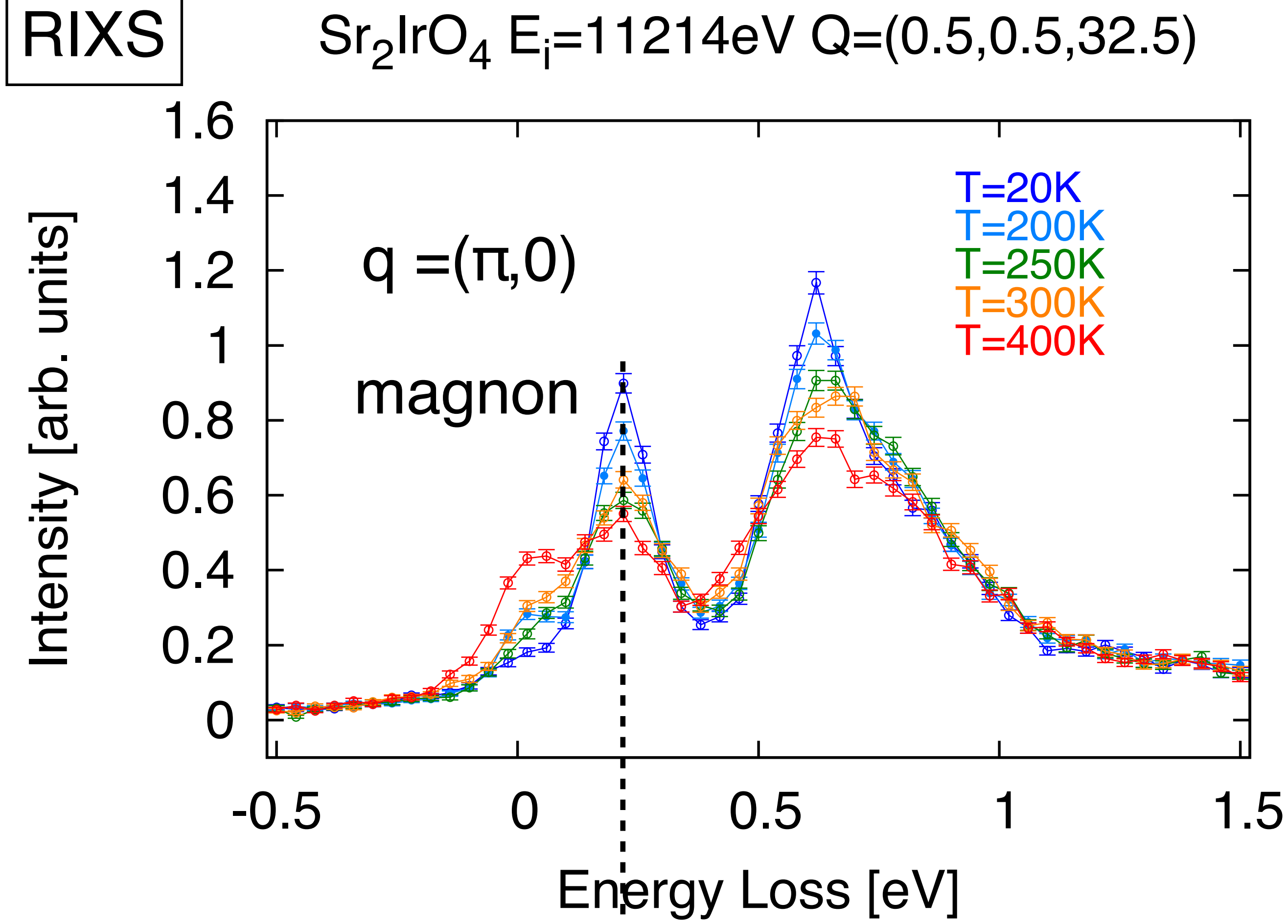


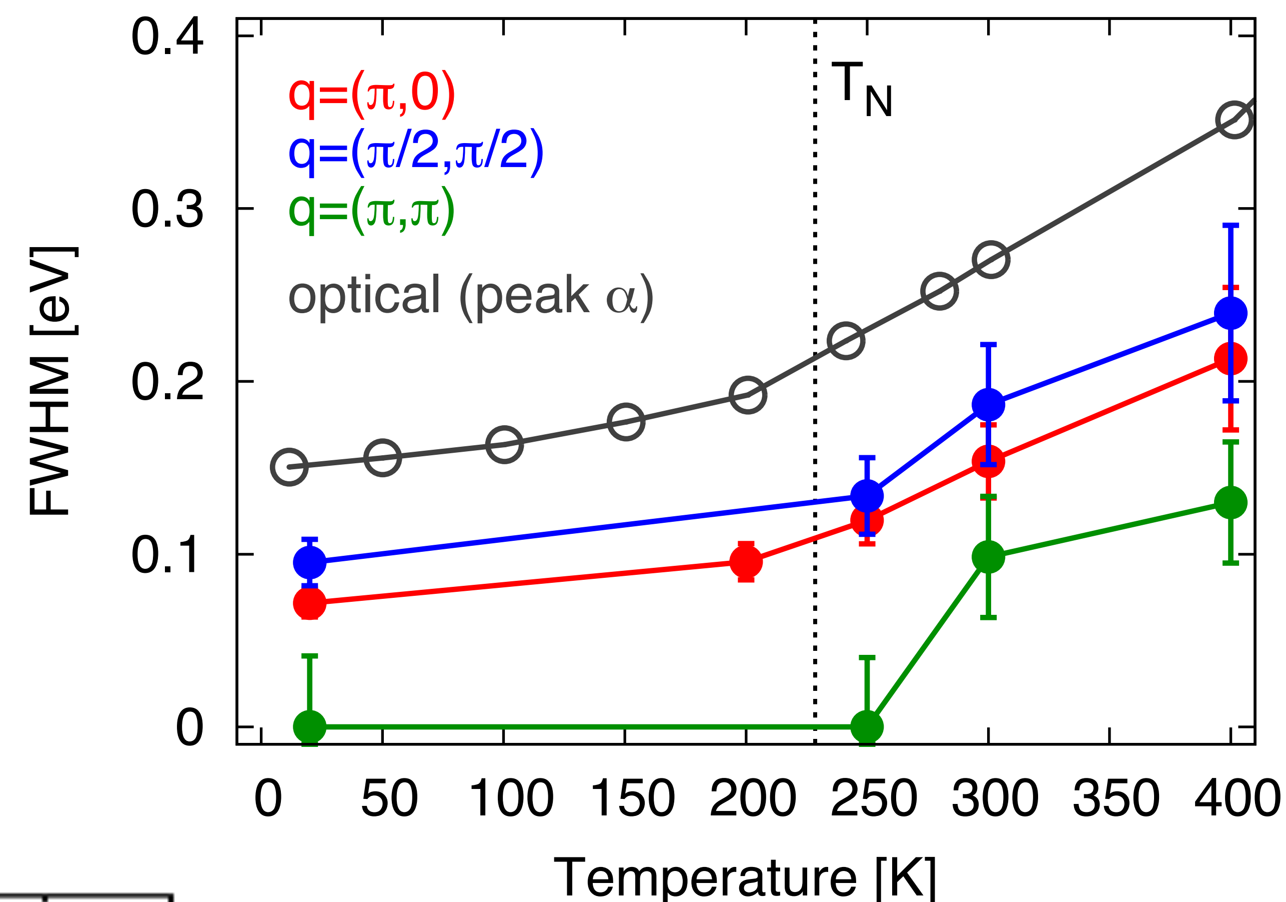
Magnon-like dispersive mode survives above T_N = 228.5 K.

⇒ strong 2D character of magnetism

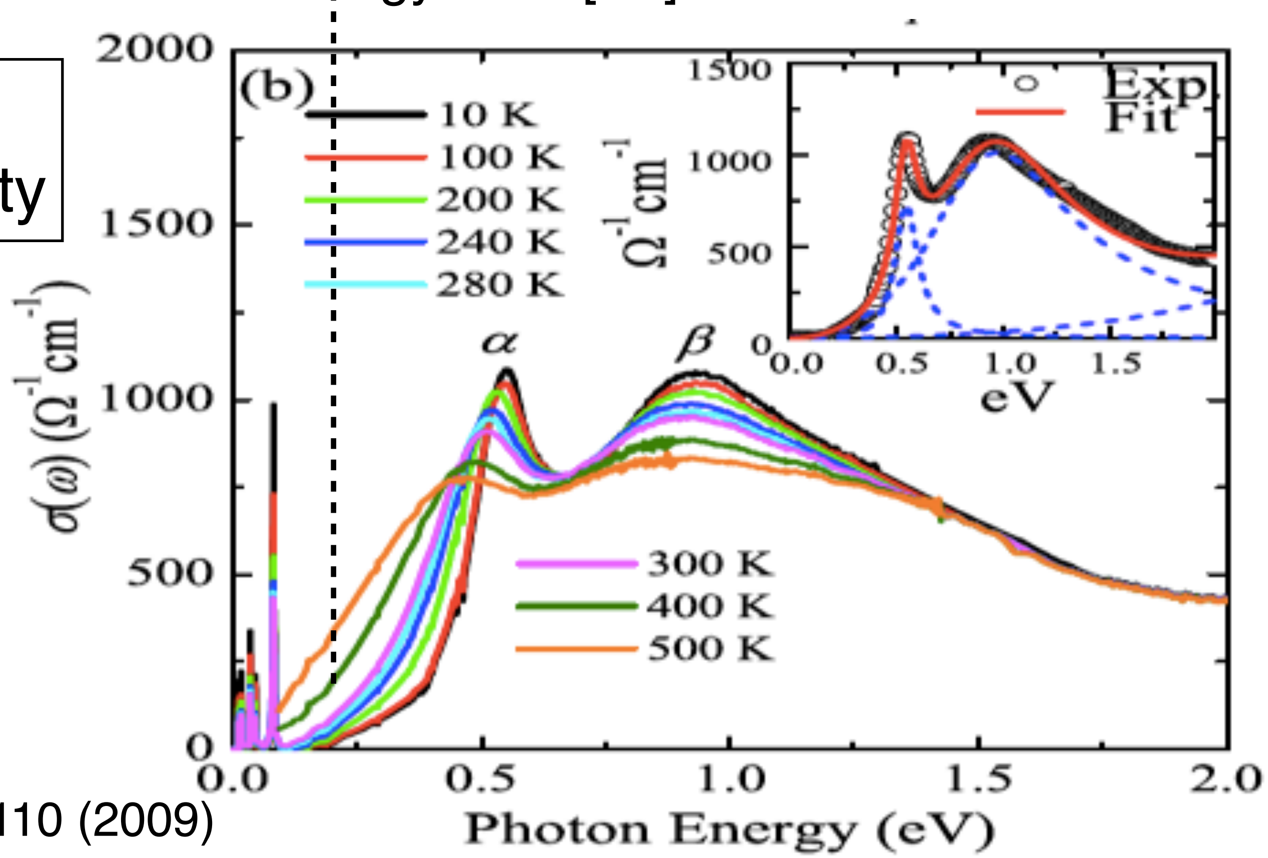
Comparison with charge dynamics

RIXS





optical conductivity



Peak width of magnetic excitations is similar to optical conductivity

Moon et al.,
PRB **80**, 195110 (2009)

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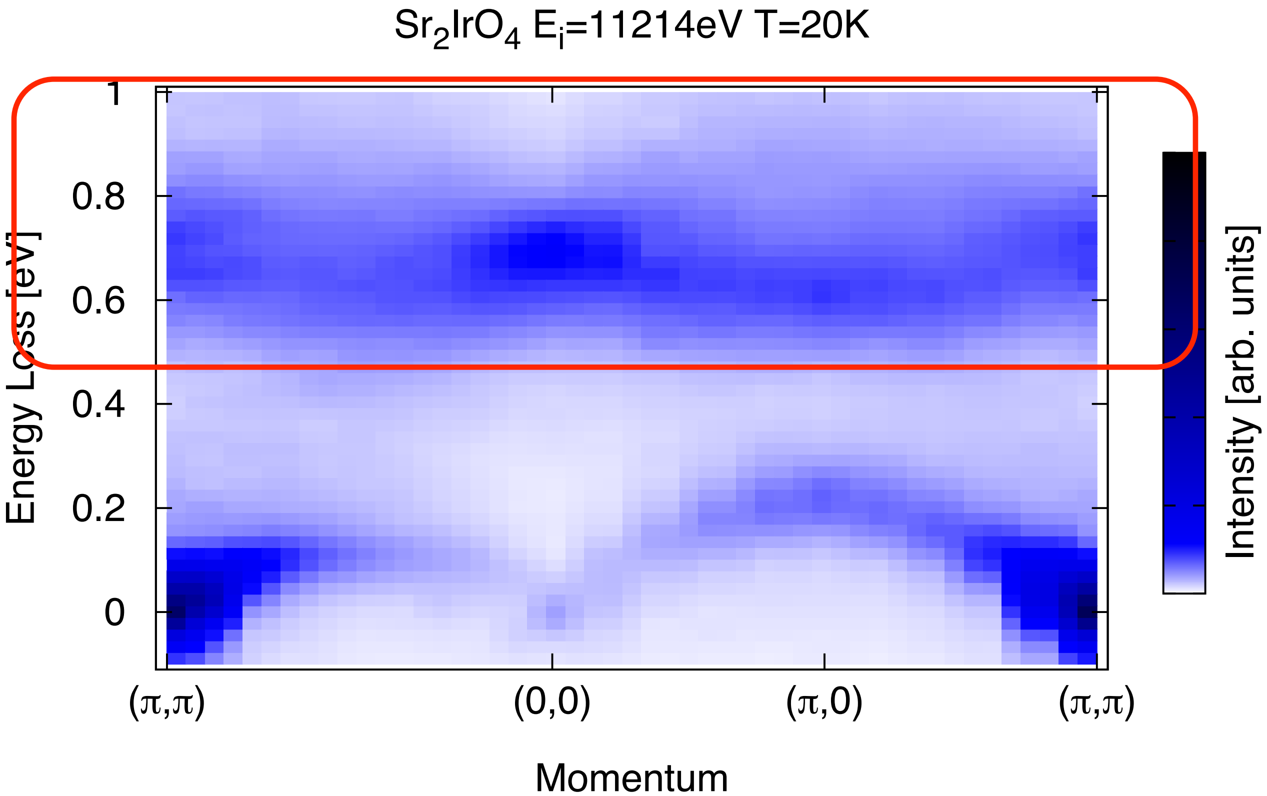
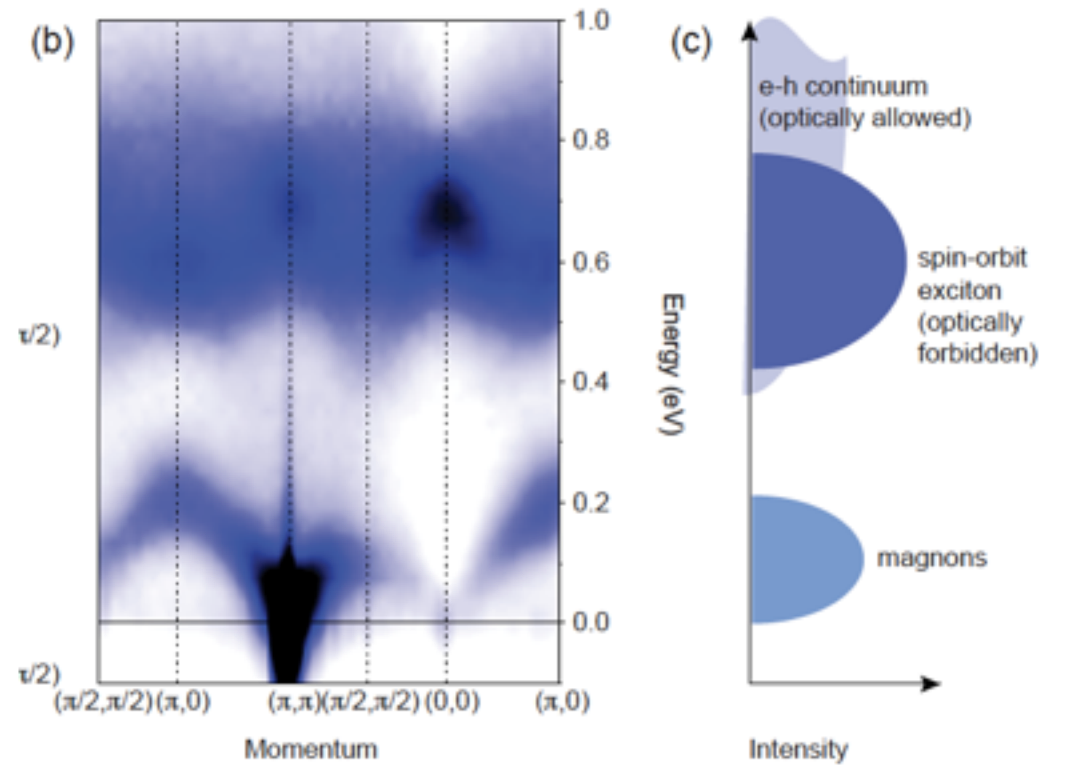
<u>BL11XU at SPring-8</u>



<u>30ID at APS</u>

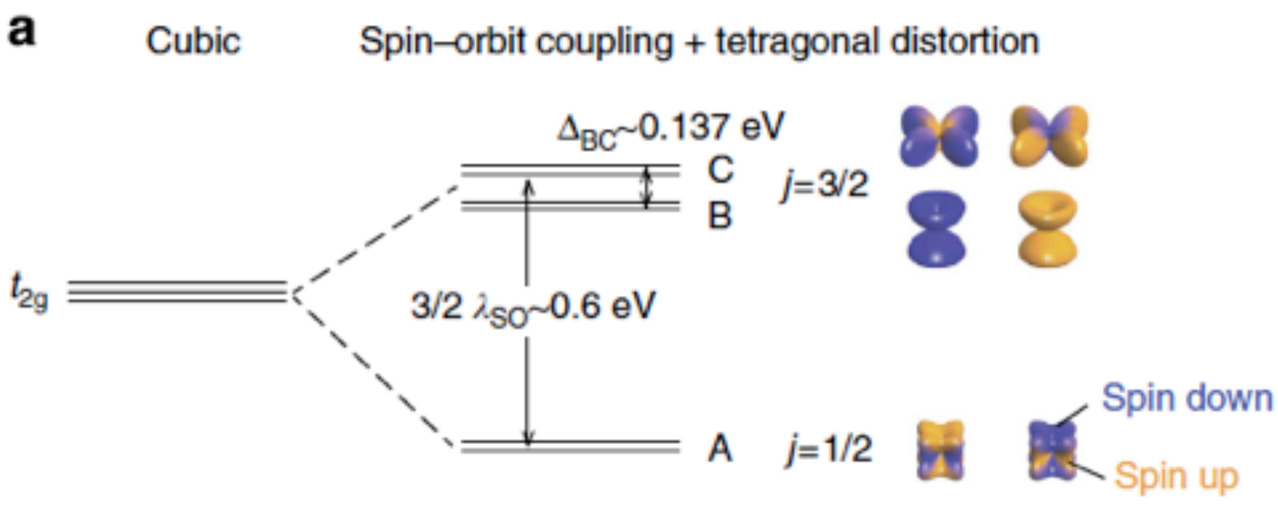
J. Kim PRL **108**, 177003 (2012)



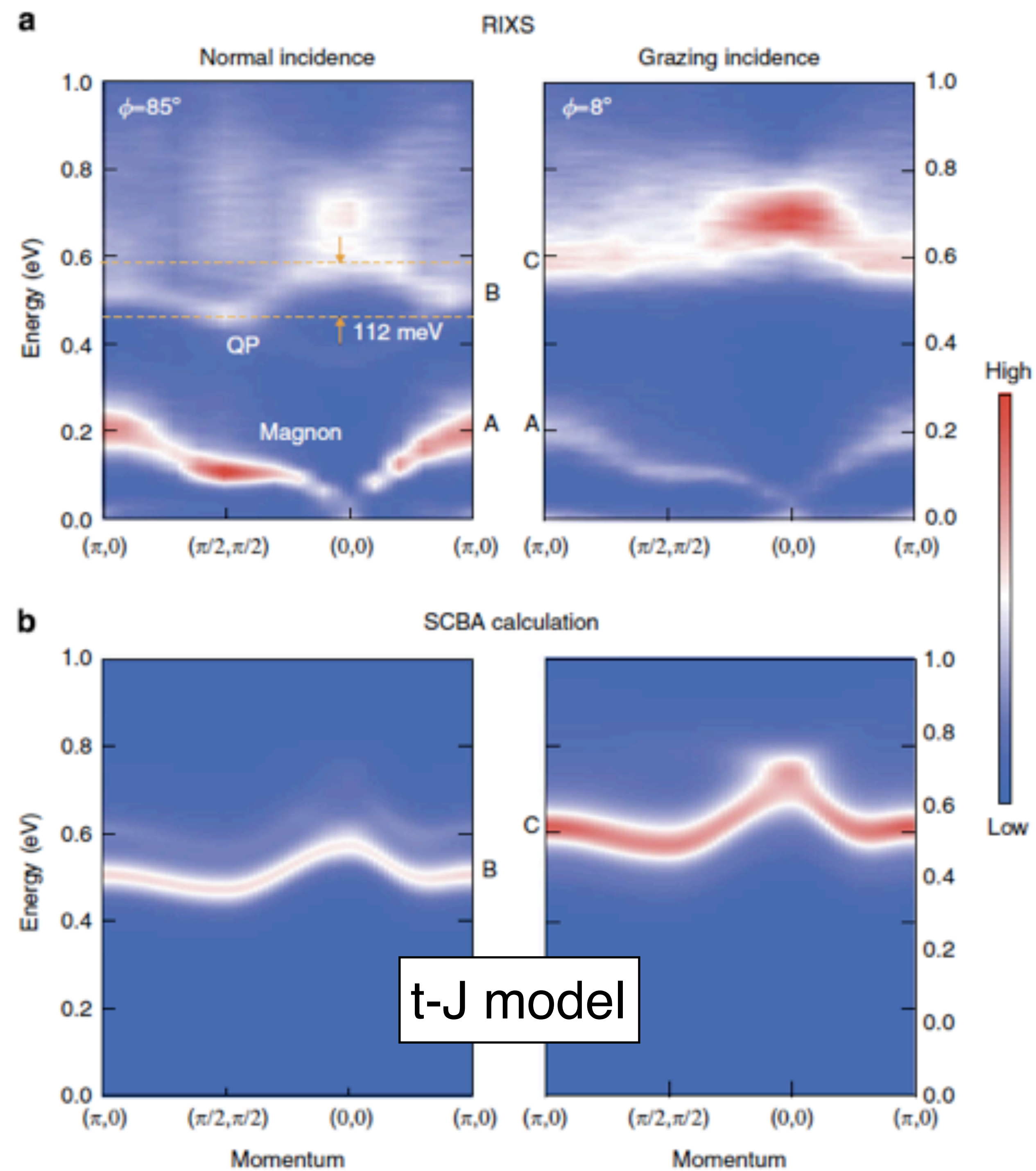
Dispersive dd excitations

Excitonic quasiparticle in Sr_2IrO_4

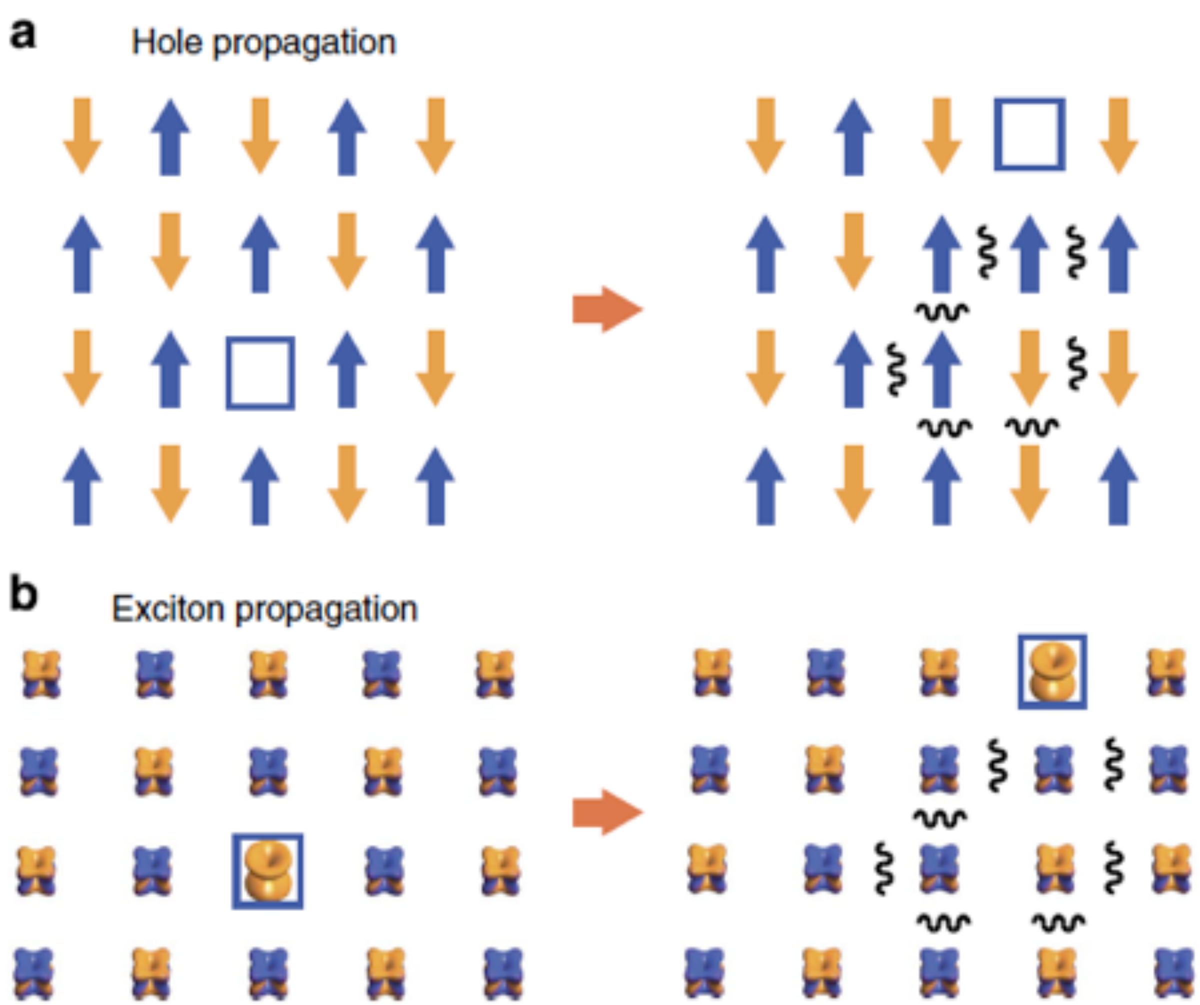
Spin-orbital level scheme



Dispersive excitonic mode



Hole vs Exciton propagation in AF background



t-J model

J. Kim et al., Nat. Commun. **5**, 4453 (2014)

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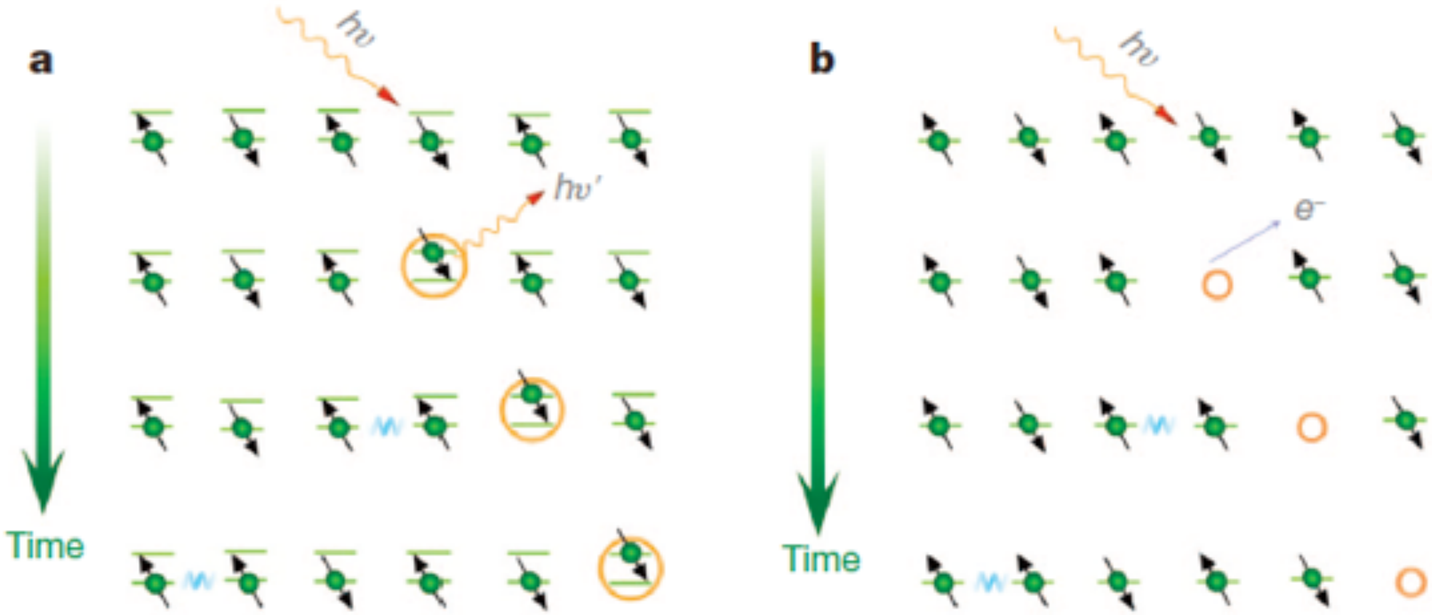
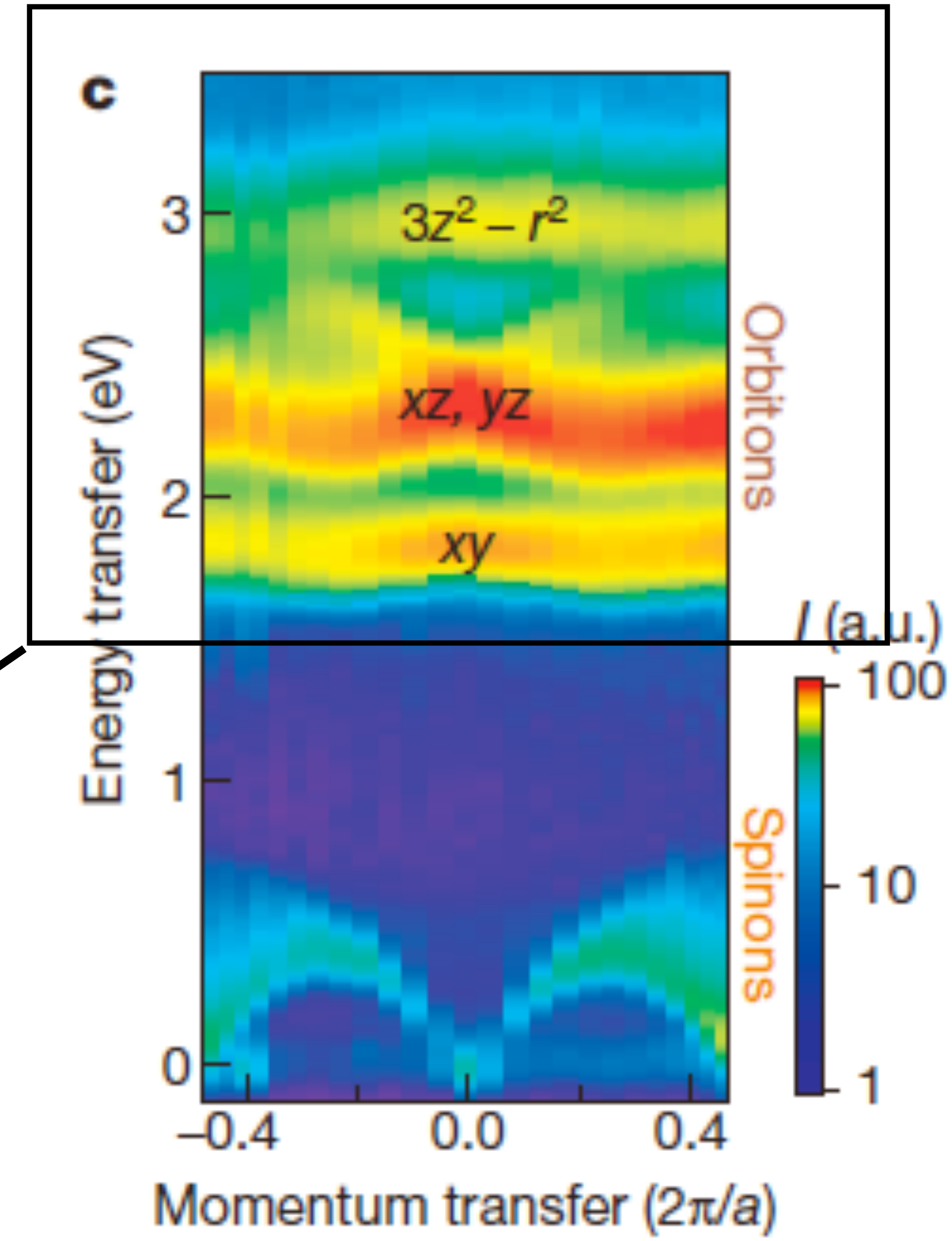
3. Layered iridate
   exciton in **spin**-**orbit** entangled states (d-d excitation)

4. 1-dimensional cuprate
   spin-**orbital** separation (d-d excitation)

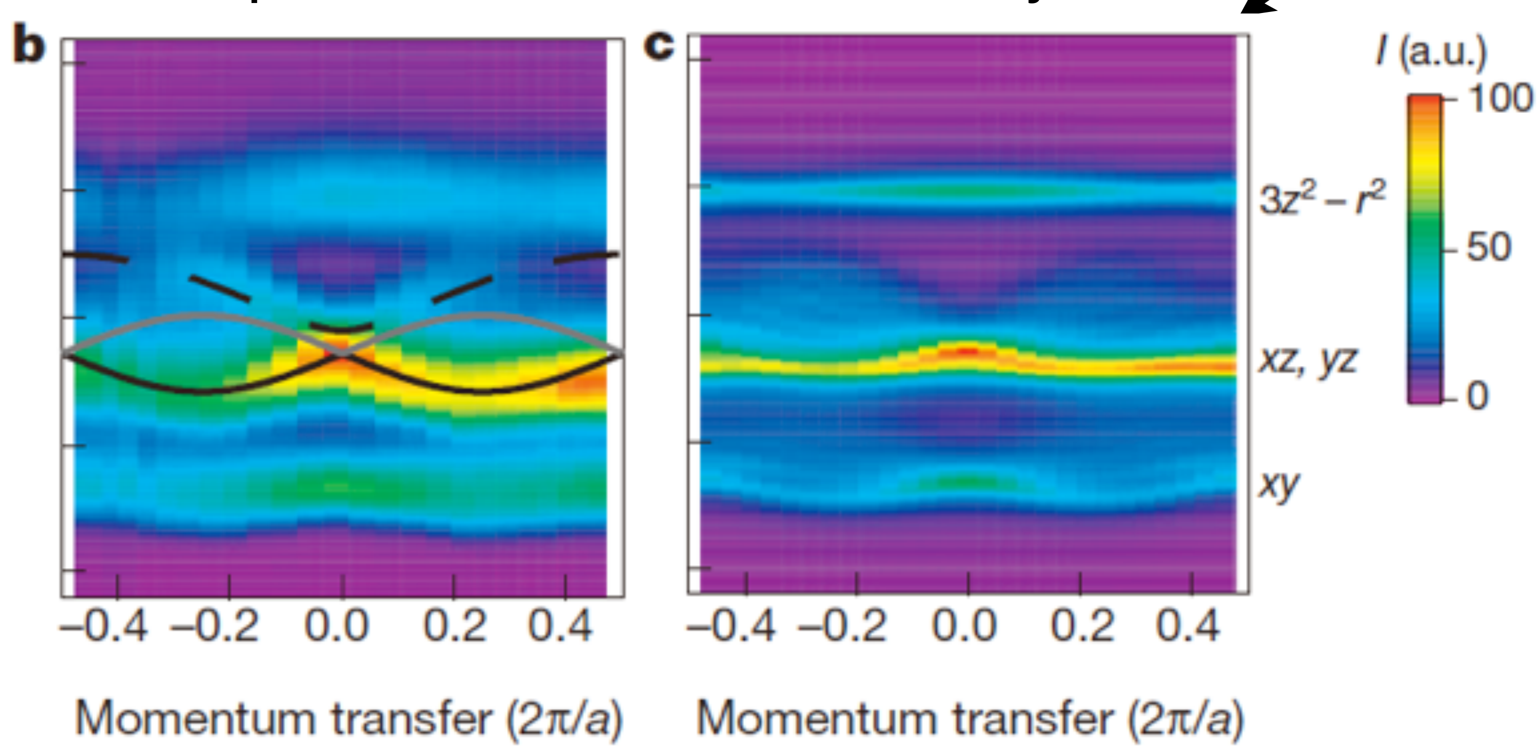
Spin-orbital separation in 1D cuprate Sr_2CuO_3

spin-orbital separation | spin-charge separation





experiment | theory



J. Schlappa et al., Nature **485**, 82-85 (2012)

まとめ

ここ十数年で、

- エネルギー分解能が顕著に向上し、大きな発展を遂げた。
- 磁気励起の観測というブレークスルーもあり、強相関電子系での利用研究が進んだ。

  <u>スピン励起</u>

  　中性子非弾性散乱による研究の蓄積があり、相補的手法として発展している。

  <u>電荷励起・軌道励起</u>

  　いくつかの例が観測され、実験的検証が可能となった。

今後、

- 方向の一つとして、**高エネルギー分解能化**の流れは続くであろう。

  （高フラックス、高指向性のＸ線が不可欠）

  次の5-10年は、数十meV領域（~k_BT）の測定へ

  物性を説明する理論モデルは、**Q**-ω空間での励起状態まで含めた検証が可能になる

  新しい励起　⇒　新しい物理へ

- 励起の同定に有用な、**偏光の積極的な利用**も進める必要がある。