

Hard x-ray RIXS study on
high-T_c cuprates
and related compounds

*Charge excitation associated with
charge ordered state*

Shuichi Wakimoto
Japan Atomic Energy Agency

Collaborators

Hiroyuki Kimura
(Tohoku Univ.)

Kenji Ishii
(JAEA/Spring-8)

Kazuhiko Ikeuchi
(JAEA/Spring-8)



@ Wild Fire Steak House

Jun'ichiro Mizuki, Kazuhisa Kakurai (JAEA)

Masaki Fujita, Kazuyoshi Yamada (IMR)

Tadashi Adachi, Youji Koike, Yukio Noda (Tohoku Univ.)

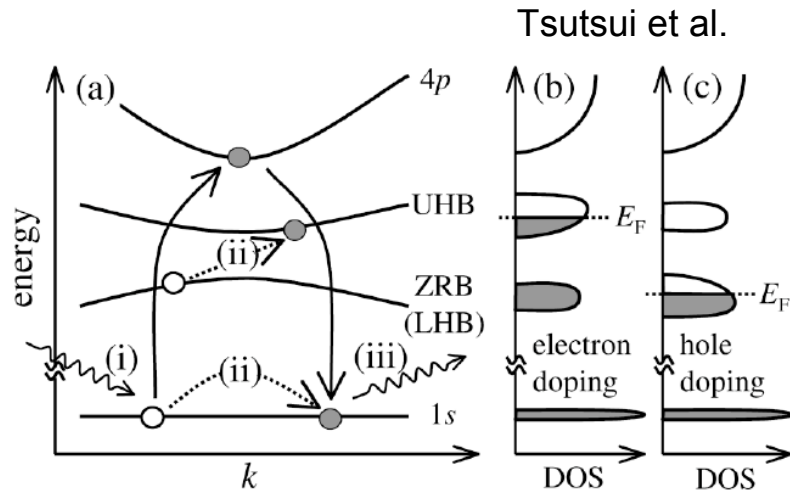
Aimon Said, Yuri Shvyd'ko (APS)

CONTENTS

1. Introduction
 - a. Hard x-ray RIXS
 - b. Stripe order in the 214 compounds
2. Experiments
 - a. Samples
 - b. Instruments (MERIX, BL11XU)
3. Results
 - a. Cuprates
 - $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$ $x=0.125, 0.08$
 - $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ $x=0.12$
 - b. $\text{La}_{5/3}\text{Sr}_{1/3}\text{NiO}_4$
 - c. Ladder system (work by Yoshida et al.)
4. Summary

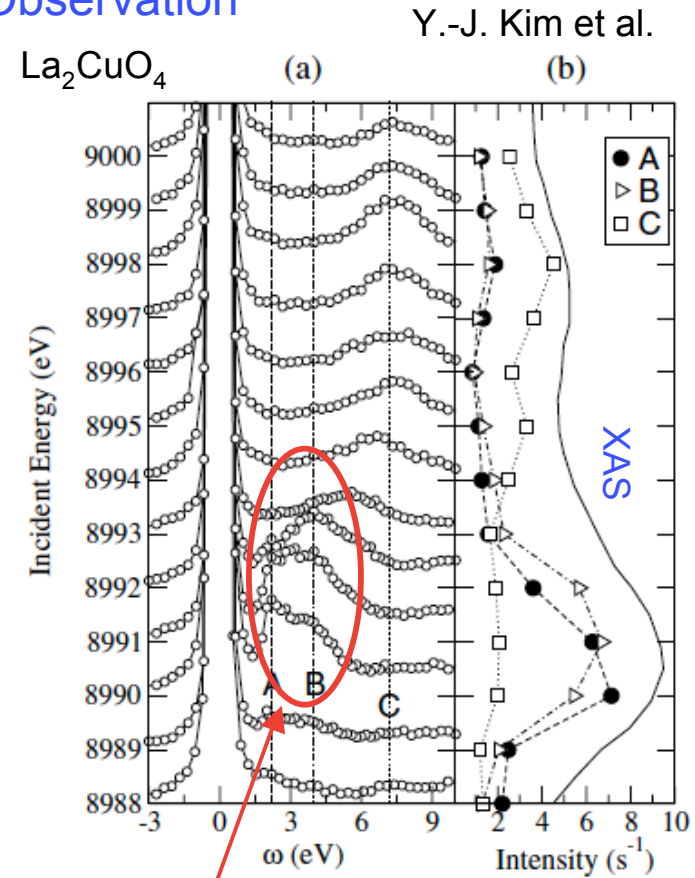
Hard x-ray RIXS (Cu, Ni, ... K-edge)

Principle



- ◆ The RIXS process involves an intermediate state where 1s electron is excited to 4p.
- ◆ In this process, many type of charge excitations are induced.
- ◆ Charge transfer (CT) type insulator -CT excitation (O2p to Cu3d) is very sensitive to this process.

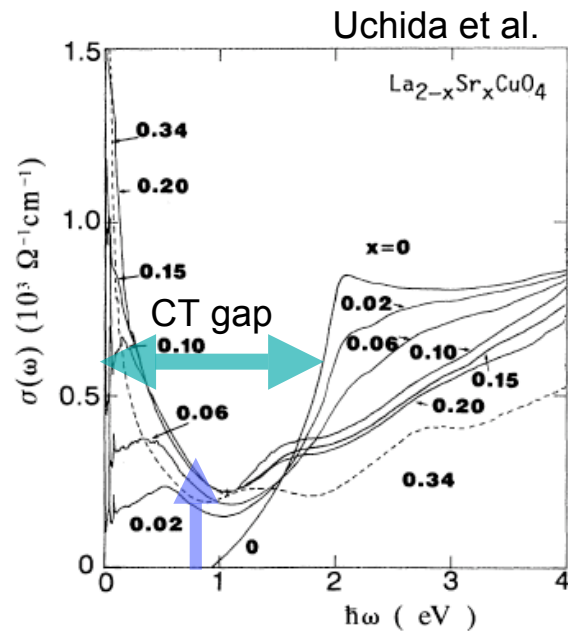
Observation



CT excitation resonates near the photon energy where XAS peak appears.

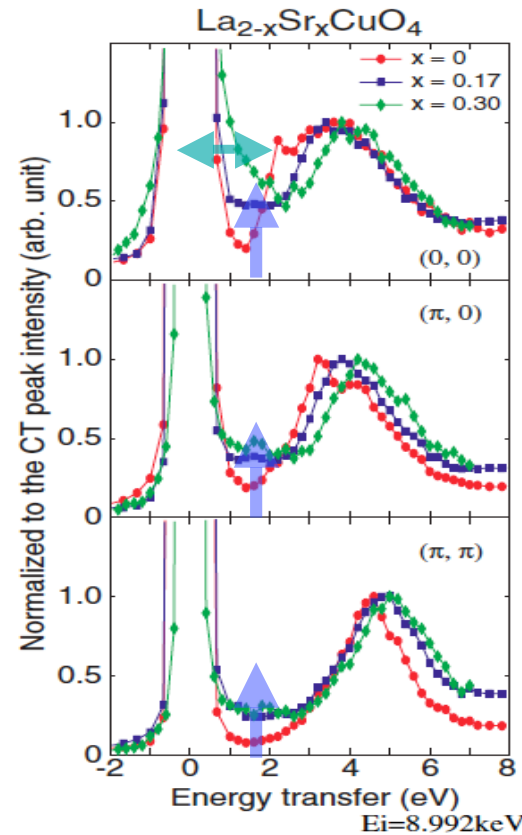
Charge transfer excitation

- Optical measurement one of the most important probes to study CT gap.



- Clear CT gap in the non-doped sample.
- Spectral weight shifts to lower energy as doping increases.

- Cu-K edge RIXS observes ...



- Consistent CT gap in the non-doped sample at $(0, 0)$.
- As doping increases, continuum-like excitation at the lower energy.
- q -dependences of charge transfer excitations.

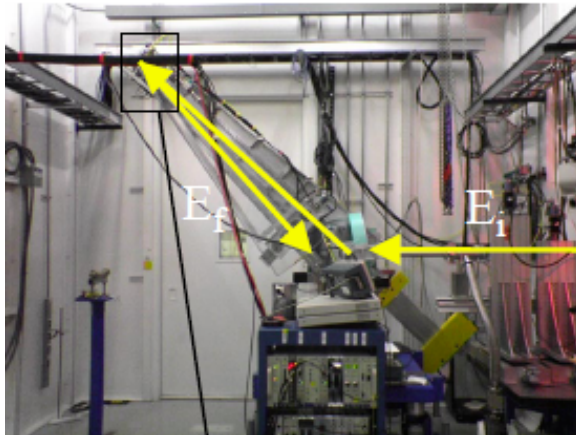
Wakimoto, Kim et al.

Advantage of RIXS
Being able to study q -dependence

Recent development

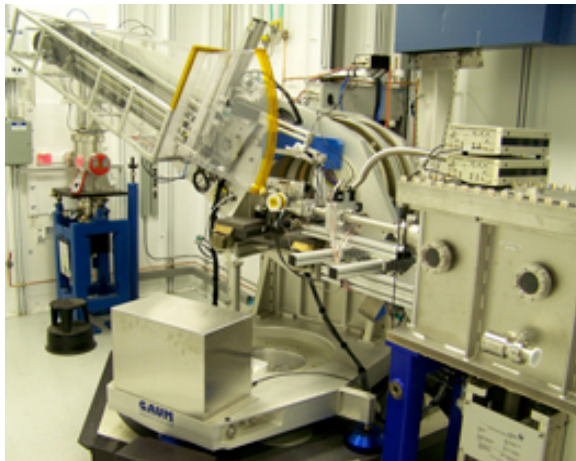
Typical energy resolution of RIXS Instruments : 300 ~ 400 meV
(APS 9ID-B with 1m-arm, SP8 BL11XU, BL12XU, ...)

- ◆ Recent development towards finer energy resolution
APS 9ID-B with 2m-arm



Cu K-edge :
 $\Delta E = 118 \text{ meV}$

APS 30ID MERIXS

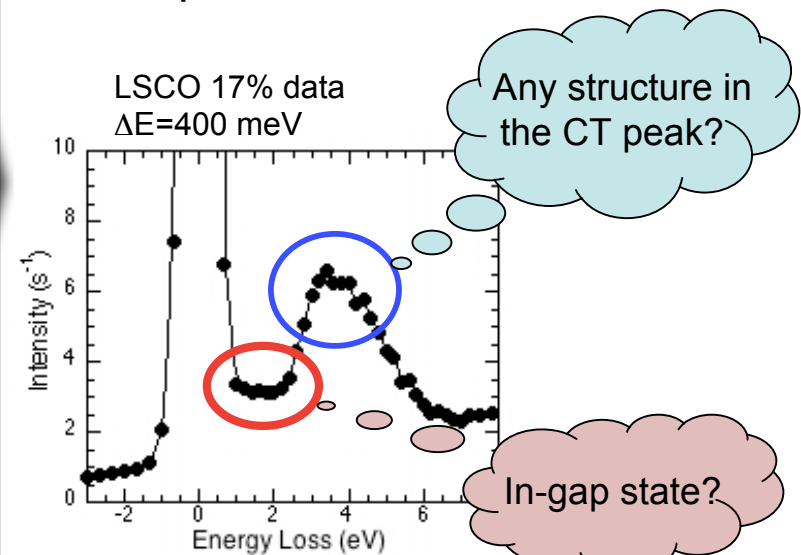


Cu K-edge :
 $\Delta E = 110 \text{ meV}$
Ni K-edge :
 $\Delta E = 150 \text{ meV}$
:
:
:

Small ΔE will ...

- ◆ reduce the elastic tail.
- ◆ help us to see smaller structure.

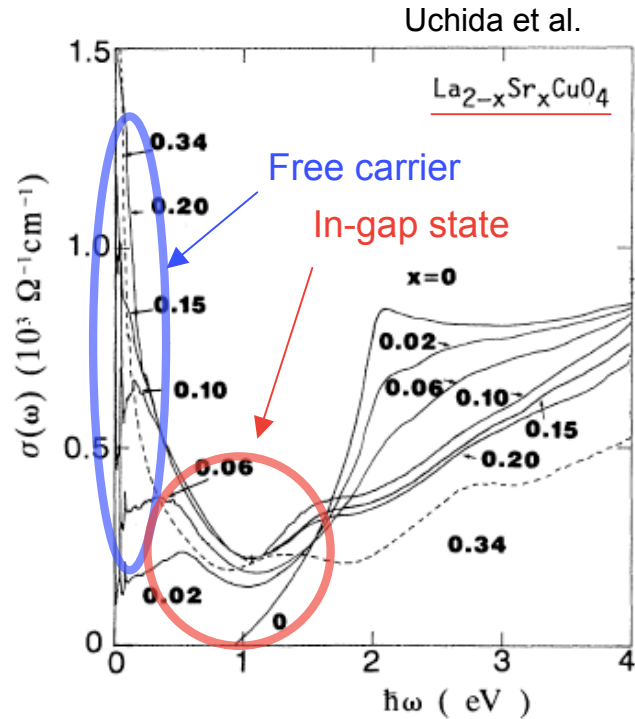
New aspect ...



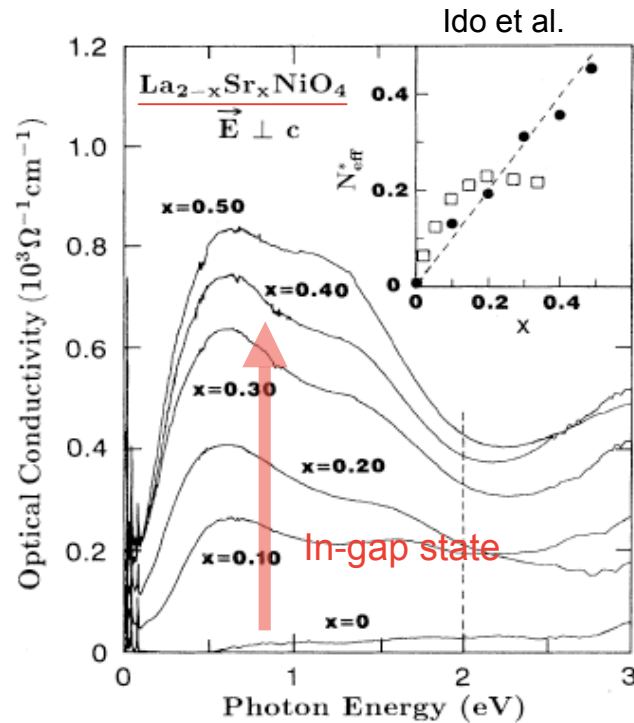
We are aiming here!

In-gap state

◆ Optical conductivity



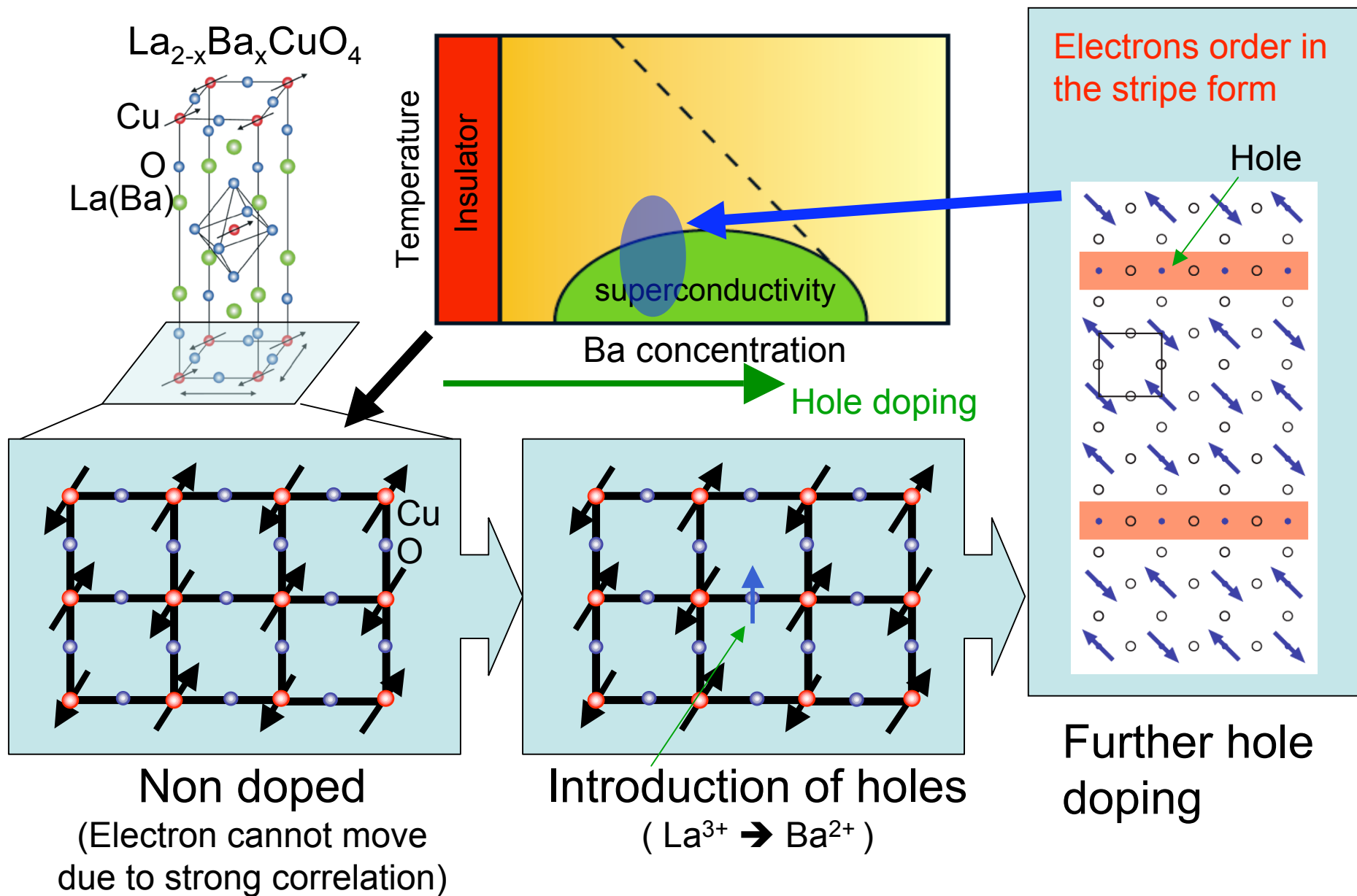
- ◆ When holes are doped into the system, in-gap state appears and grows with doping.



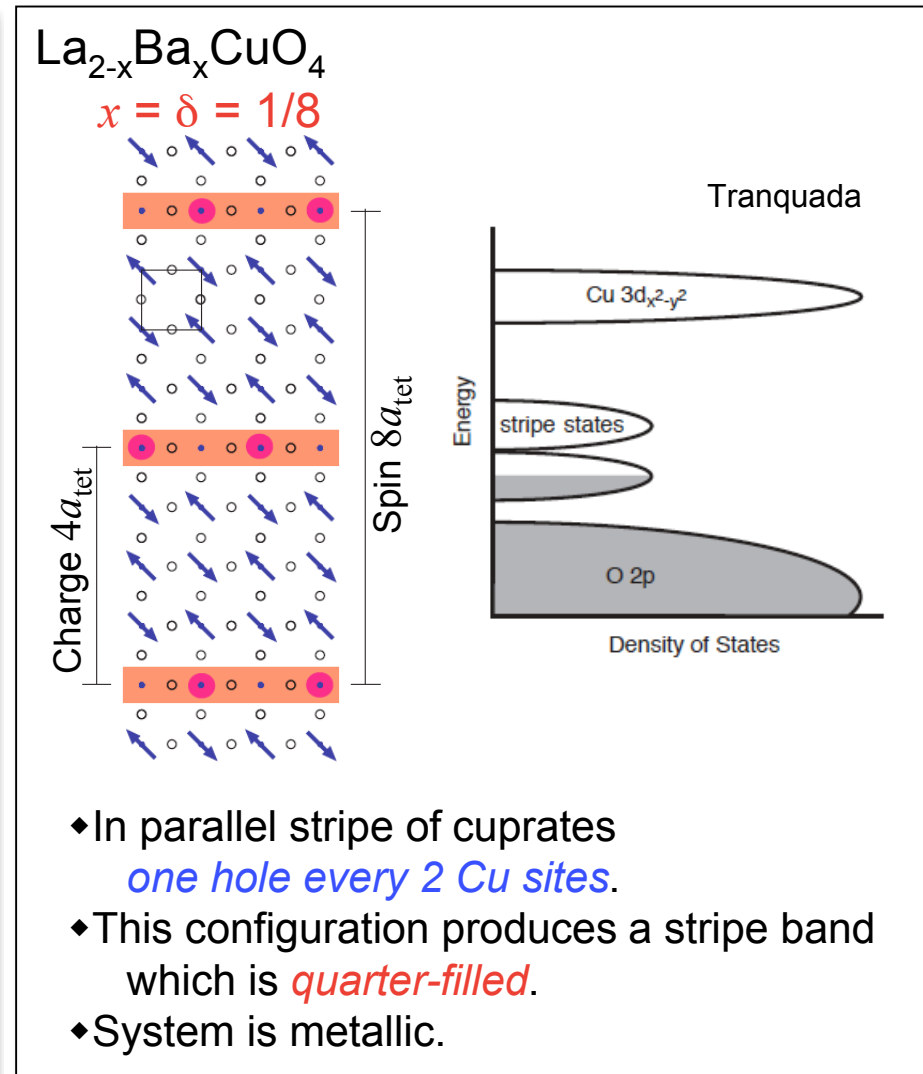
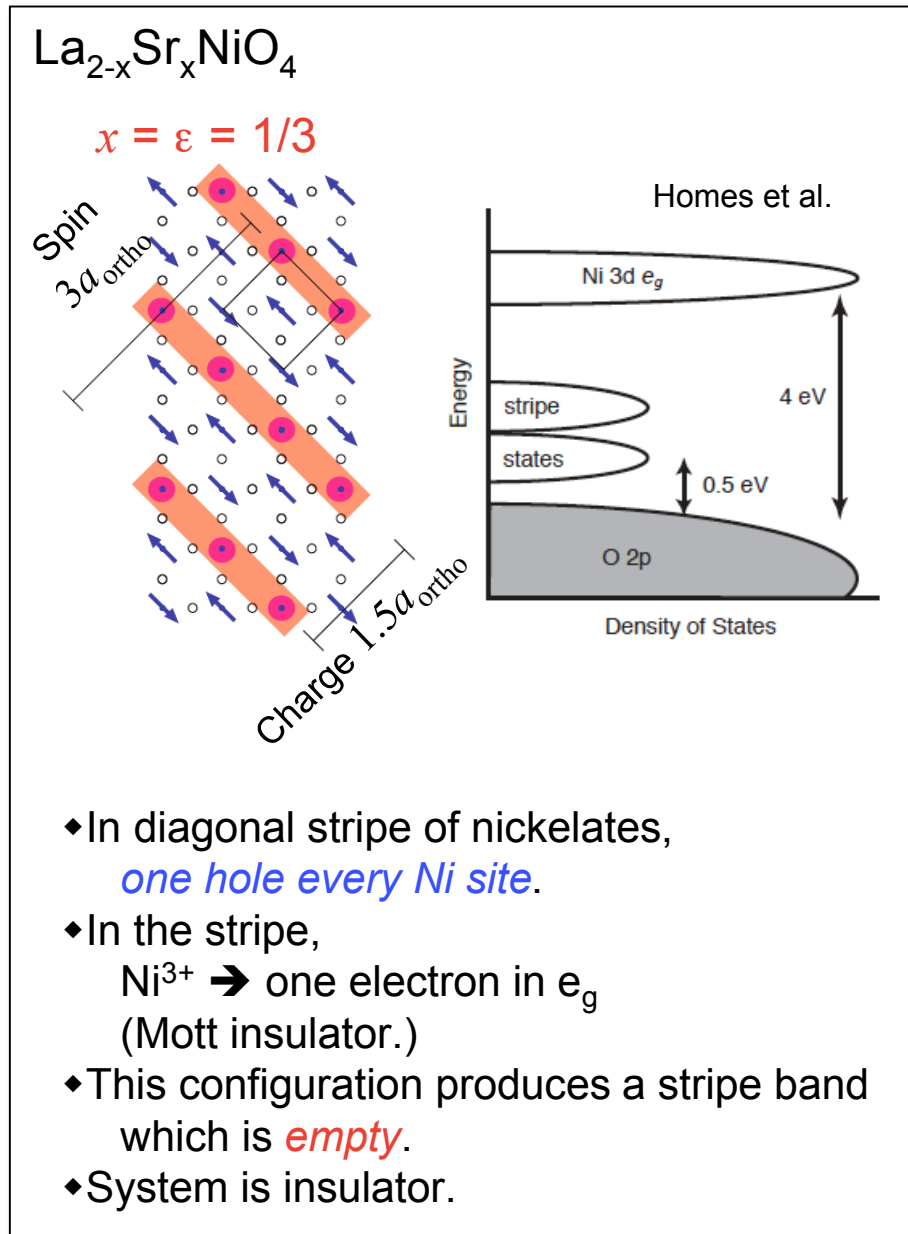
- ◆ This was reported by various experimental probes.
 - Optical measurements
 - Photo-emission
 - x-ray absorption
 - and so on ...

- ◆ Interpreted in several ways.
 - Impurity band
 - Polaron band
 - d-d excitation
 - Stripe band
 - ...

Stripe order



Schematic picture of stripe band



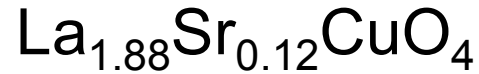
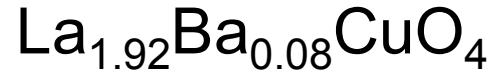
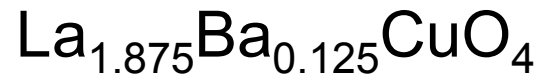
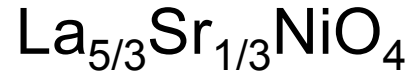
Can we detect charge excitations to the stripe band?

Motivation

- ◆ Ingap state by stripe order?
- ◆ Dynamics of stripes?
 - relation to the superconductivity
 - strongly correlated electron system
 - cf. CDW by electron-phonon coupling

Experiments

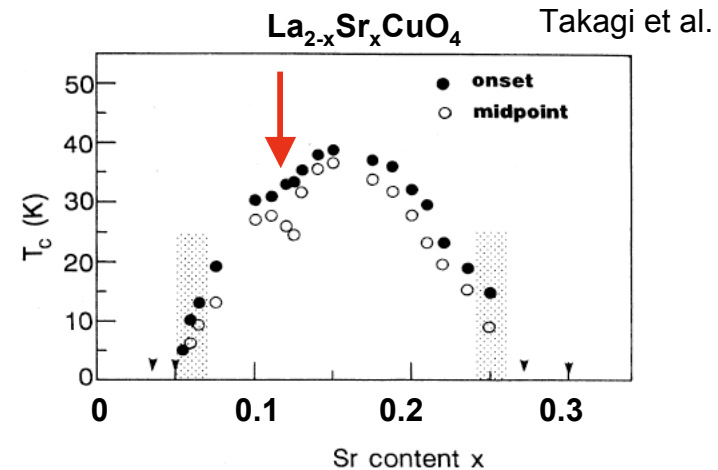
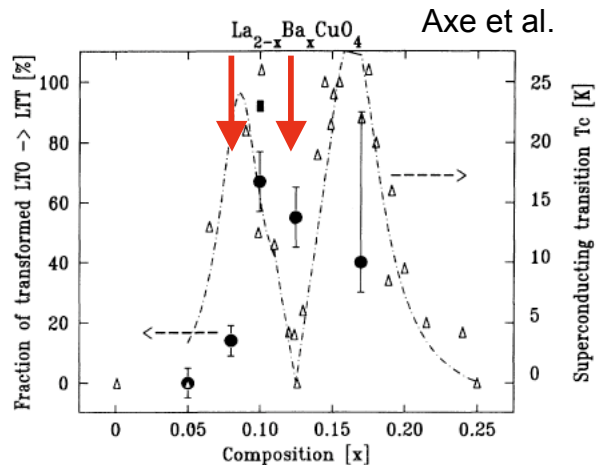
Samples



Robust stripe order

No stripe order

Stripe order expected



Instruments

MERIX@APS

Nickelate

BL11XU@SP8

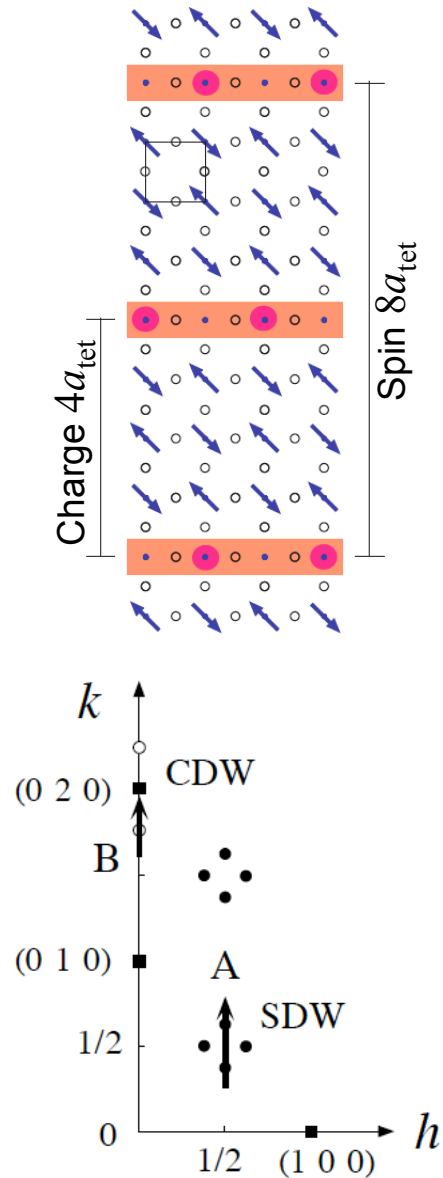
Cuprates

Horizontal scattering plane

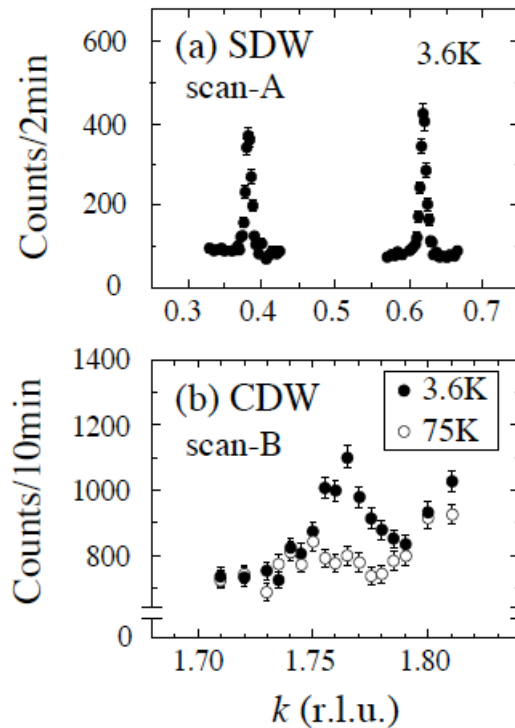
La_{1.875}Ba_{0.125}CuO₄ sample

Single crystal

- ◆ Grown by TSFZ method.
- ◆ Cut in a disk shape with the c-axis normal to the disk.
- ◆ Stripe order was checked by neutron.

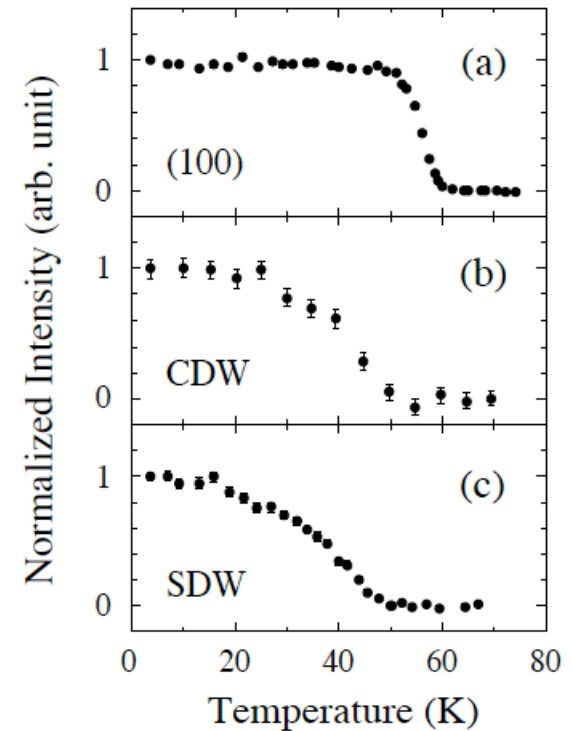


La_{1.875}Ba_{0.125}CuO₄, $\omega=0\text{meV}$

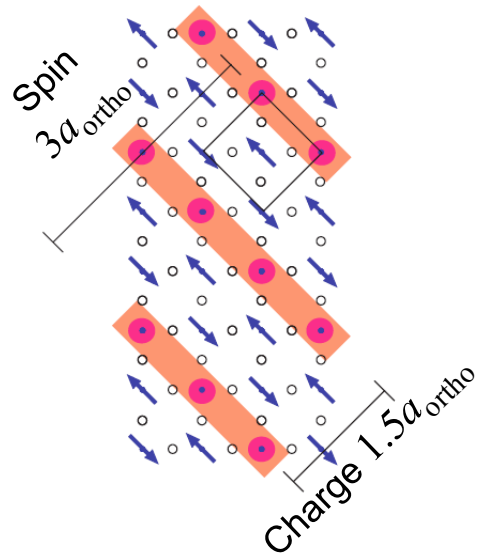


Fujita et al.

La_{1.875}Ba_{0.125}CuO₄, $\omega=0\text{meV}$

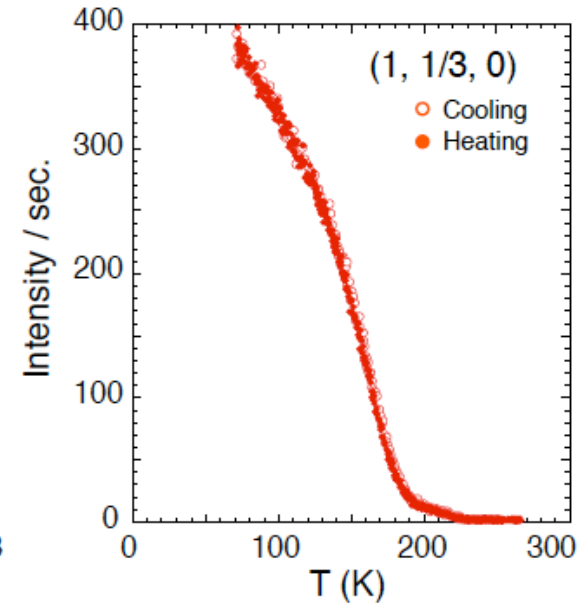
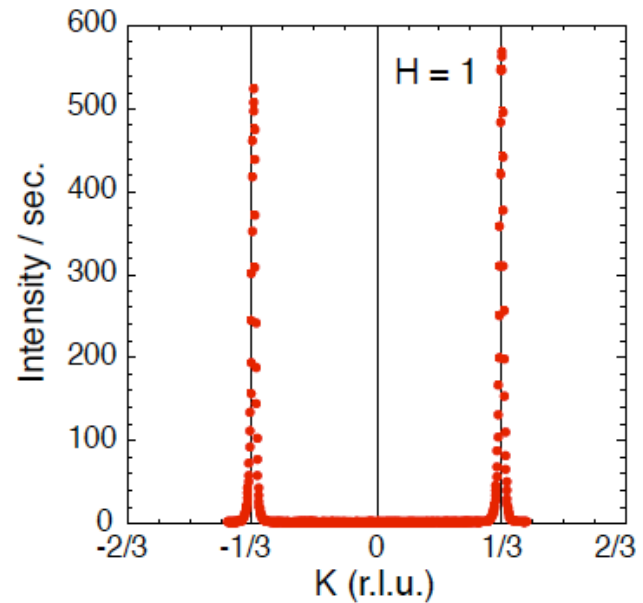
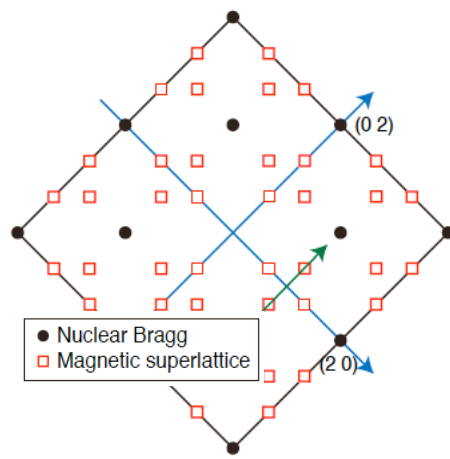


La_{5/3}Sr_{1/3}NiO₄ sample



Single crystal

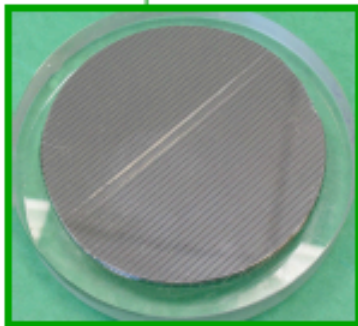
- ◆ Grown by TSFZ method.
- ◆ Cut in a disk shape with the c-axis normal to the disk.
- ◆ Stripe order was checked by neutron.



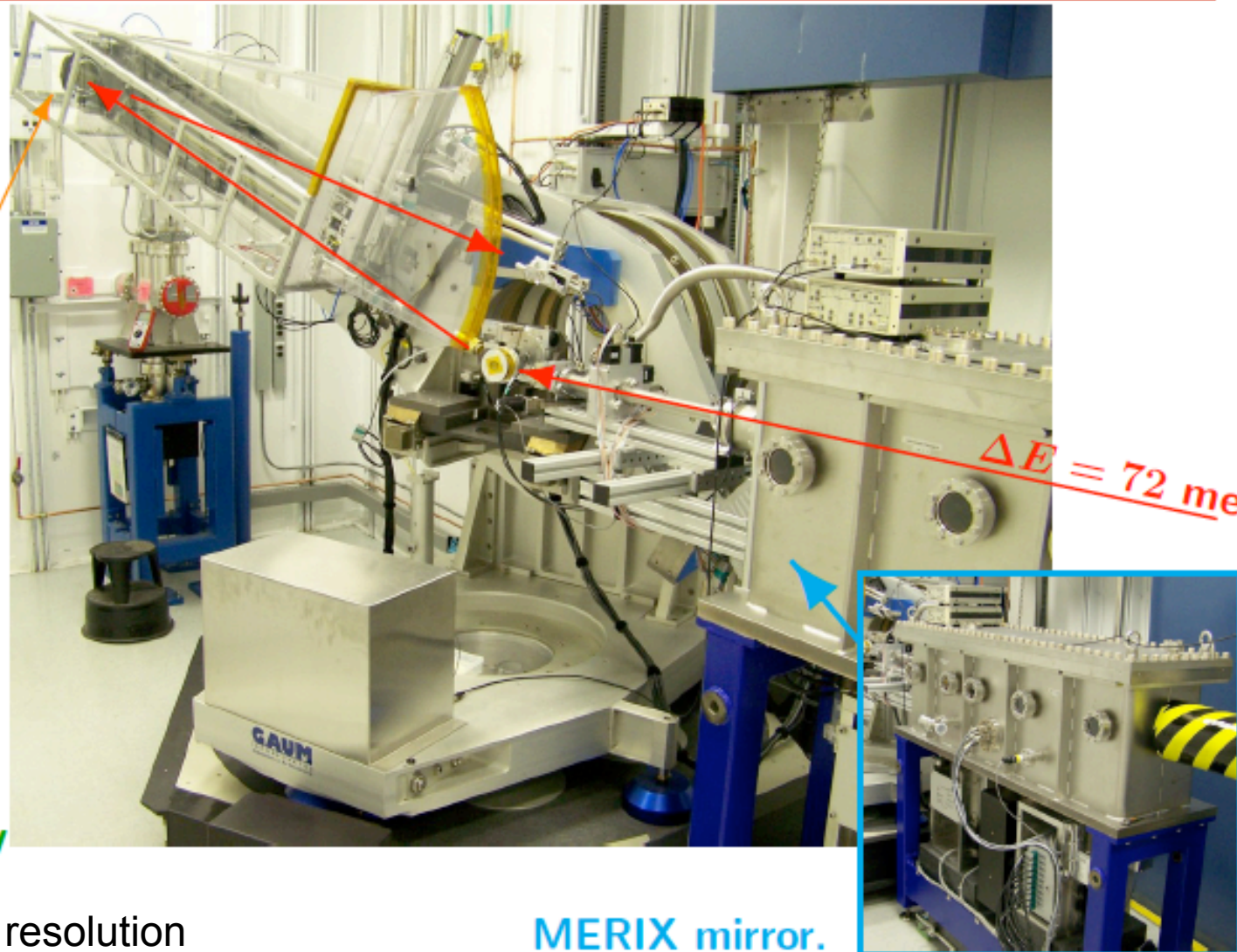
MERIX Spectrometer@30-ID.APS

(LSNO experiments)

Analyzer gimbal



Ge(337)
diced analyzer:
 $\Delta E = 115 \text{ meV}$

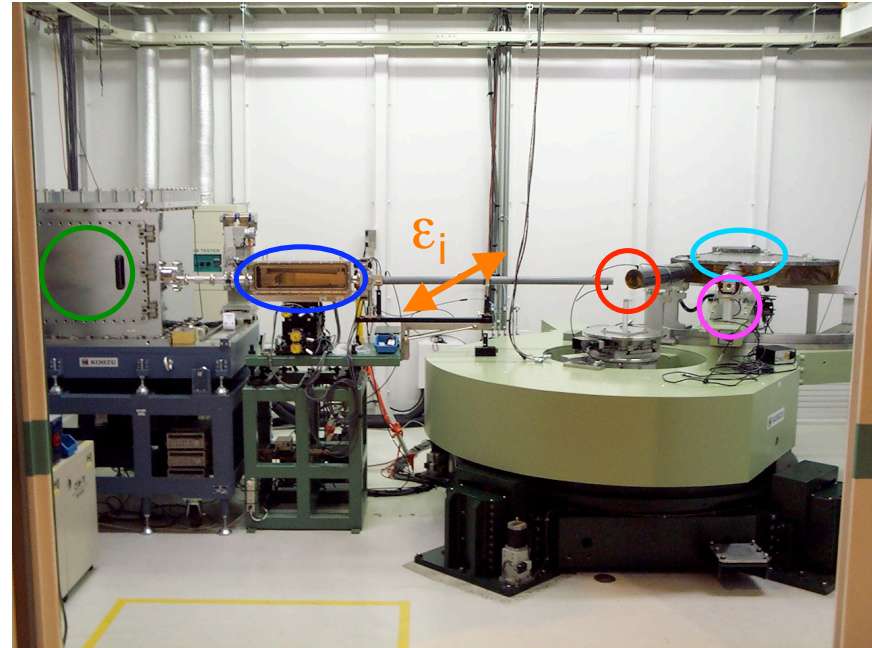
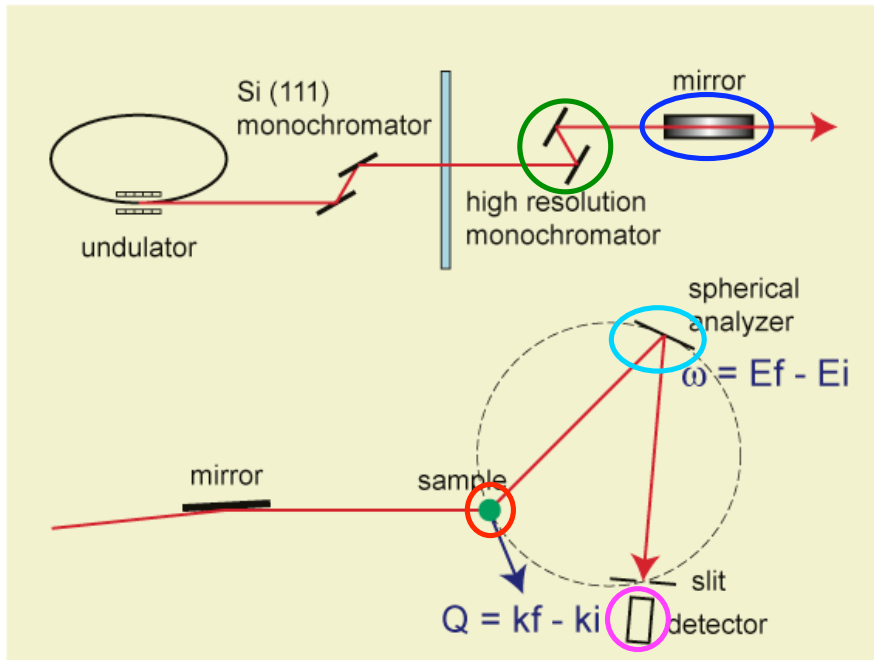


$\Delta E = 72 \text{ meV}$

MERIX mirror.
Focus: $5 \mu\text{m}$ (V) $\times 40 \mu\text{m}$ (H)

- ◆ Fine energy resolution
Cu K-edge : $\Delta E = 110 \text{ meV}$
Ni K-edge : $\Delta E = 150 \text{ meV}$ (Ge(642) analyzer)
- ◆ High efficiency by line detector

BL11XU at SPring-8 (L(B,S)CO experiments)

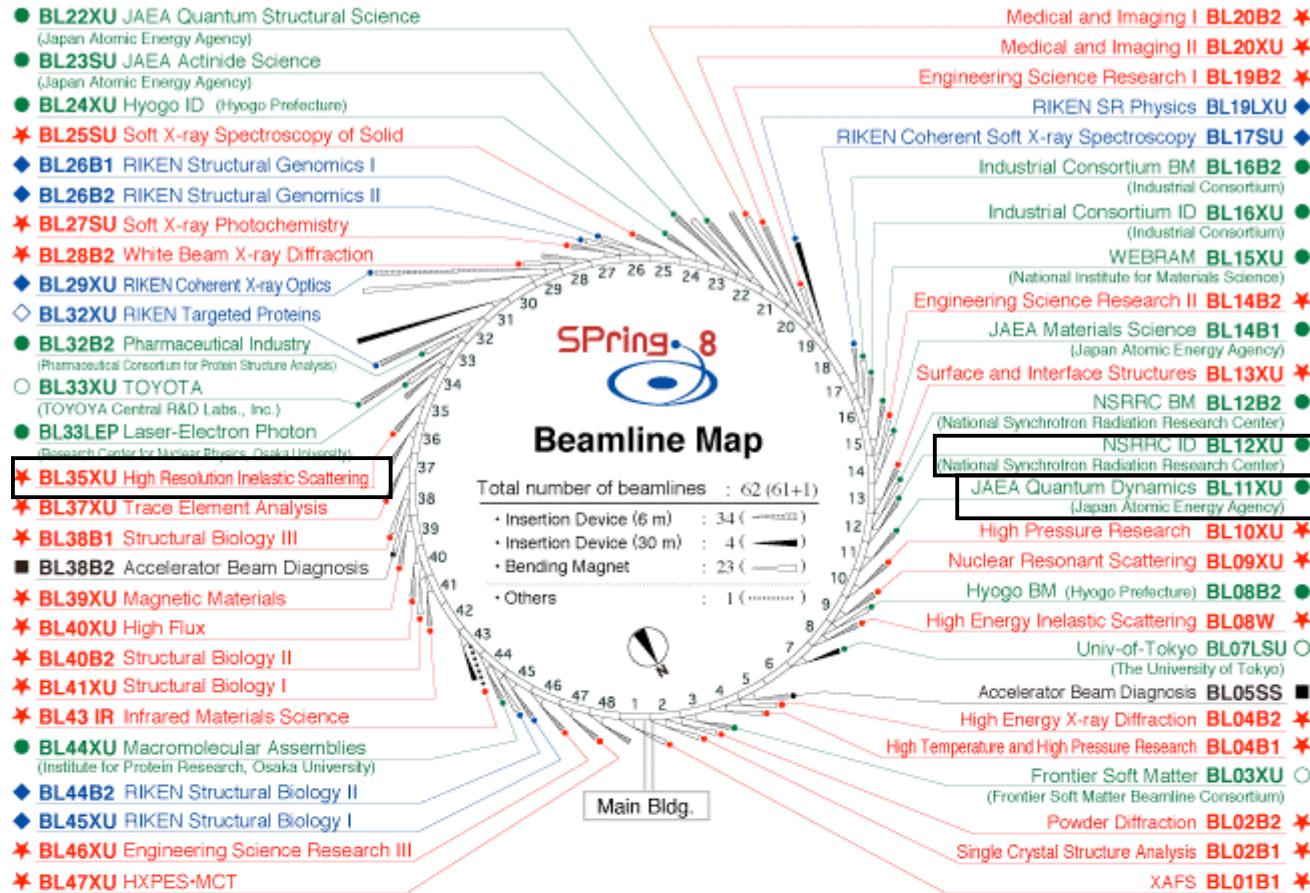


- Horizontal Scattering geometry
($\epsilon_i //$ scattering plane)
- Polarization analyzer
- Sample condition
 - ✓ Temperature 10 - 400 K
 - ✓ Magnetic Field < 8 T

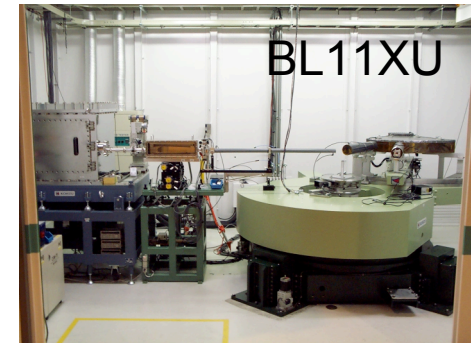
Energy resolution in practical set up

element	mono.	analyzer	ΔE (meV)
Cu	Si (400)	Ge(733)	~ 400
Mn		Ge(531)	~ 300
V		Ge(422)	~ 300

IXS beamlines at SPring-8



- medium resolution
BL11XU ... JAEA
BL12XU ... Taiwan NSRRC
(contact beamlines)
 $\Delta E \sim 100$ meV
charge excitations

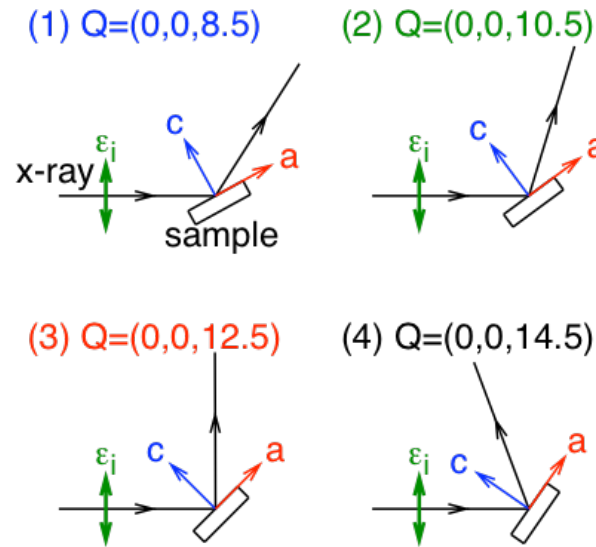
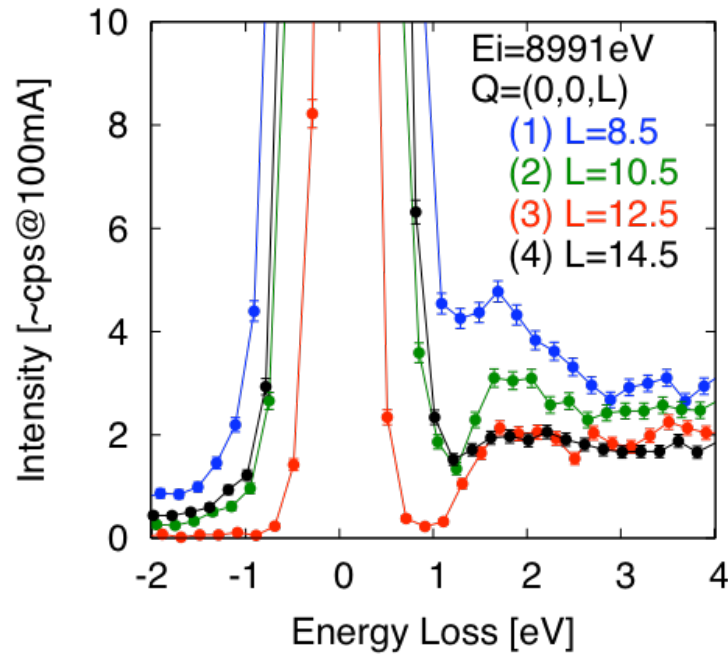


- high resolution
BL35XU ... public beamline
 $\Delta E \sim 1$ meV
phonon

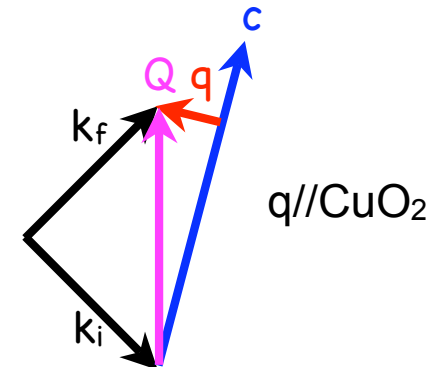
Scattering Geometry

Ishii et al.

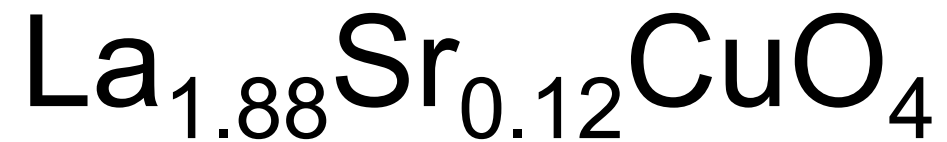
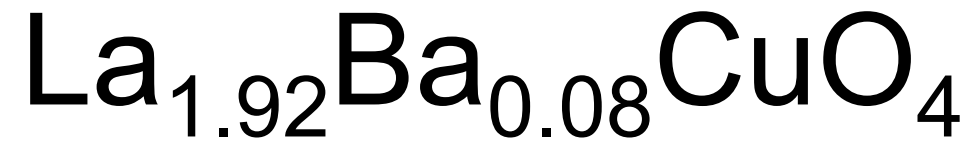
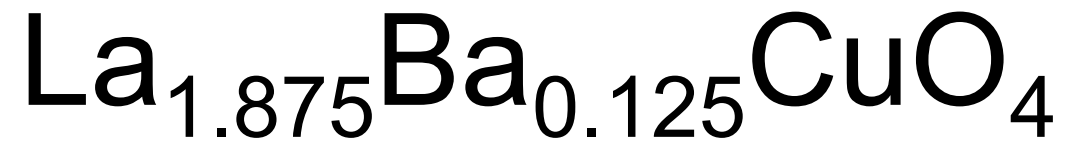
Nd_2CuO_4 momentum scan along c^* -direction



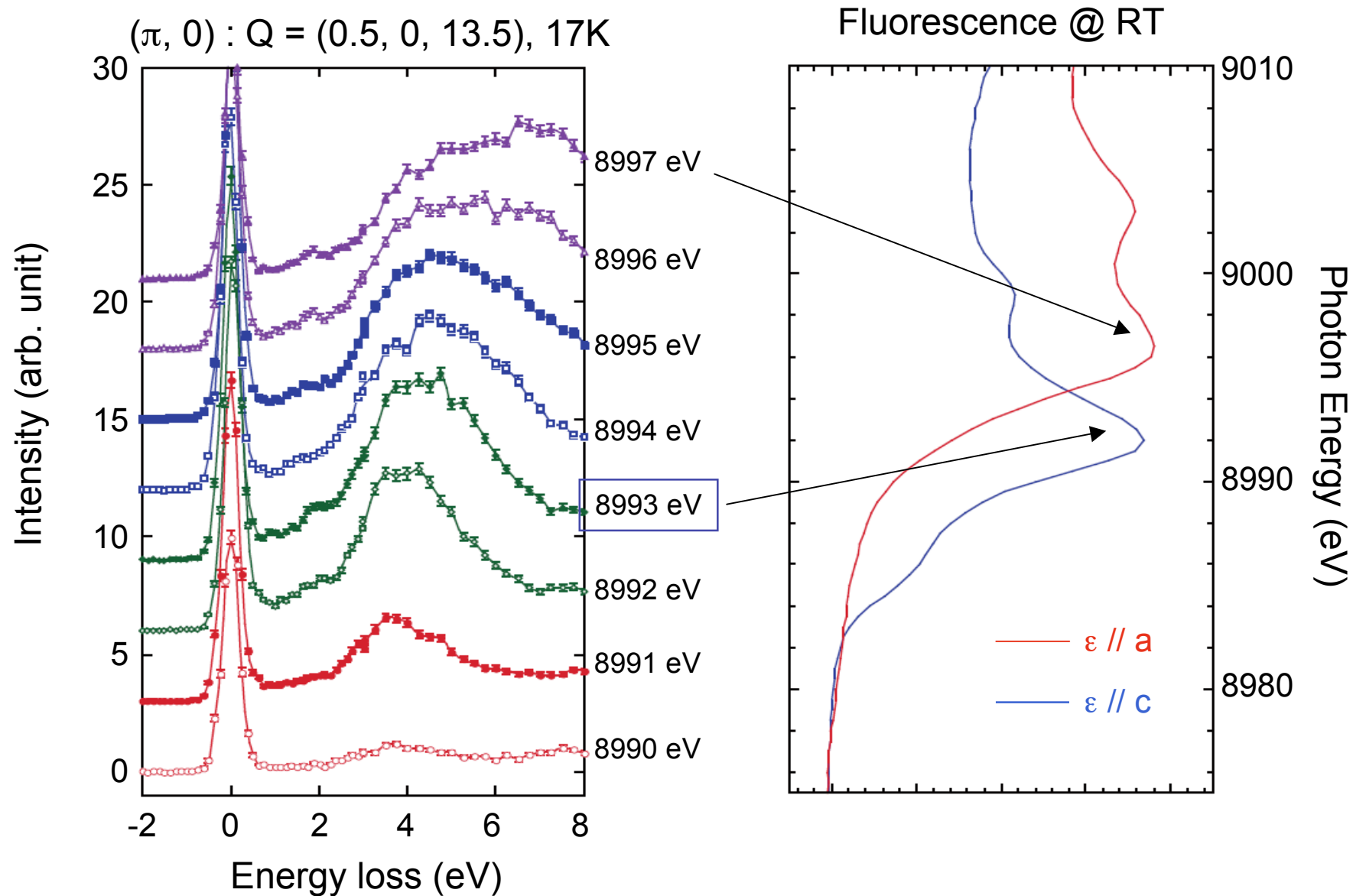
Transverse geometry



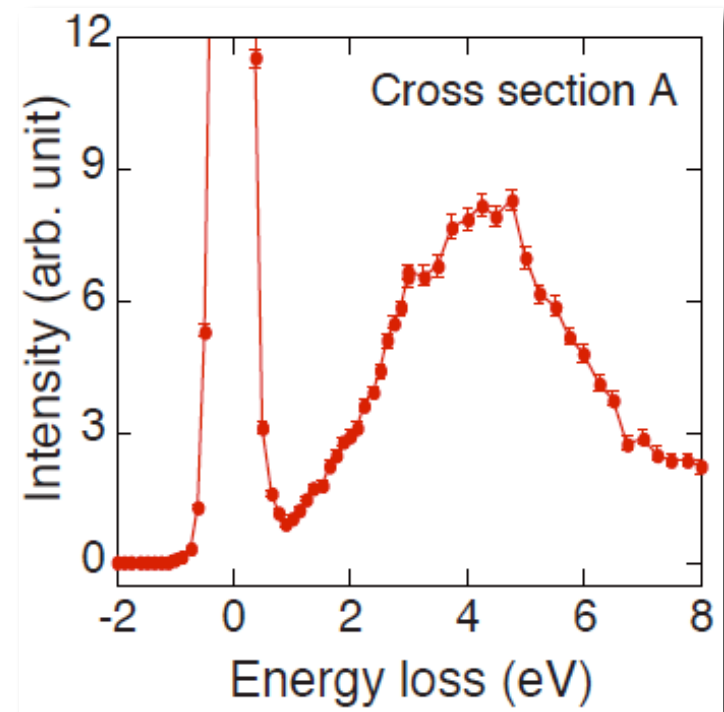
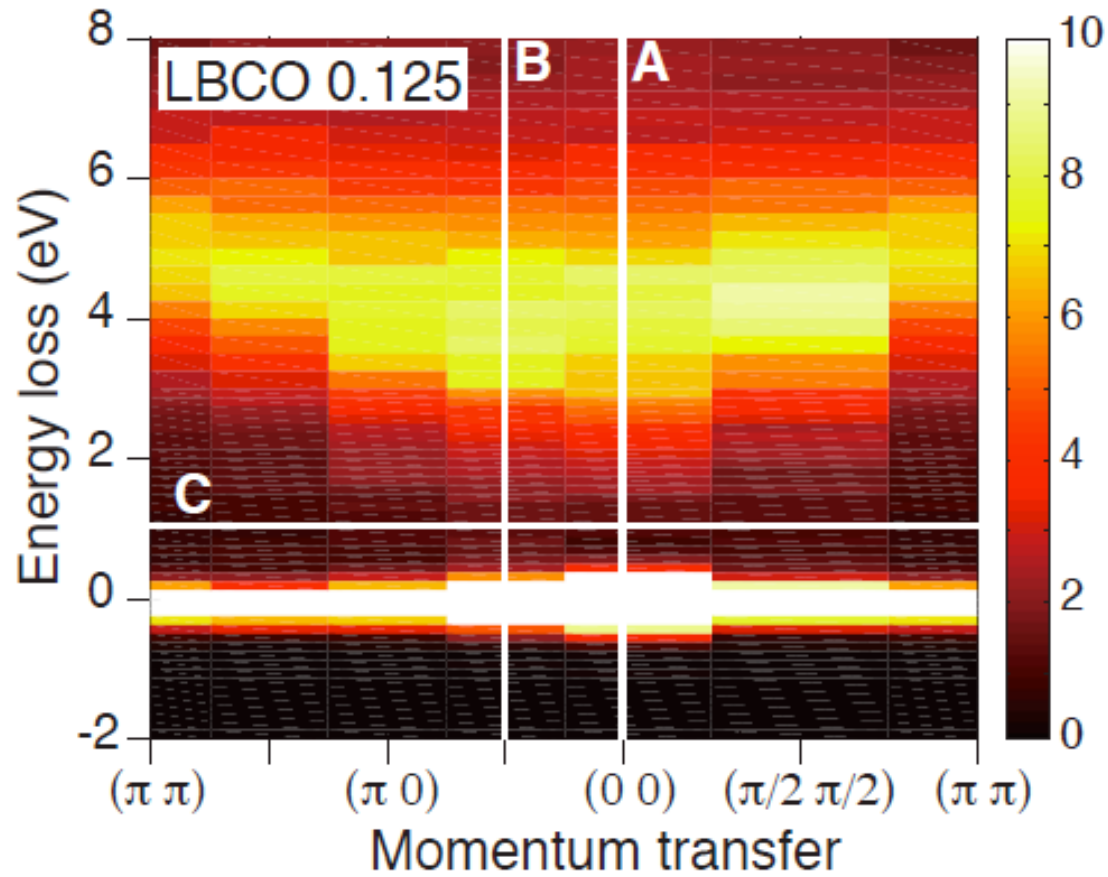
- Elastic intensity is minimum at $2\theta = 90^\circ$.
Clear gap feature at $Q = (0,0,12.5)$.
 q -scan in transverse geometry
 \Rightarrow work well for low-dimensional materials



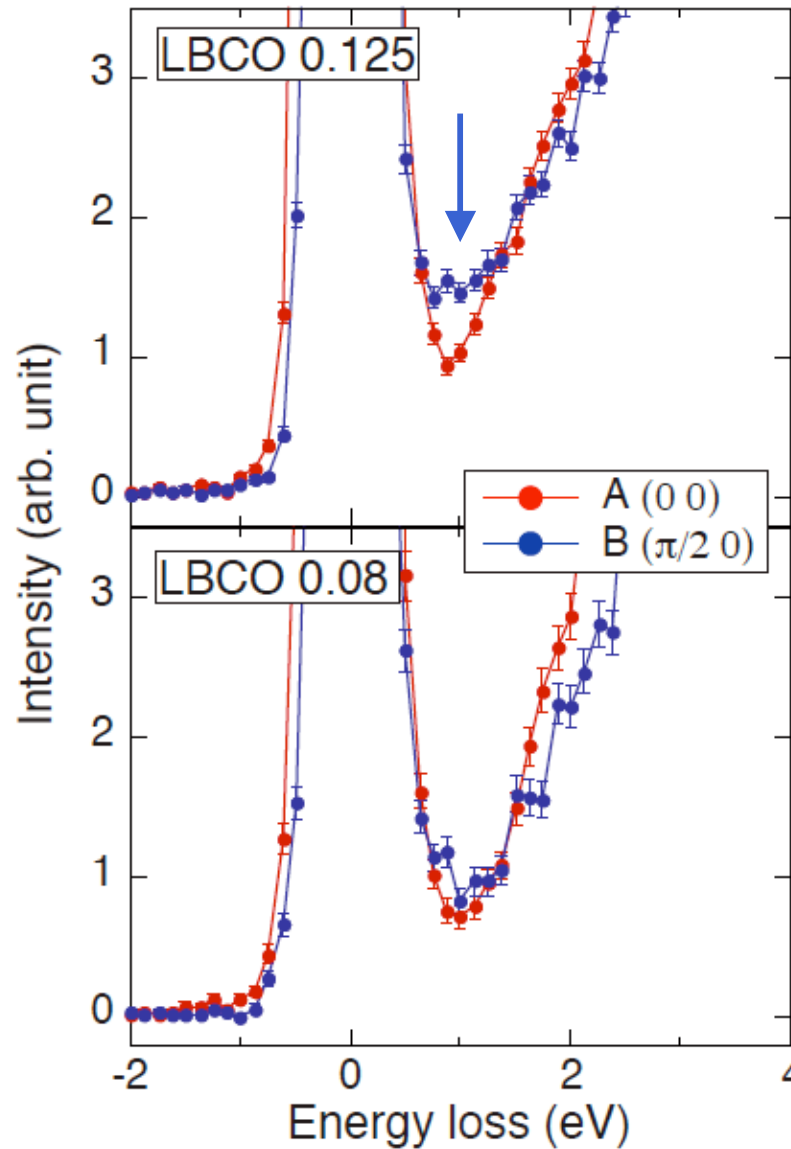
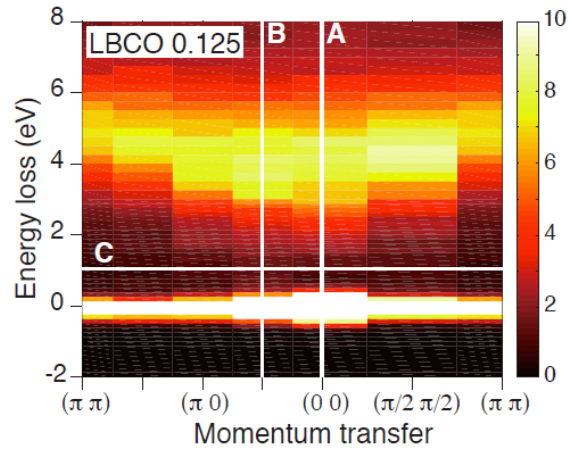
E_i - dependence : $\text{La}_{1.875}\text{Ba}_{0.125}\text{CuO}_4$



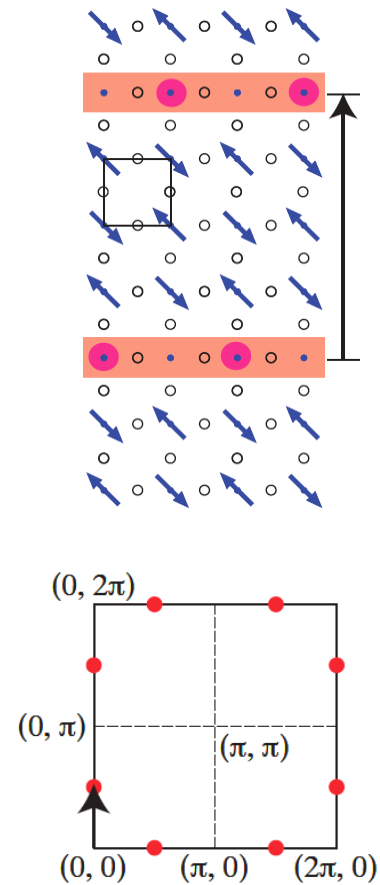
Overall spectra



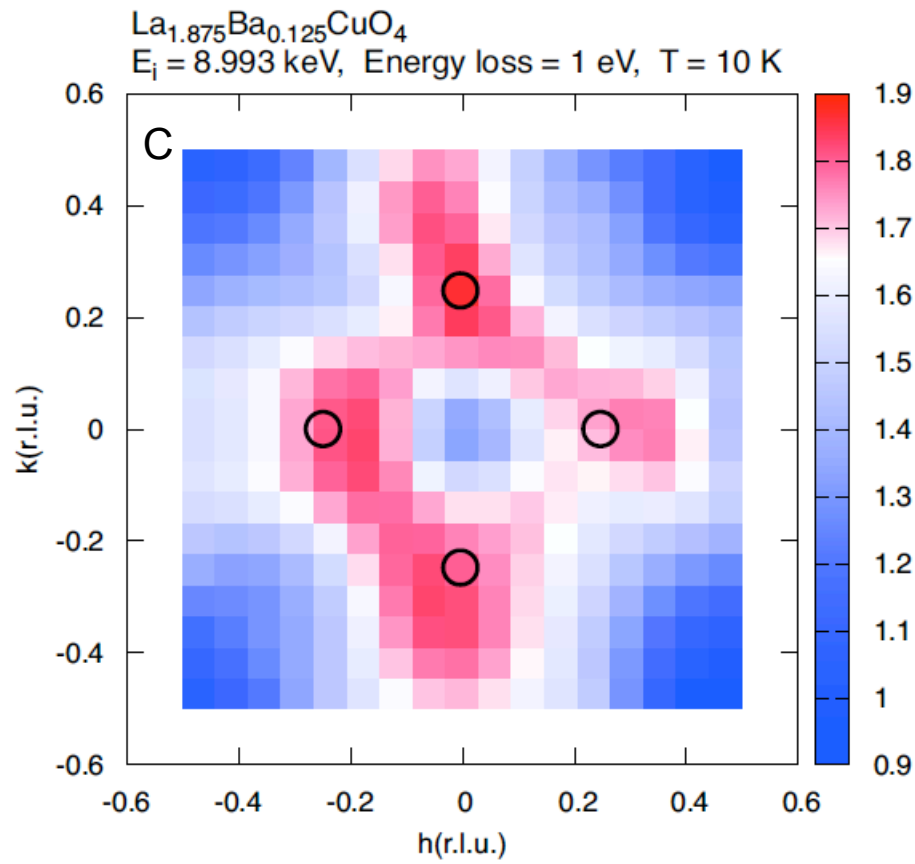
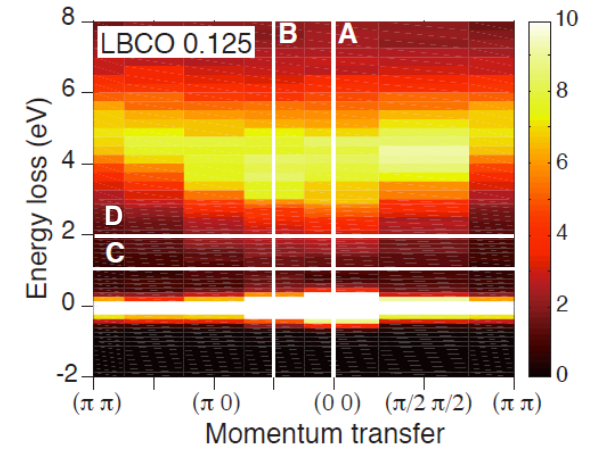
Comparison between $(0, 0)$ & q_s



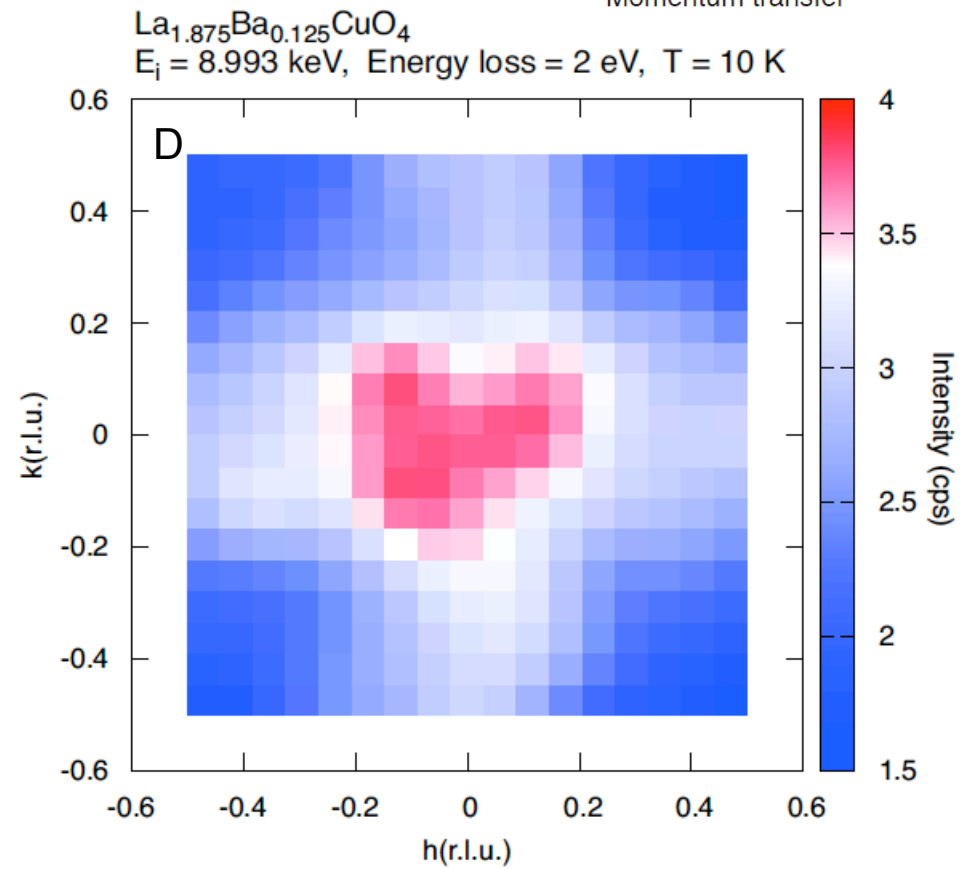
Stripe spacing :
 $d_s = (0, 4a)$
 Stripe q :
 $q_s = 2\pi(0, 1/4)$



Excitation at q_s

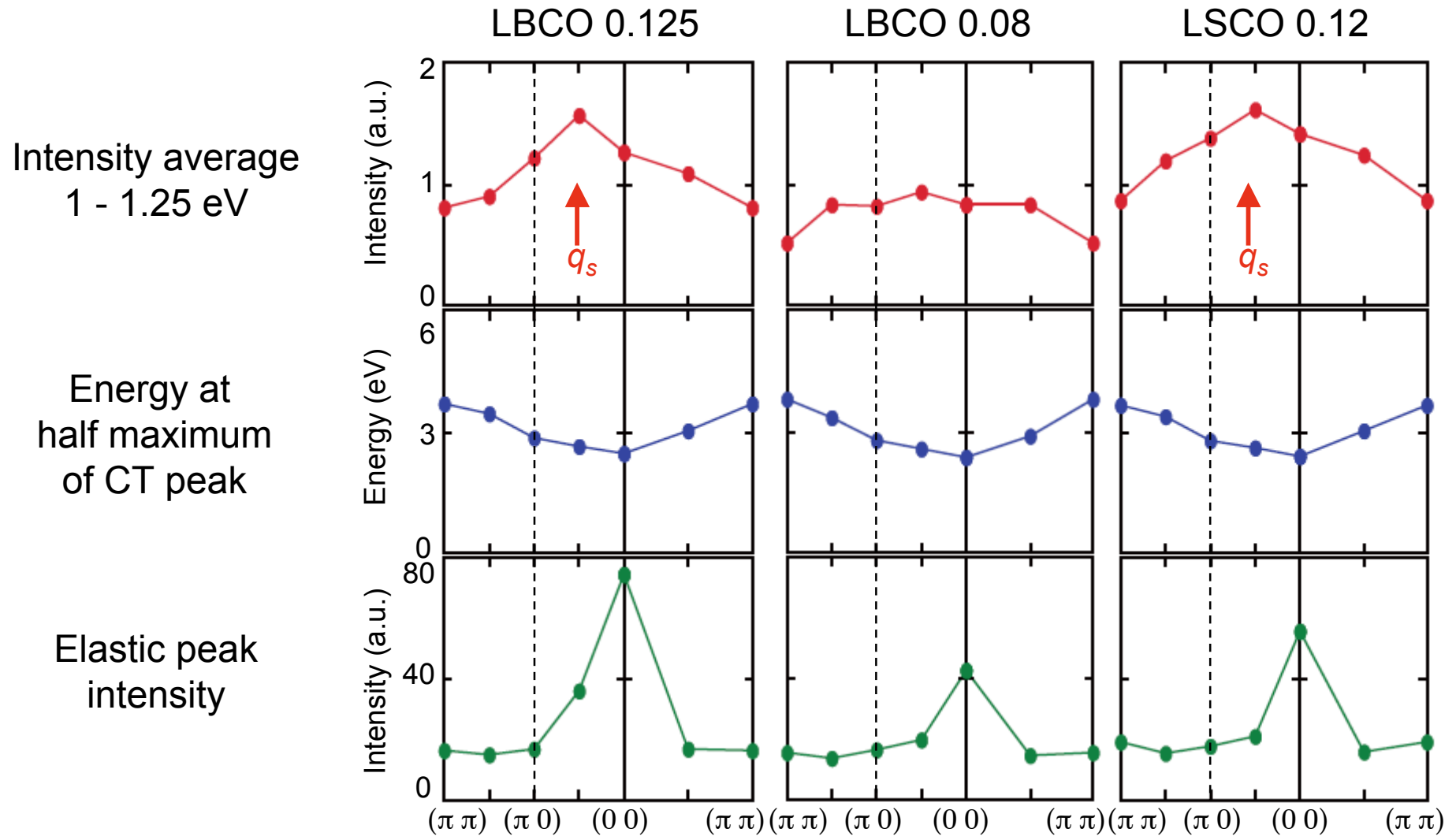


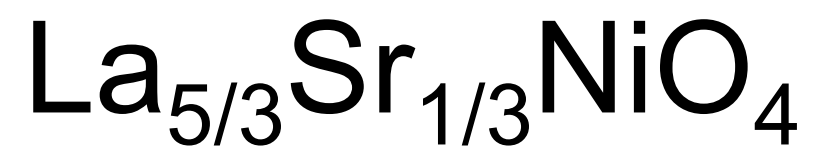
1 eV



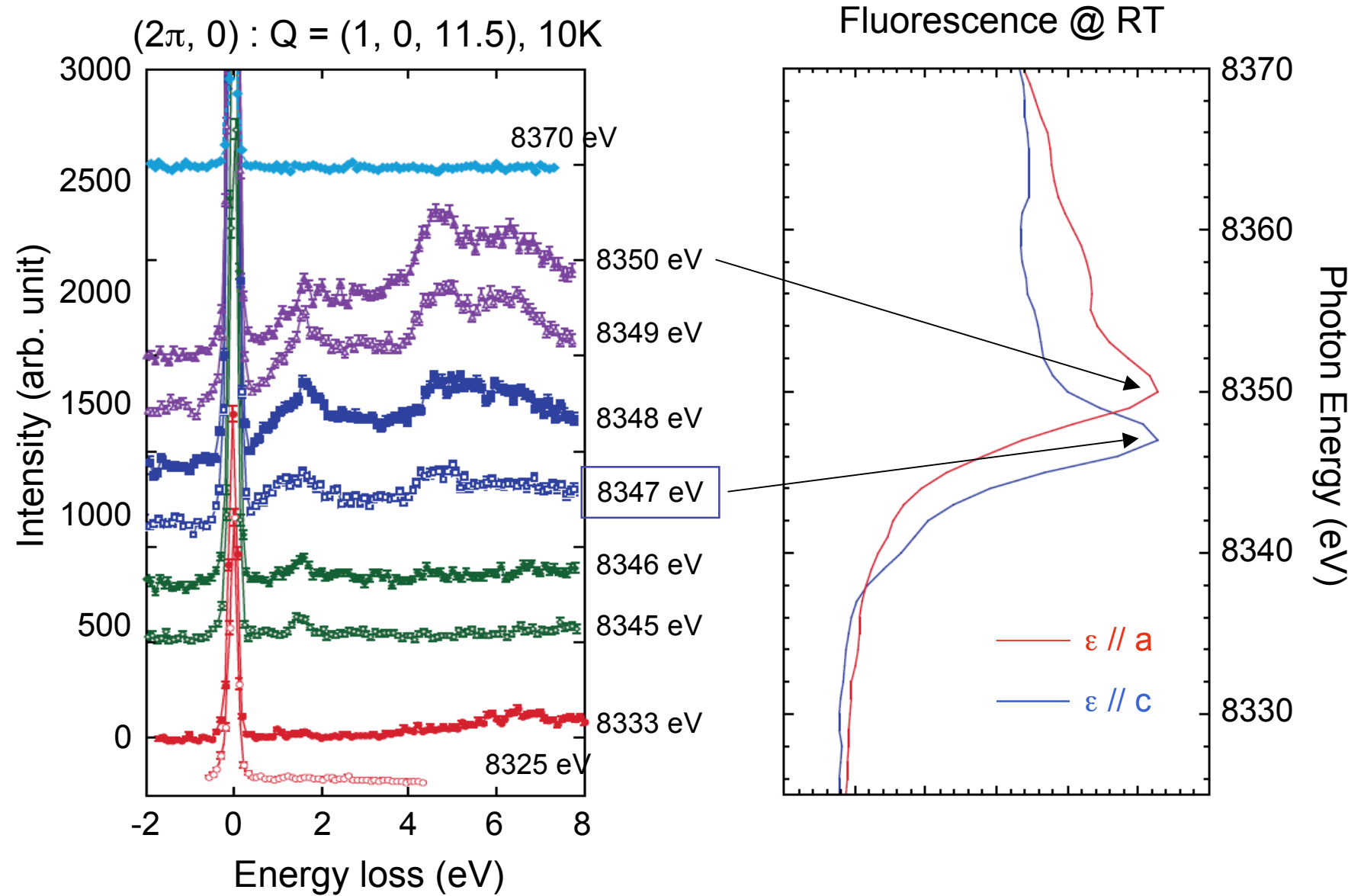
2 eV

q - dependence (2)

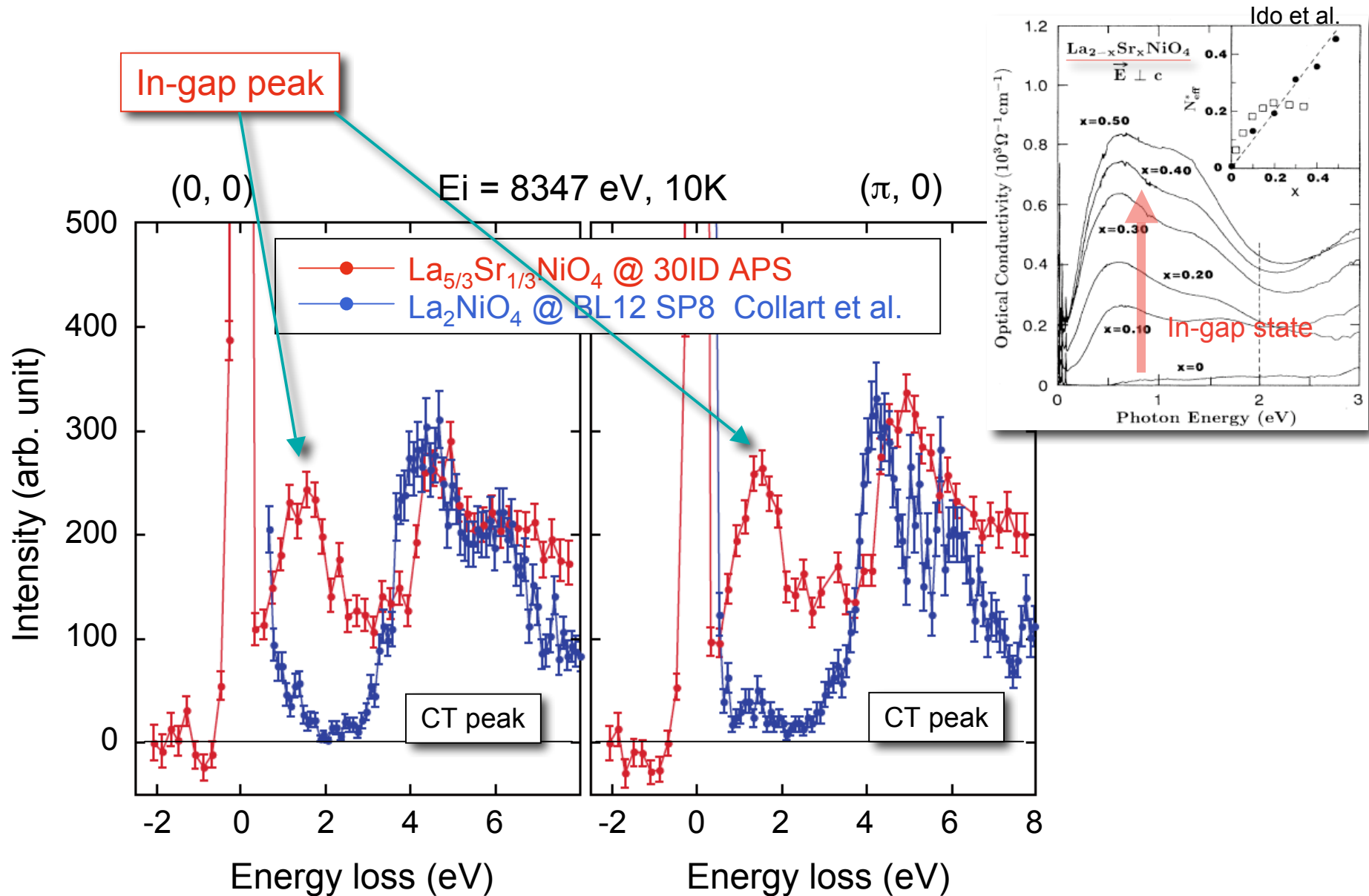




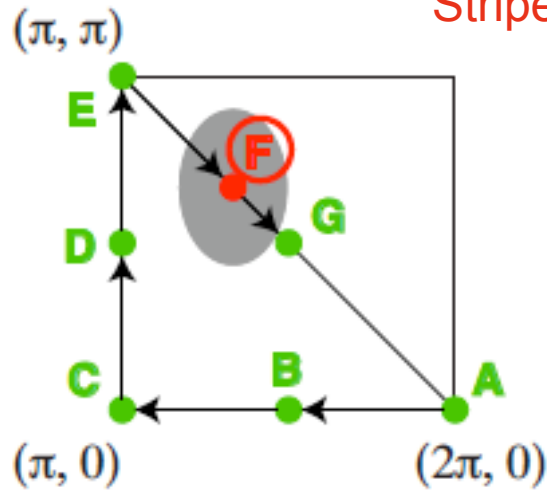
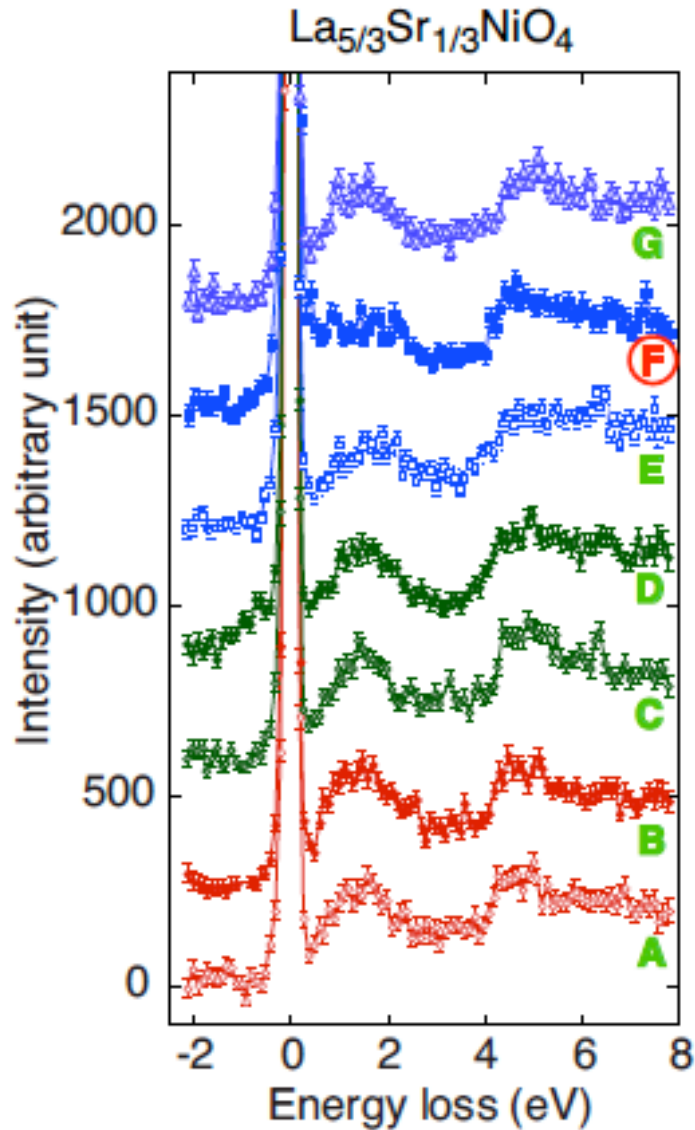
E_i - dependence



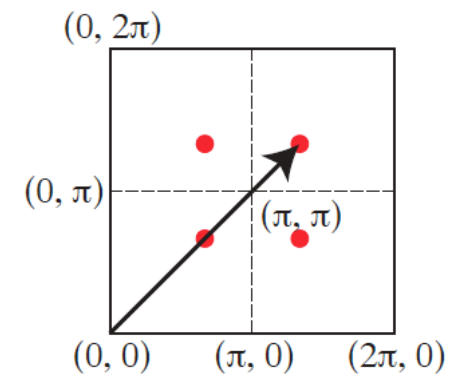
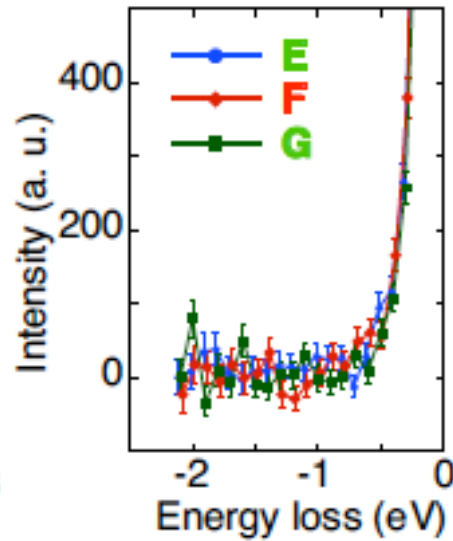
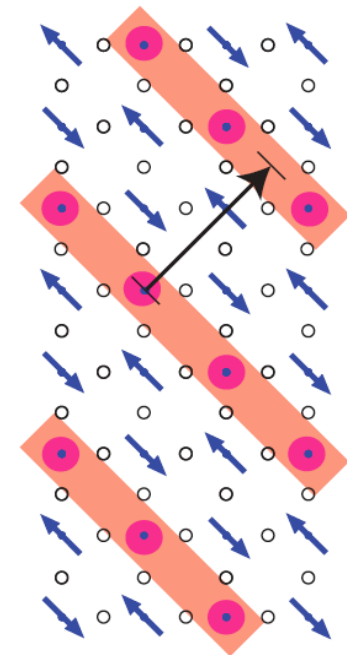
Comparison to the non-doped La_2NiO_4



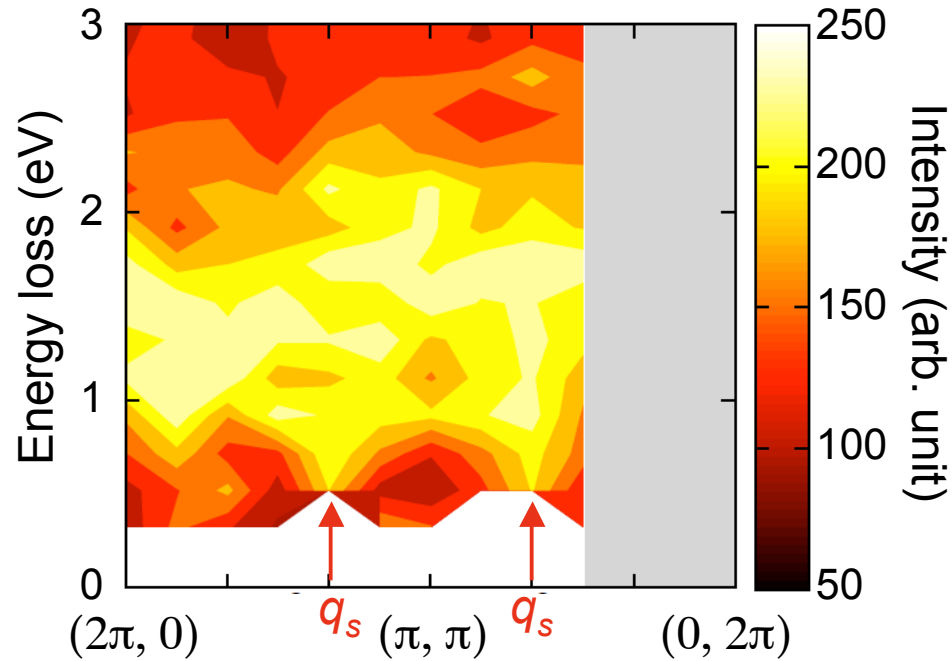
q - dependence (1)



Stripe spacing : $d_s = (3a/2, 3a/2)$
 Stripe q : $q_s = 2\pi(2/3, 2/3)$

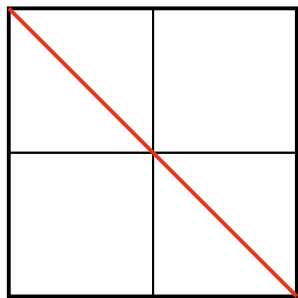


q - dependence (2)



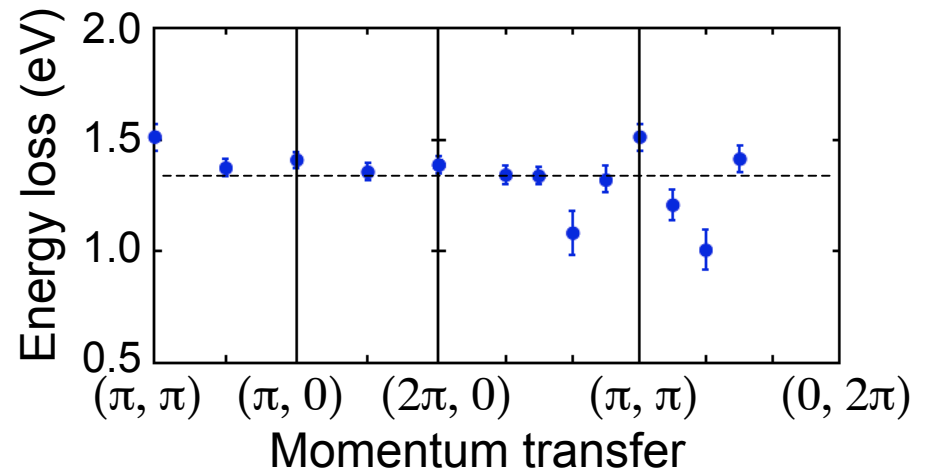
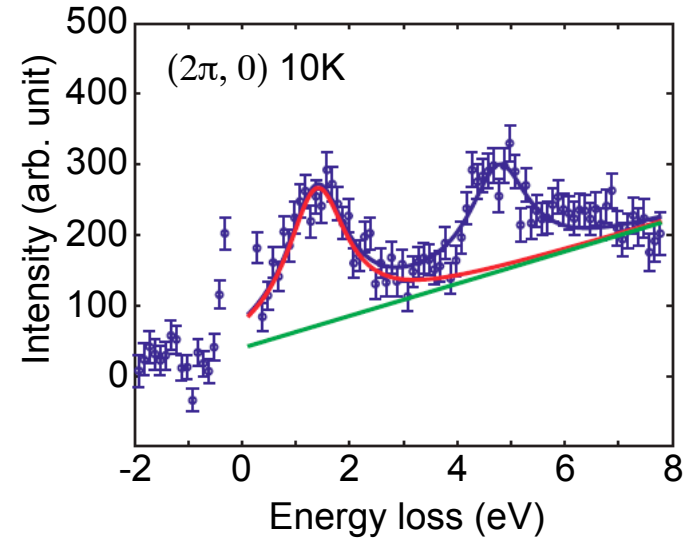
Momentum transfer

$(0, 2\pi)$

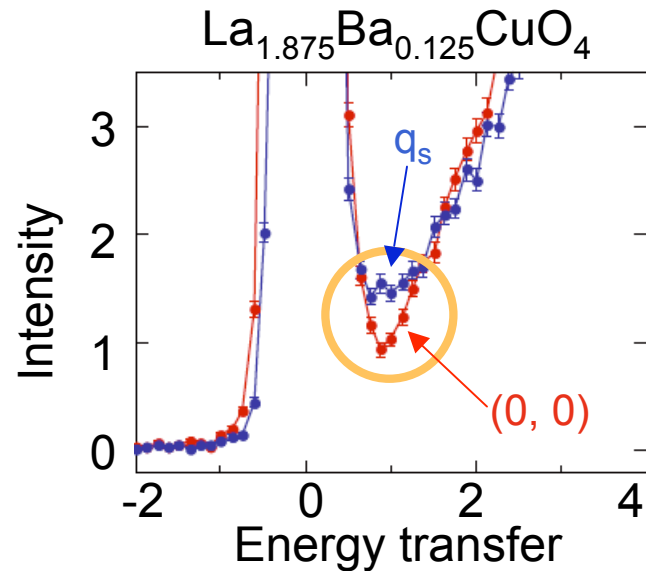


$(2\pi, 0)$

Fit by Lorentzian function



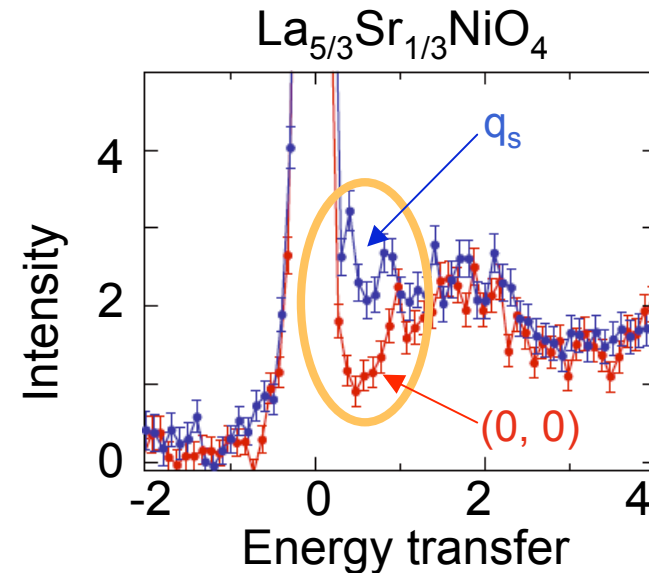
Experimental results summary



Cuprates

BL11XU 300meV resolution

- ◆ In-gap state is not observed.
- ◆ Continuum intensity increases at q_s for LBCO 0.125 and LSCO 0.12.



LSNO 1/3

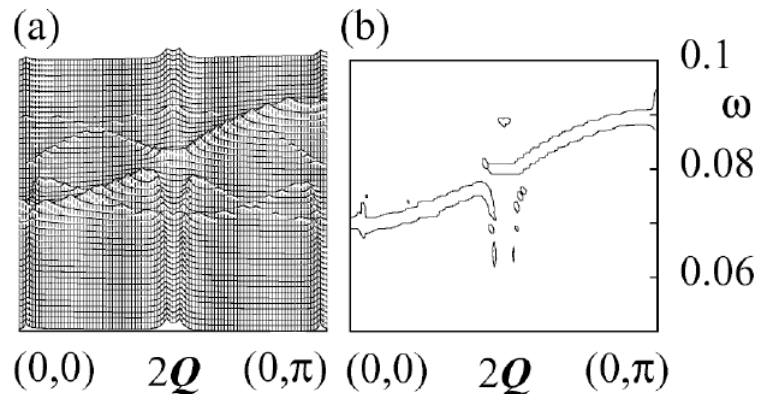
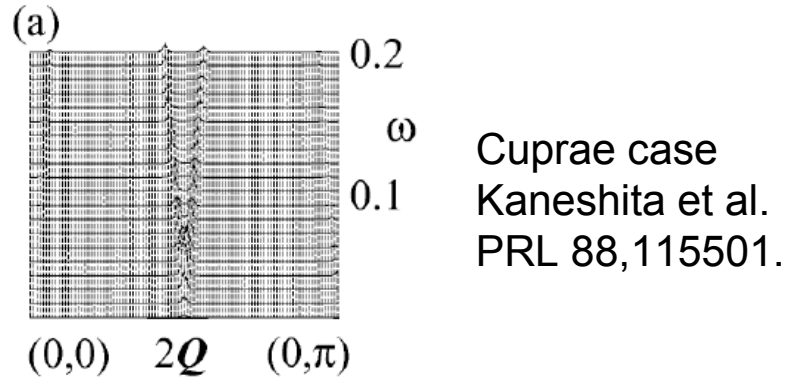
MERIX 150meV resolution

- ◆ In-gap state is clearly observed.
- ◆ Gap-like structure is absent at q_s .

Both nickelate and cuprate, stripe order gives additional spectral weight at q_s around 1 eV.

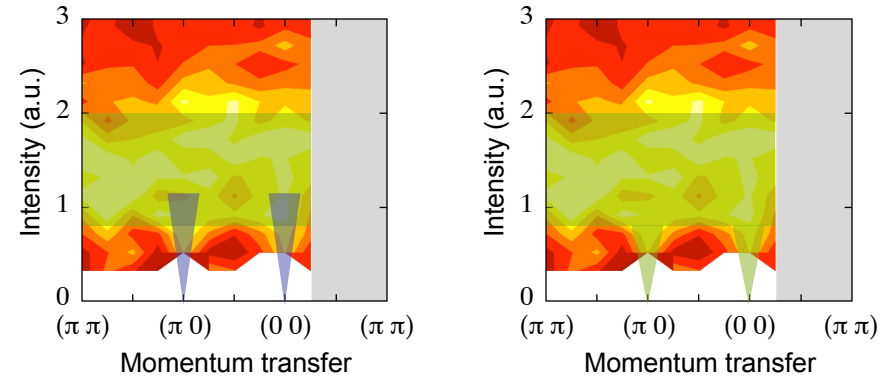
Possibilities

Collective charge excitation?



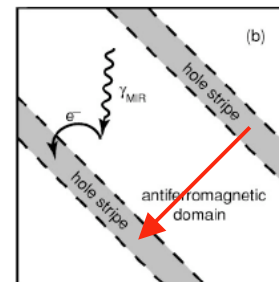
- ◆ Dynamic fluctuation of charge stripes produces collective charge excitation.
- ◆ In principle, this should be observed in non-resonant scattering.

Same or different ?



Stripe band excitation?

Homes et al.



- ◆ The excitation at the stripe- q corresponds to the charge excitation between charge stripes.

May such excitation produces small gap??
(Gap < 340 meV)

Other origin?

Does introduction of the stripe structure into the models account for the observation?

Summary

- Hard x-ray RIXS on charged ordered system.
- Charge ordered compounds show additional RIXS spectra at q_s at $\sim 1\text{eV}$.
- More doping dependence, more material dependence will be studied soon.
- Soft x-ray RIXS can be an candidate to study this feature.