國家同步輻射研究中心
National Synchrotron Radiation Research Center

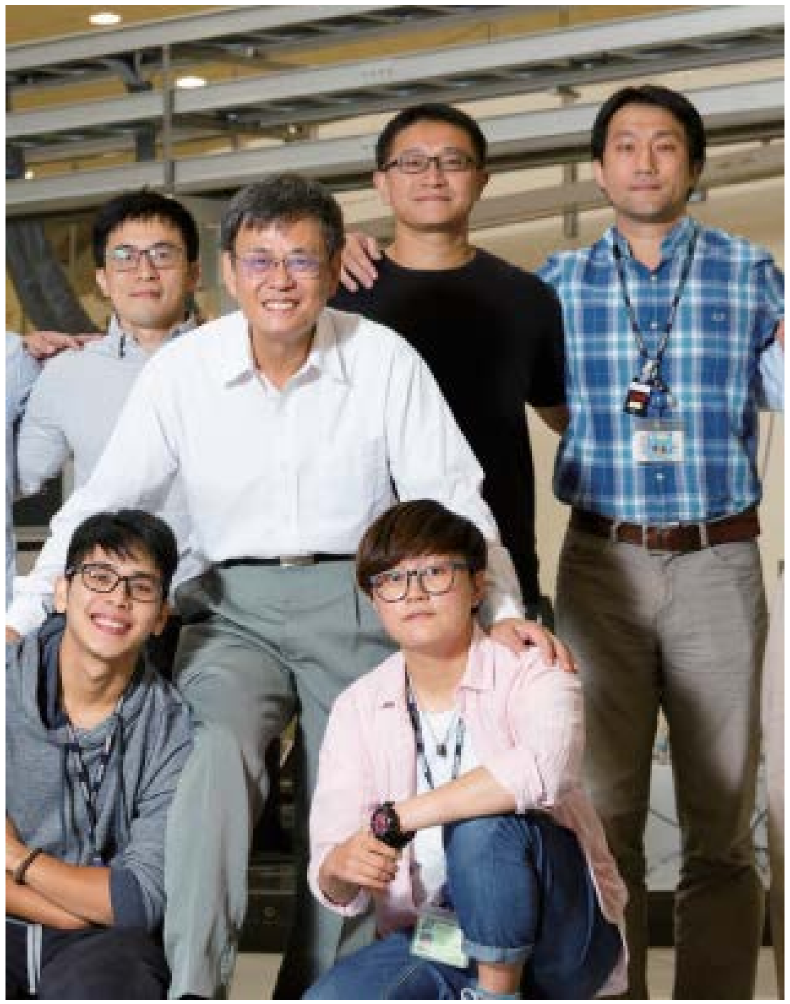
High-resolution momentium-resolved RIXS at Taiwan Photon Source

Di-Jing Huang

NSRRC, Taiwan

IXS 2019, June 26, 2019

Our Team

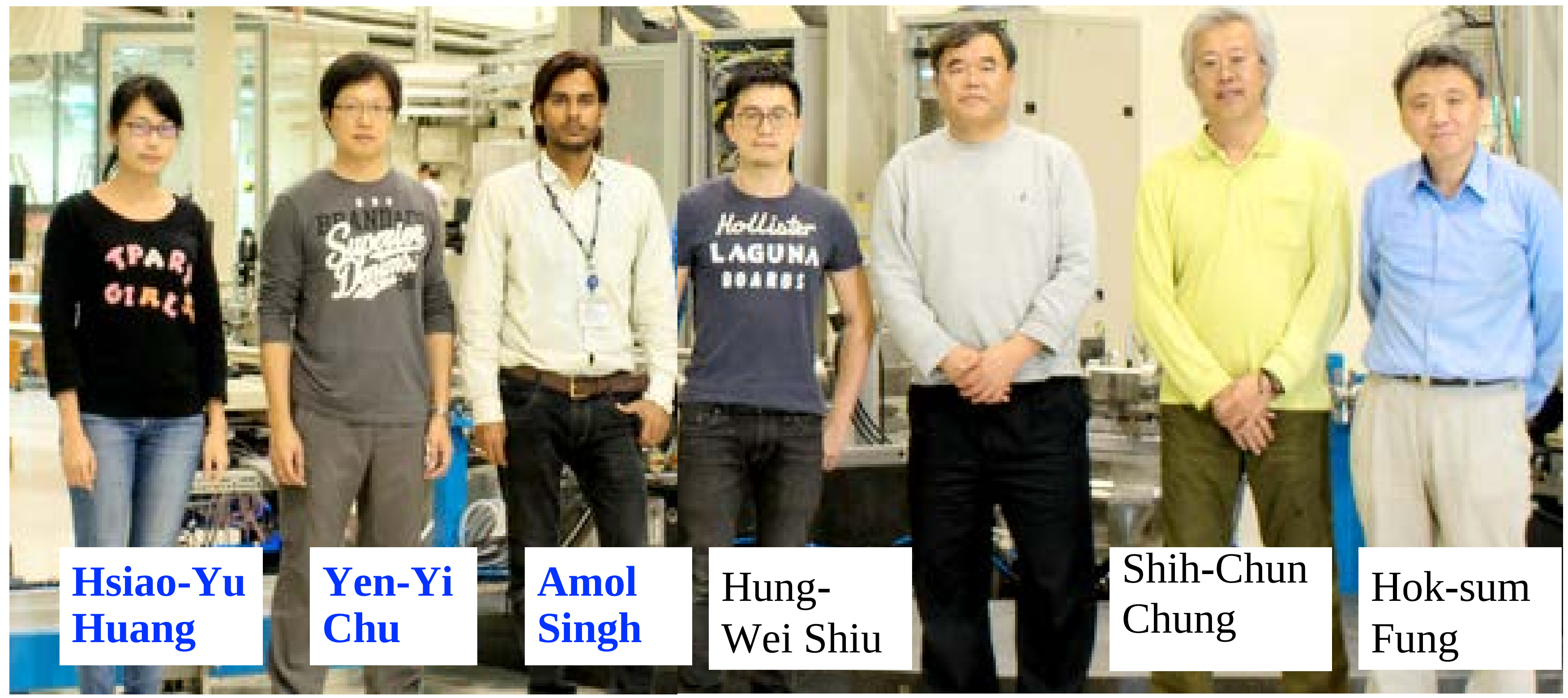


Chih-Yu Hua, Kai-Yang Kao

Chien-Te Chen

Hsuan-Yao Chao

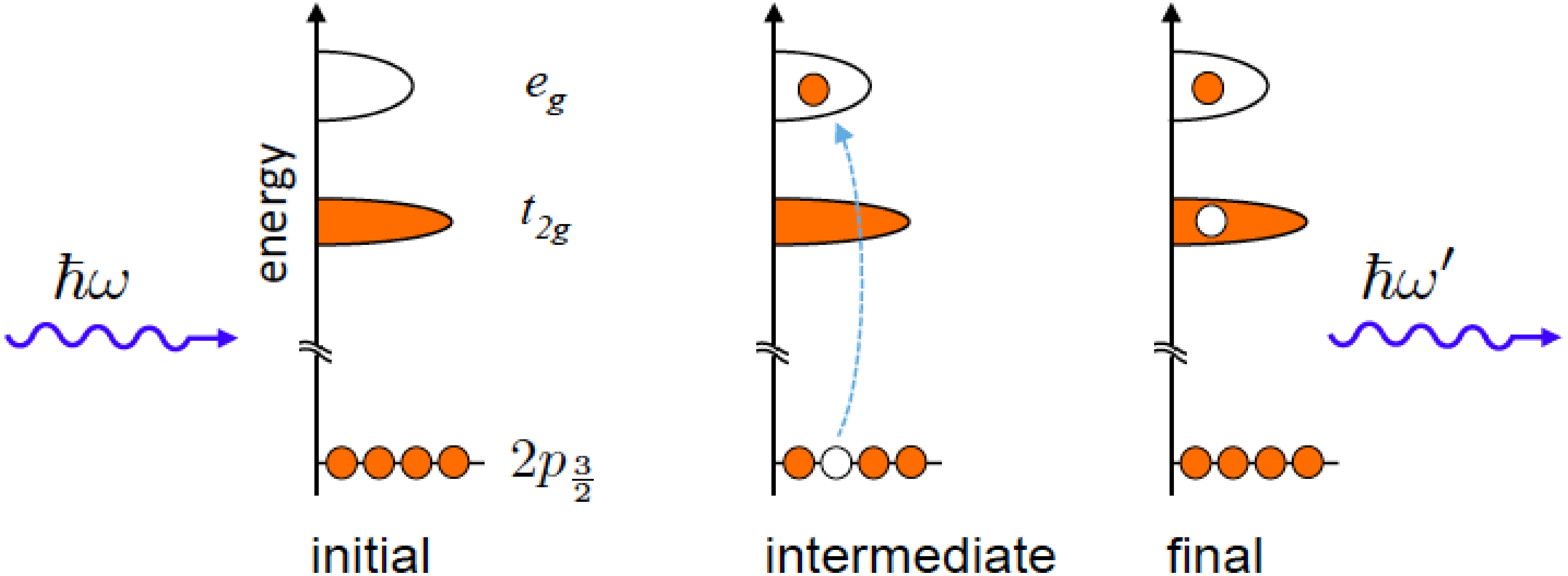
Shang-Ching Yeh



Seiki KOMIYA


National Synchrotron Radiation Research Center
Taiwan
1.5 GeV
Taiwan Light Source
3 GeV
Taiwan Photon Source

Resonant Inelastic X-ray Scattering (RIXS)

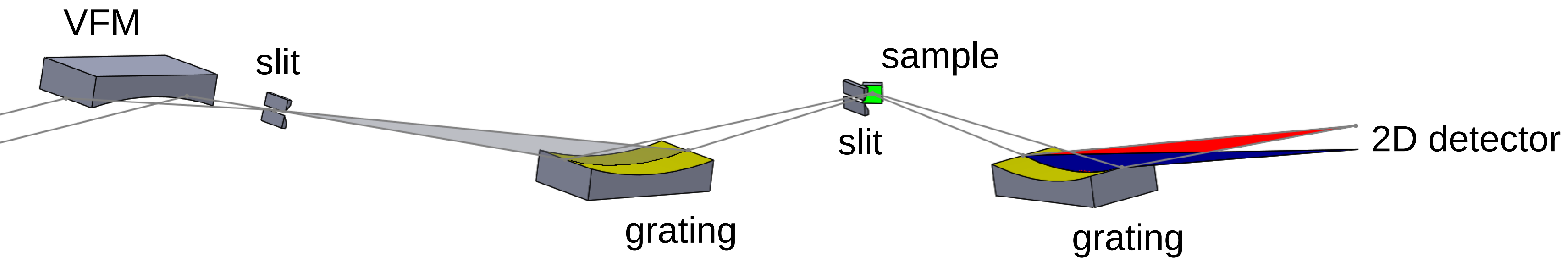


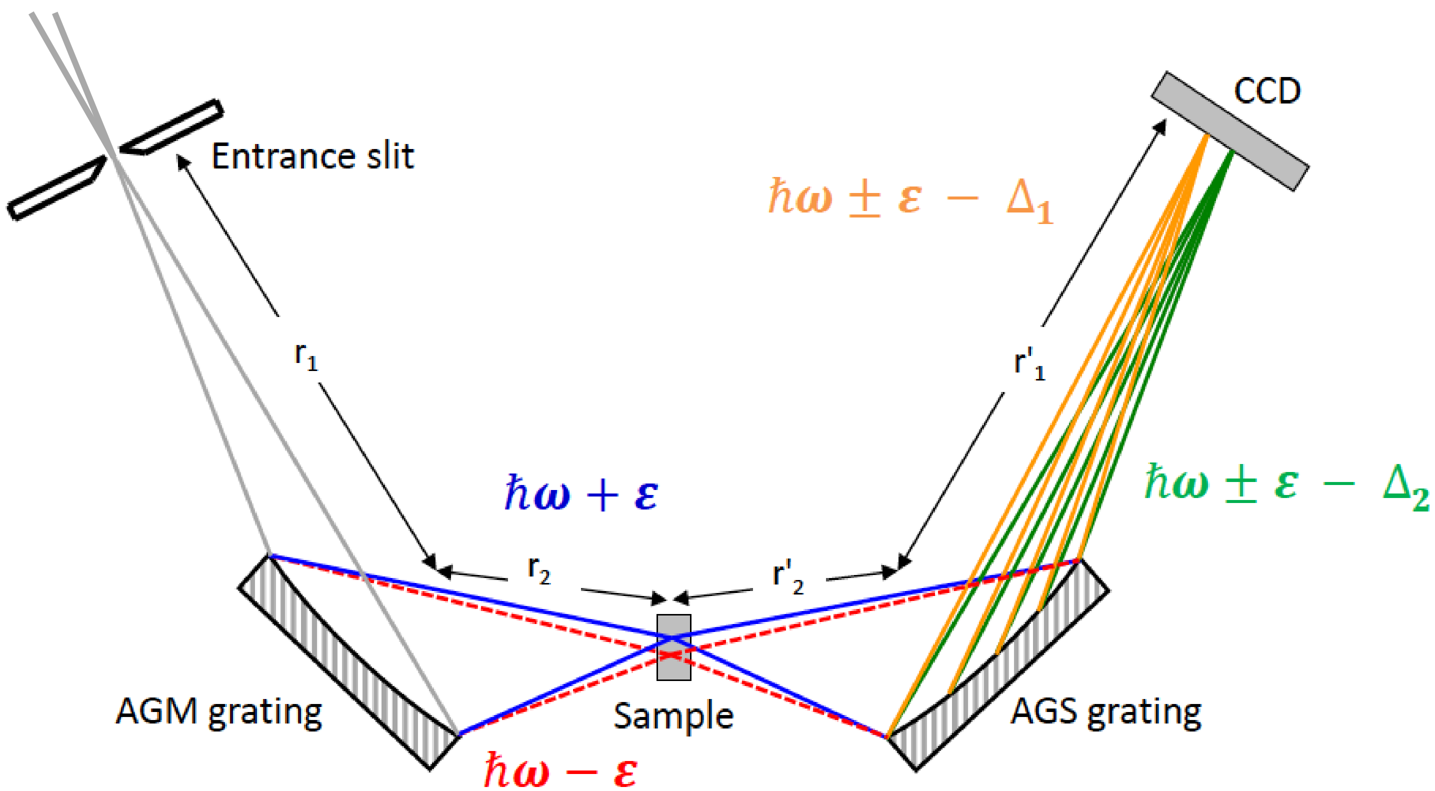
- two-particle correlations
- photon-in photon-out

resolution → ← count rate

Optical Concept of the AGM-AGS Beamline

Energy Compensation Principle



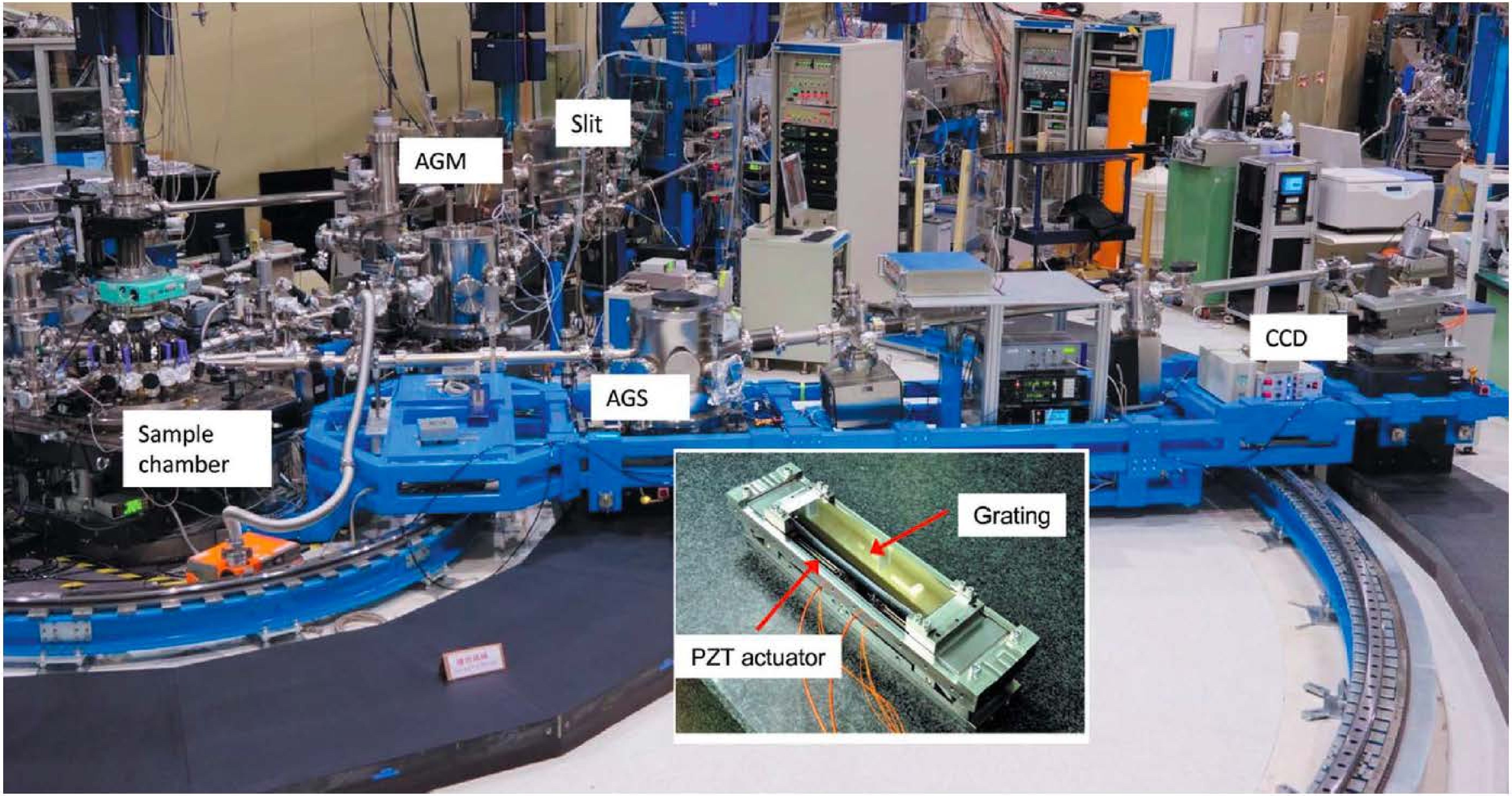


$$\Delta E_{\mathrm{total}} = \sqrt{\Delta E^2_{\mathrm{mono}} + \Delta E^2_{\mathrm{spectr}}}$$

”large” exit slit
e.g. 50 or 100 µm

H. S. Fung et al. AIP Conf. Proc. **705**, 655 (2004).

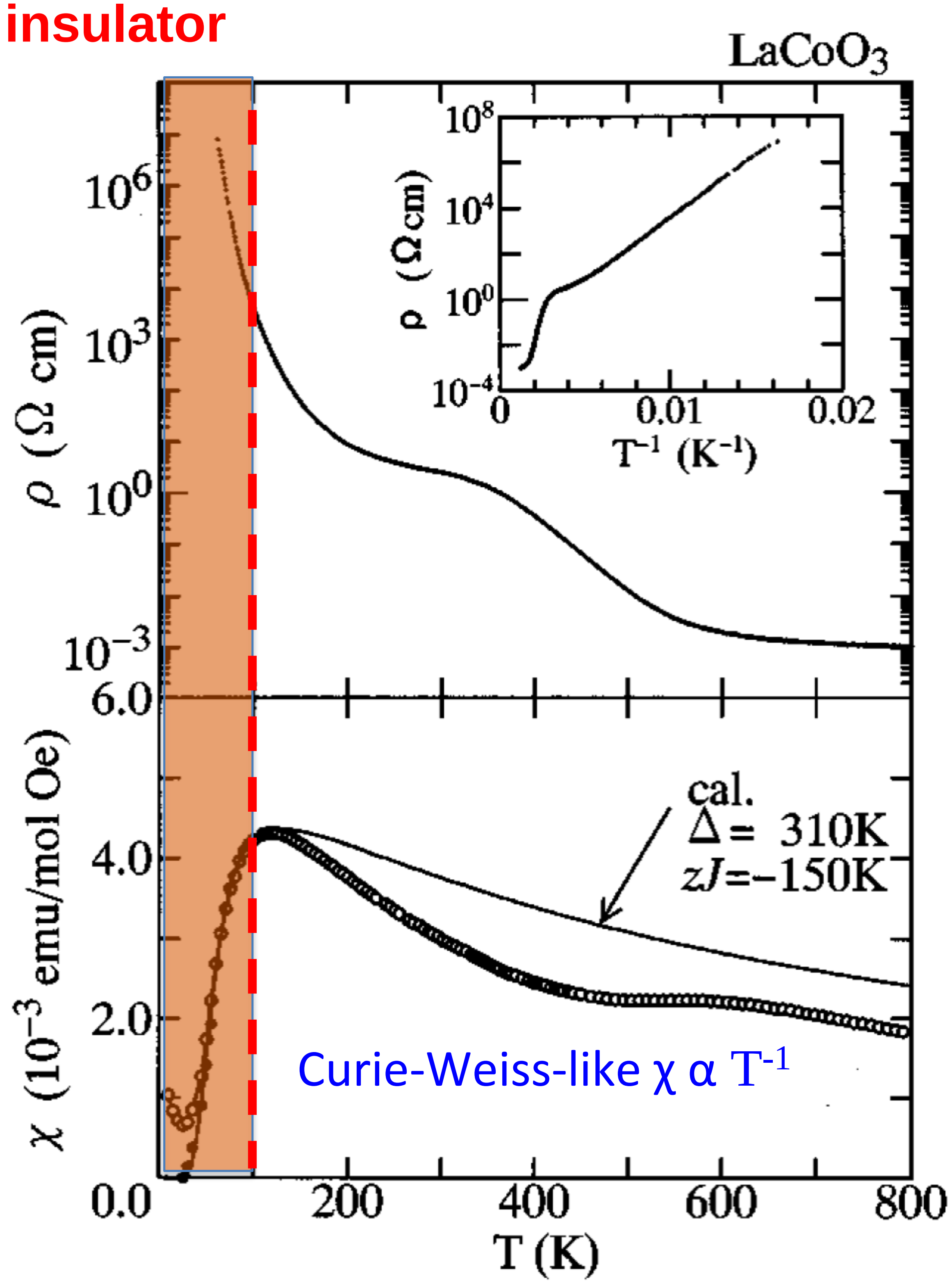
Test setup of AGM-AGS RIXS at the TLS



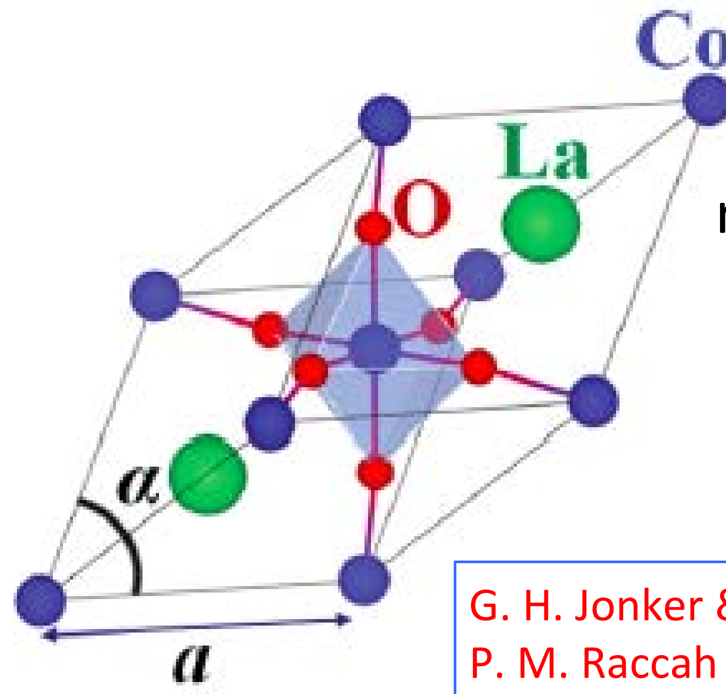
C. H. Lai, J. of SR (2014)

spin-state transition of $LaCoO_3$

diamagnetic insulator



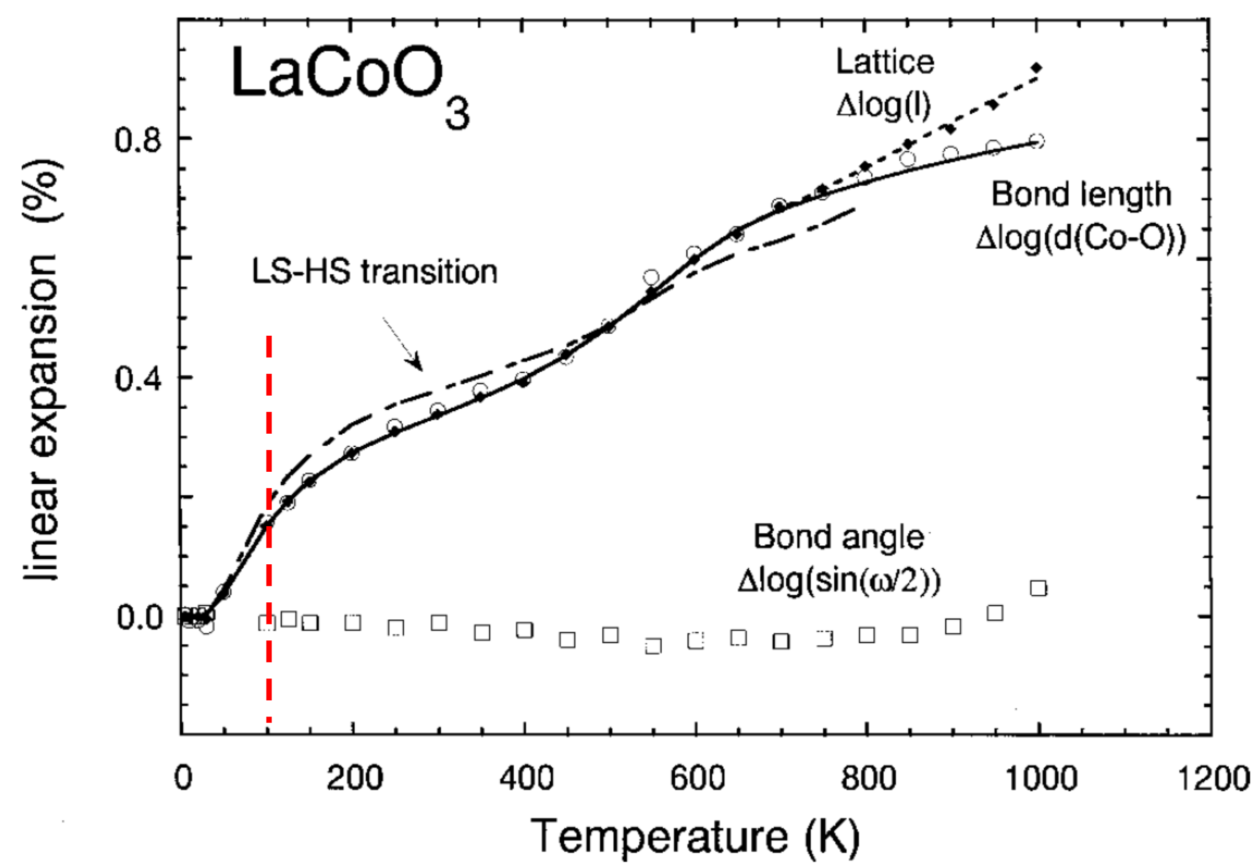
S. Yamaguchi *et al.*, Phys. Rev. B **53**, R2926 (1996).



rhombohedral unit cell

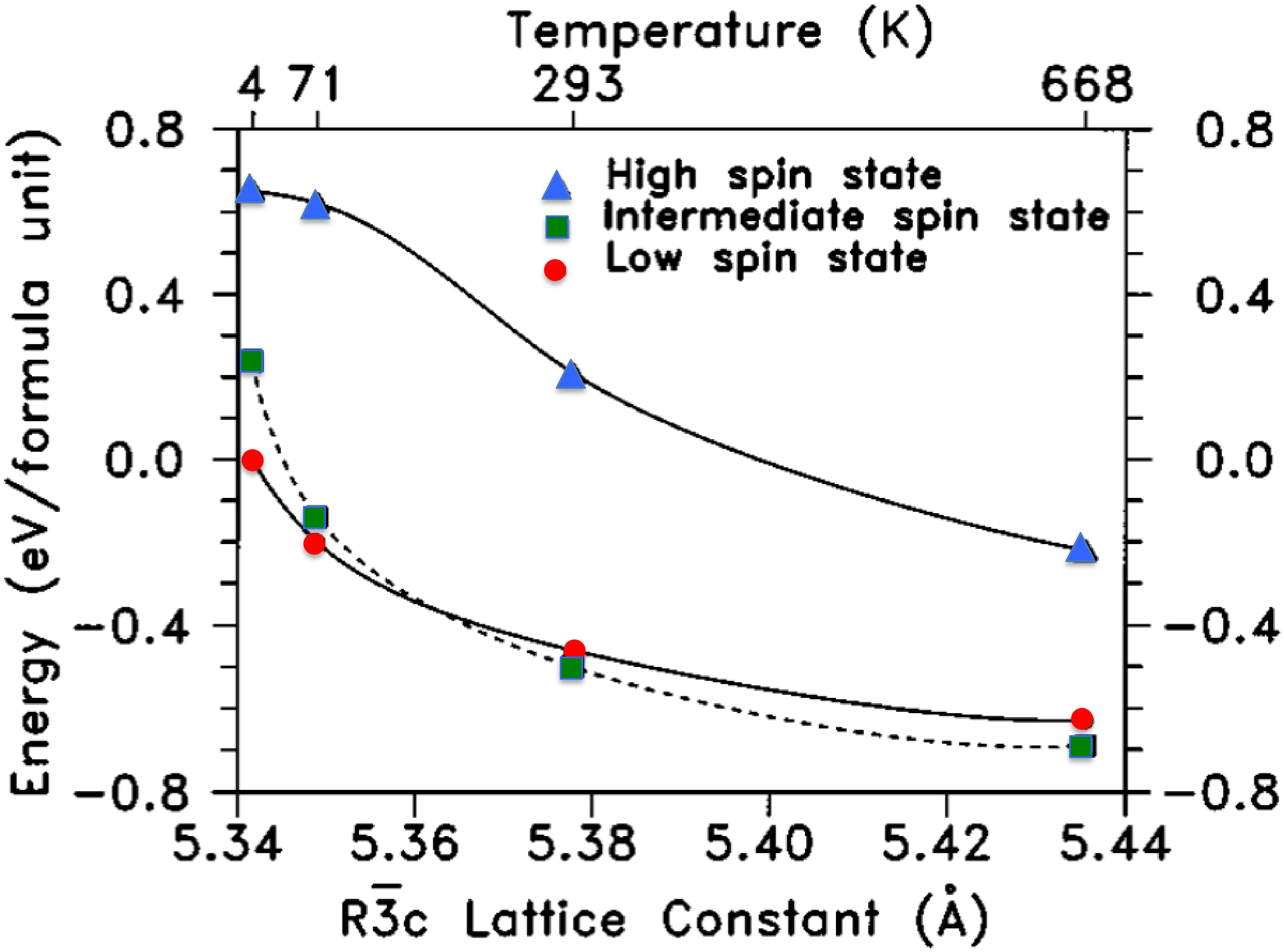
Co^{3+}, d^6

G. H. Jonker & J. H. van Santen (1953)
P. M. Raccah & J. B. Goodenough (1967)



P. G. Radaelli and S.-W. Cheong, Phys. Rev. B **66**, 094408 (2002).

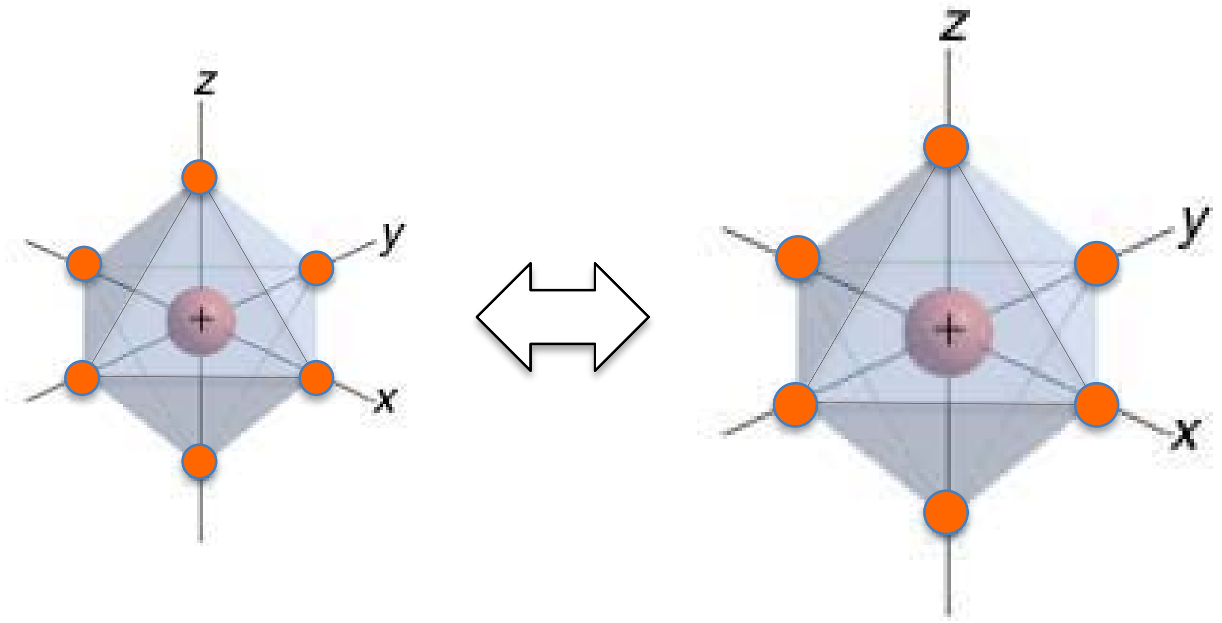
LDA + U calculations:
Energy of IS state is lower than that of LS state above transition temperature.



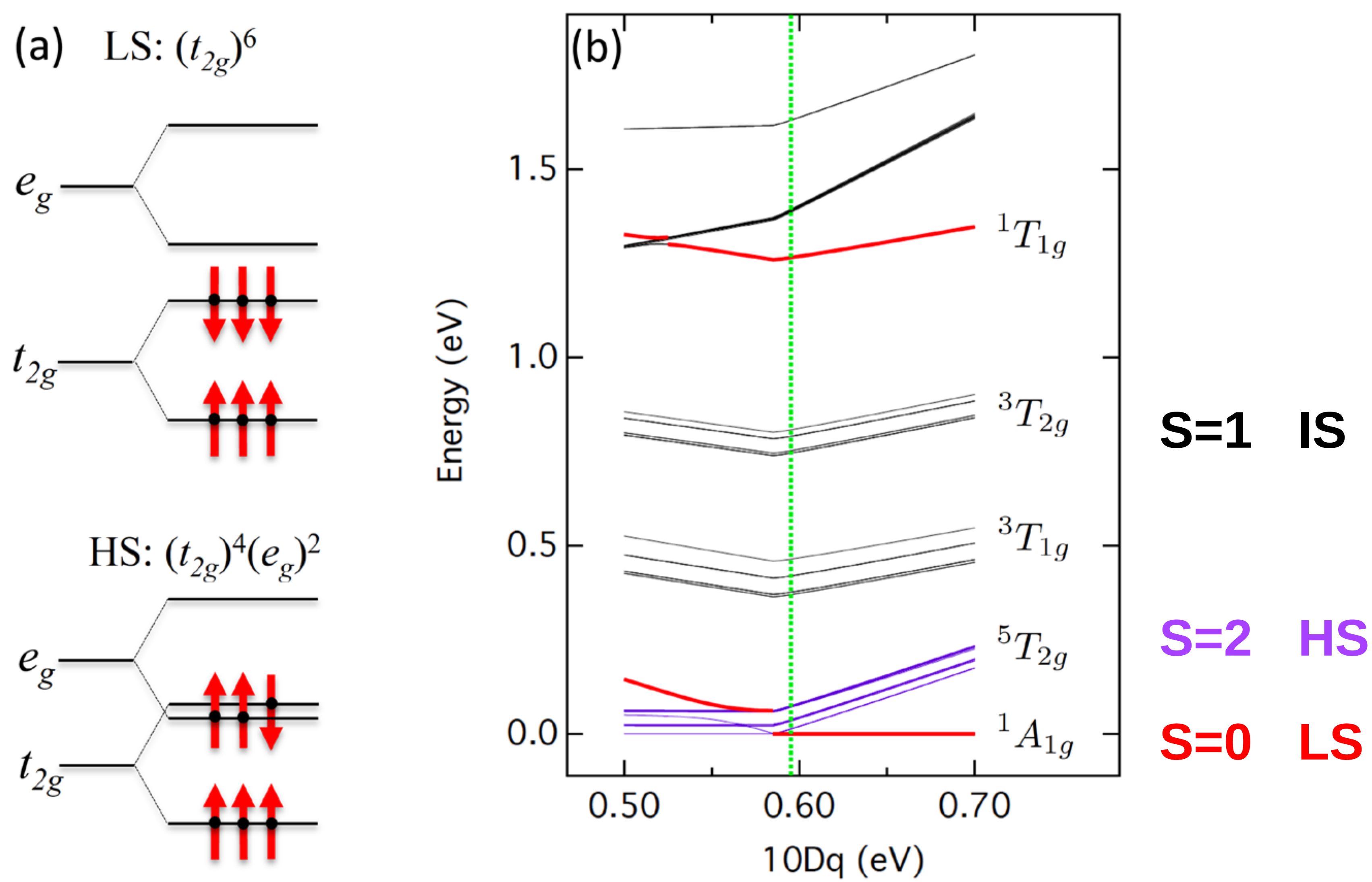
M. A. Korotin *et al.*, Phys. Rev. B **54**, 5309 (1996).

Spin-state transition of a d^6 system

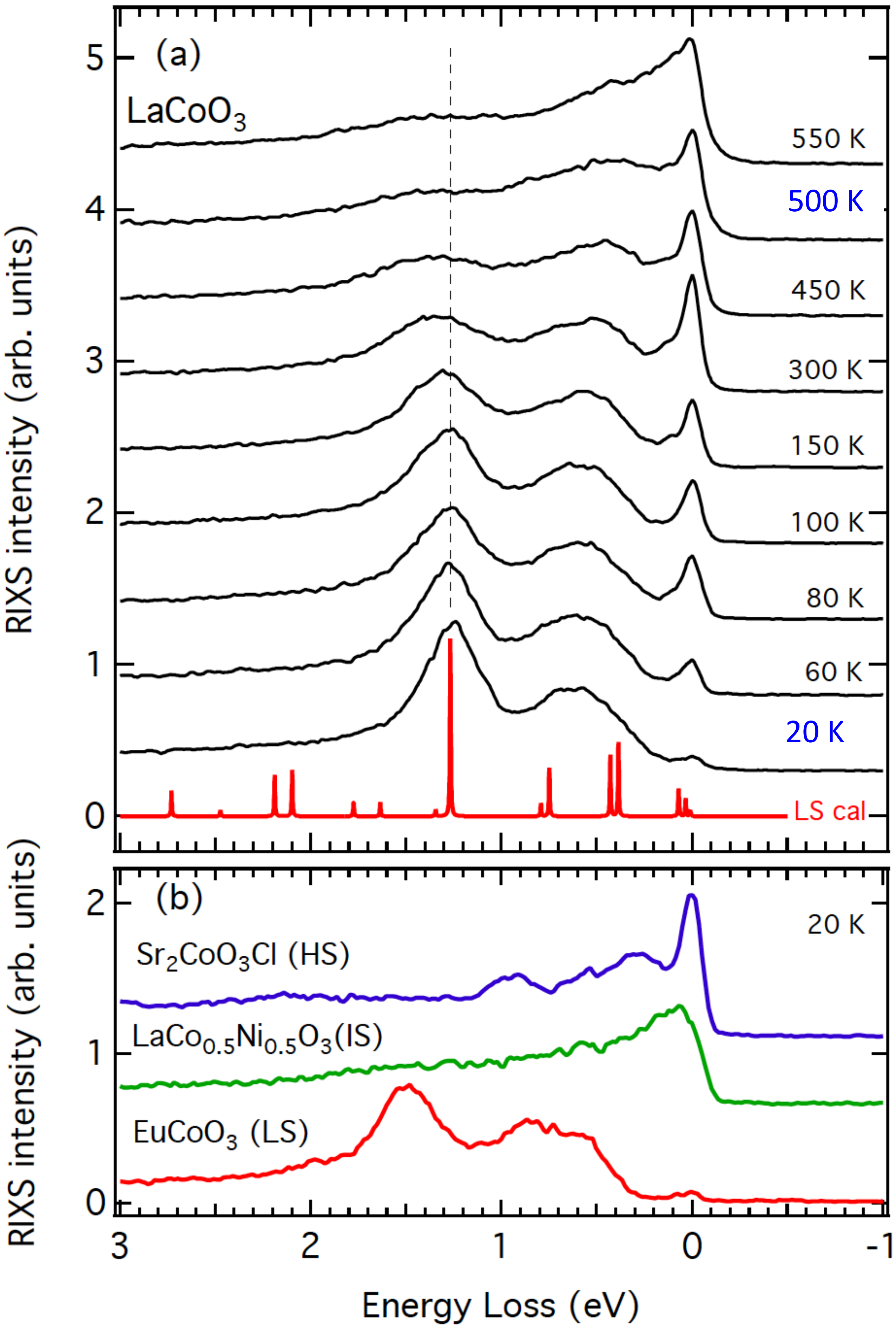
low T
large 10Dq



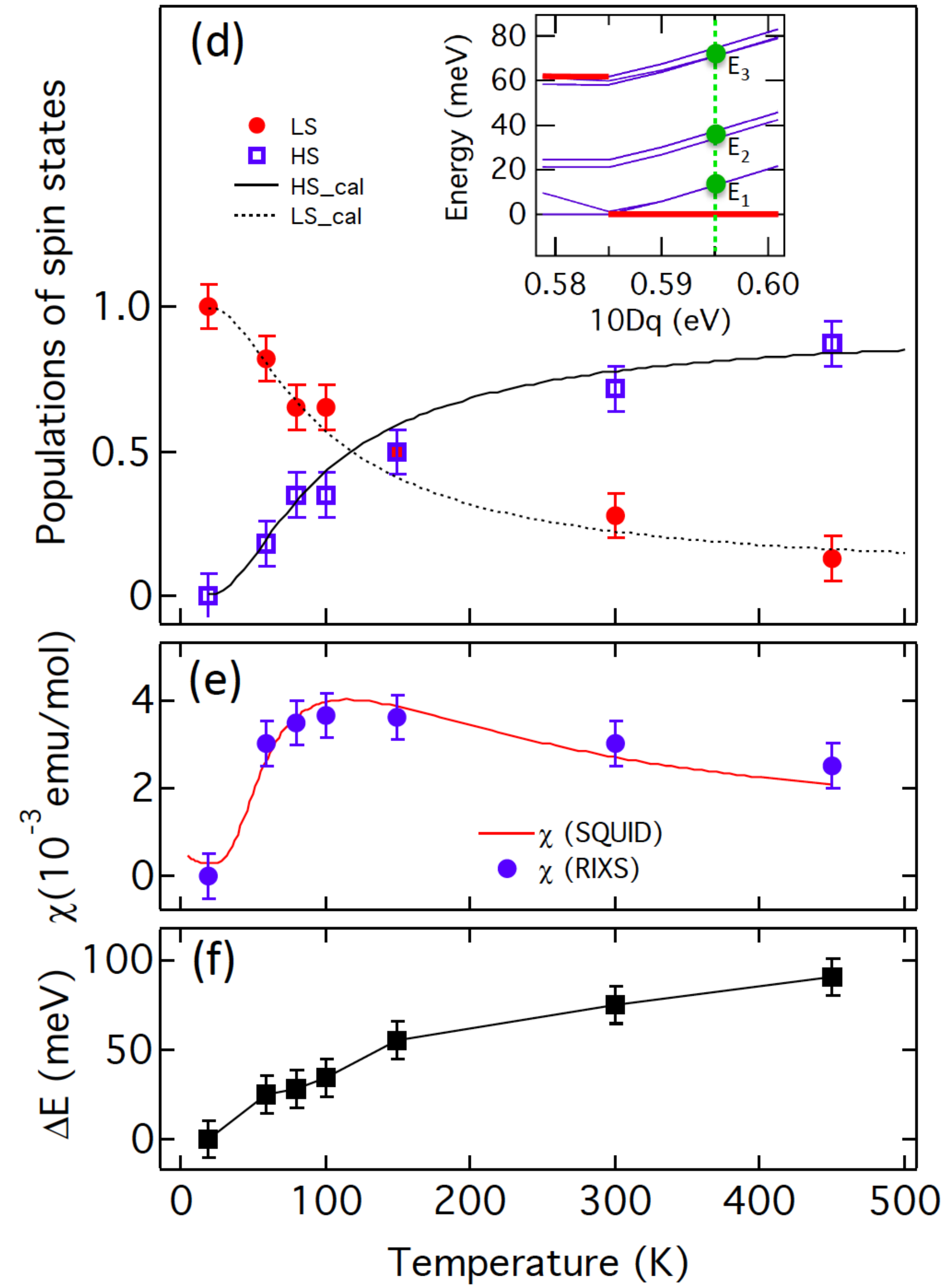
high T
small 10Dq



K. Tomiyasu et al., PRL 119, 196402 (2017)

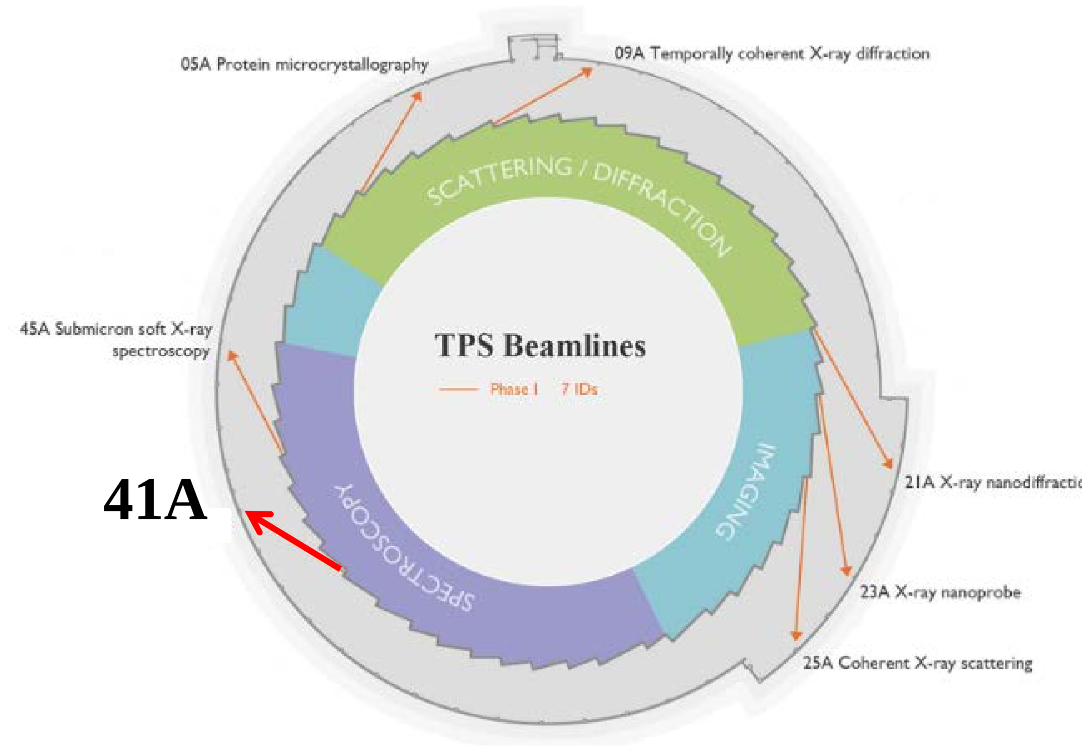


- The ground state is a LS state.
- At 100 K, mixture of LS & HS states
- At 500 K, close to a HS state.



HS population: $\sum_{i=1}^{3} \nu_i e^{-E_i/k_B T}$

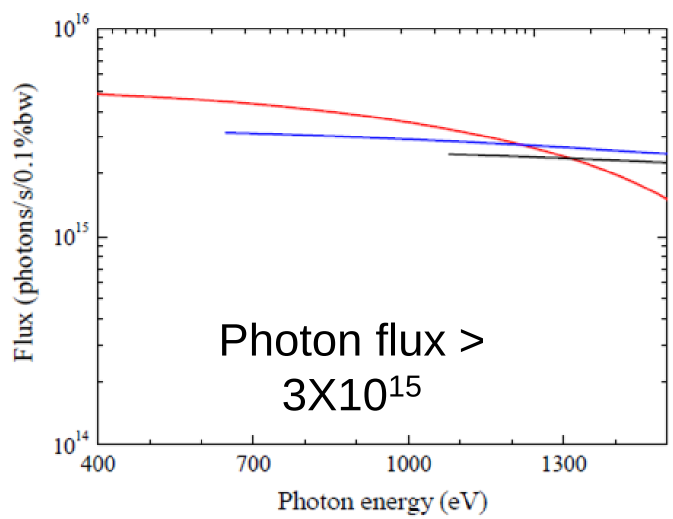
TPS 41A Soft X-ray Scattering

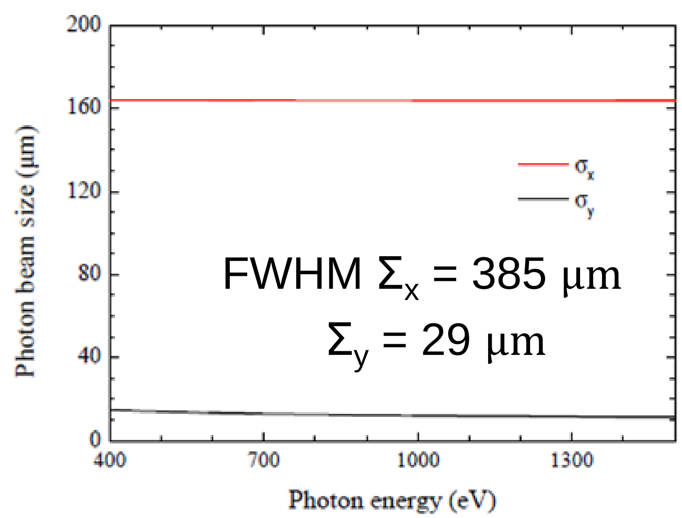


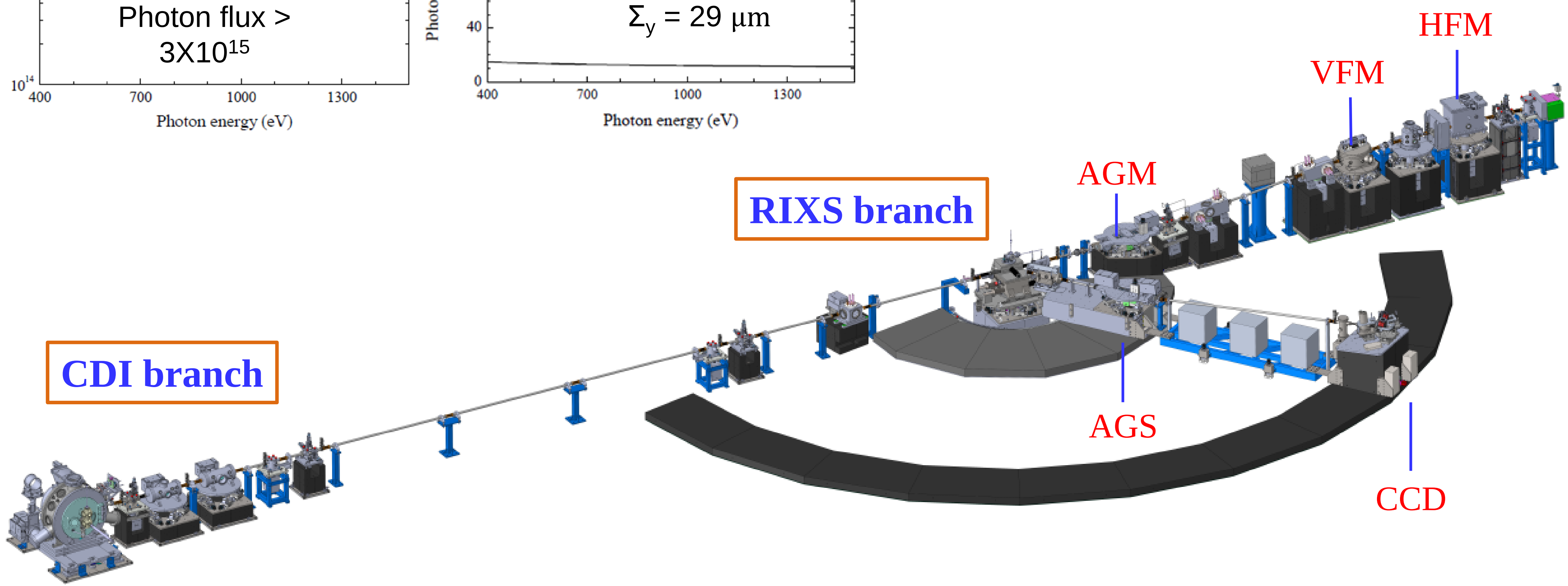
Photon energy: 450 eV – 1200 eV

Photon flux @900 eV: 5.6×10^{12} Photons · s^{-1} · $(0.01\%\ BW)^{-1}$

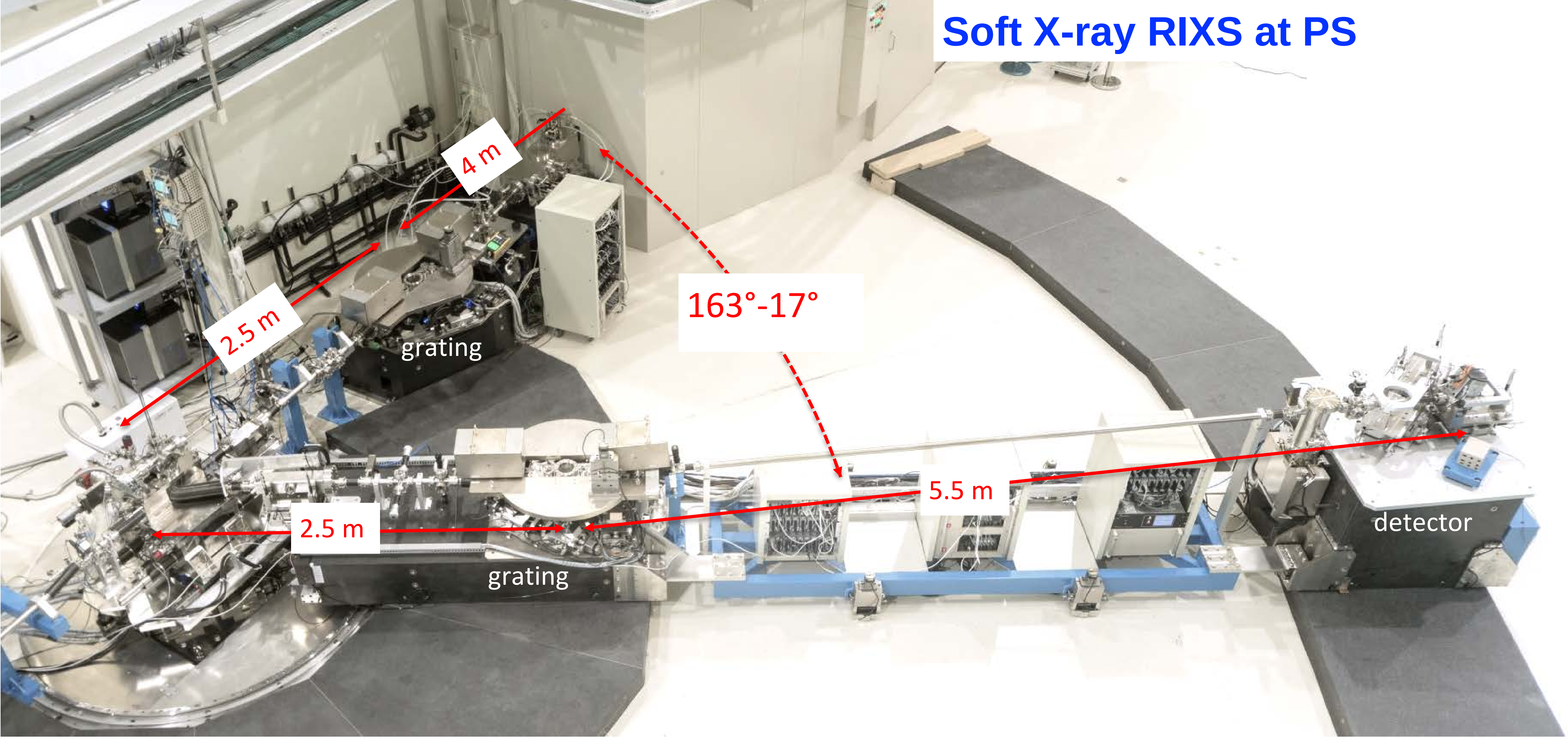
Tandem EPUs (3.5 m + 3.5 m)







Soft X-ray RIXS at PS



Momentum-resolved RIXS

$$17^\circ < 2\theta < 163^\circ$$

Active Grating

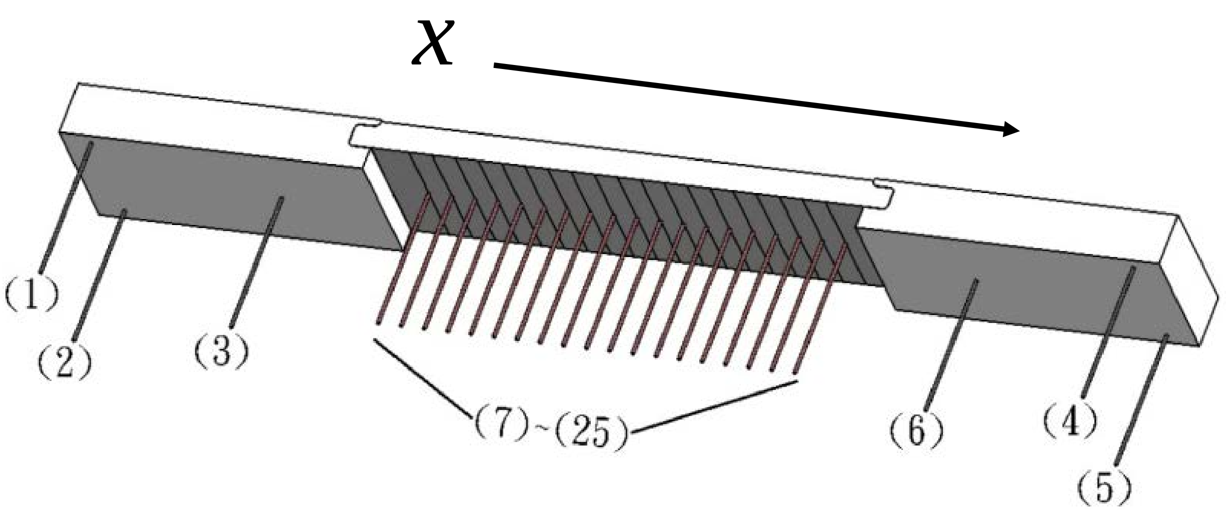
- **"active" (bendable) grating**
- **Varied-line-spacing (VLS) grating**

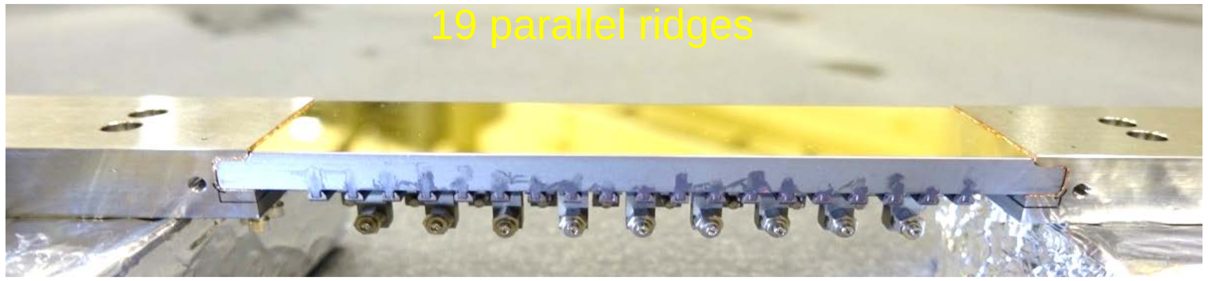
$$n(x) = n_0 + n_1 x + n_2 x^2$$

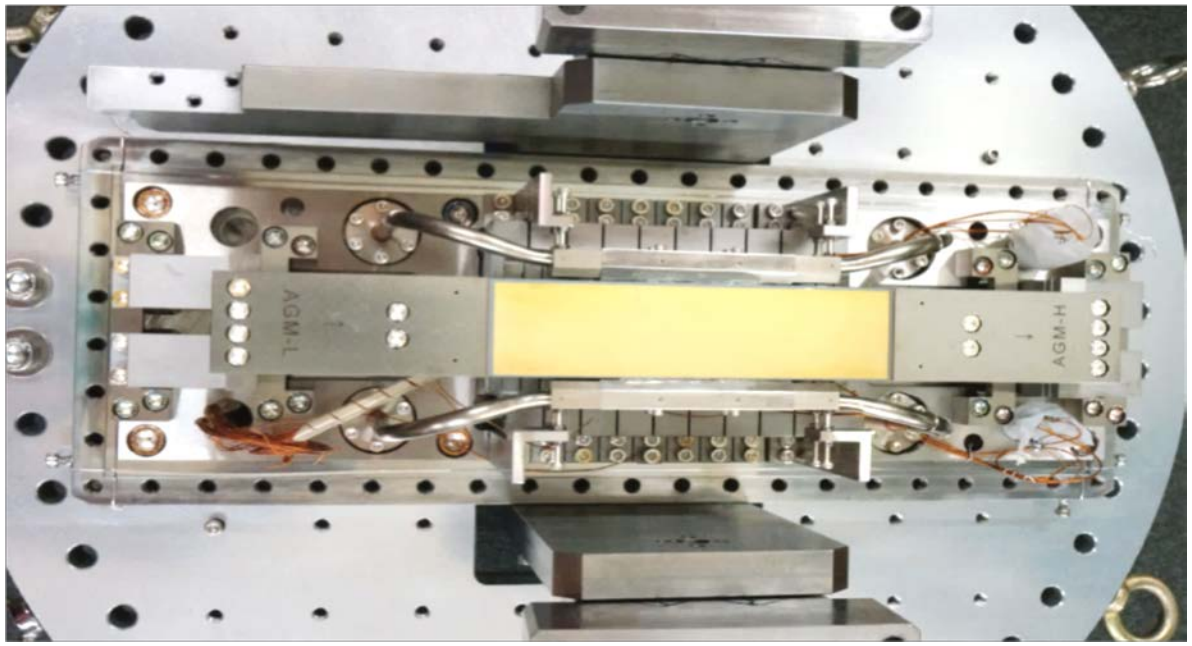
$$n_0 = 1200\ mm^{-1}$$

- **grating surface equation**

$$f_0(x) = c_0 + c_1 x + c_2 x^2 + c_3 x^3$$

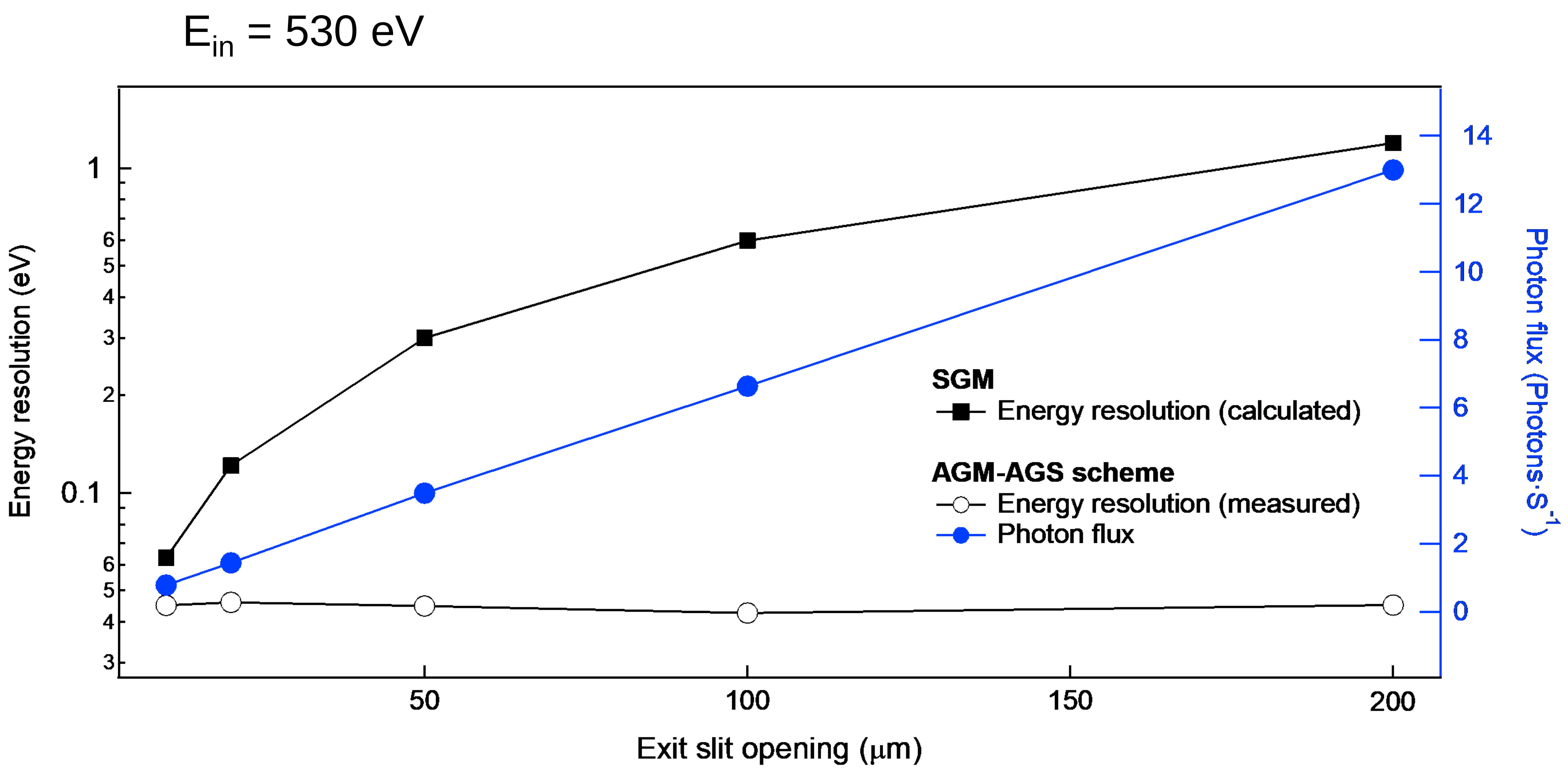




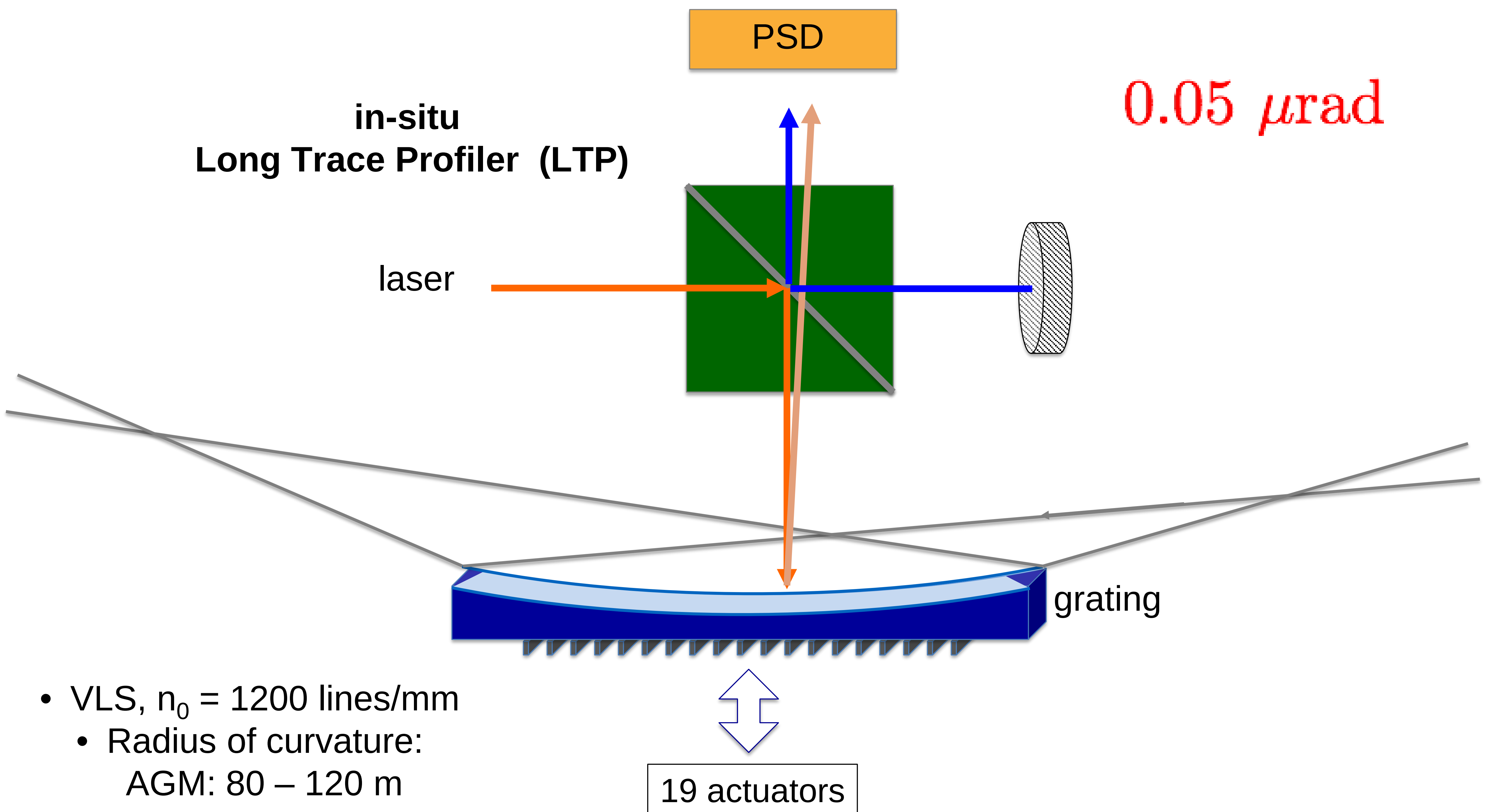


Test of energy compensation principle

The efficiency of RIXS is increased without sacrificing the energy resolution.



"active" grating monochromator/spectrometer + LTP



- VLS, n_0 = 1200 lines/mm
  - Radius of curvature:
    AGM: 80 – 120 m
    AGS: 40 – 50 m

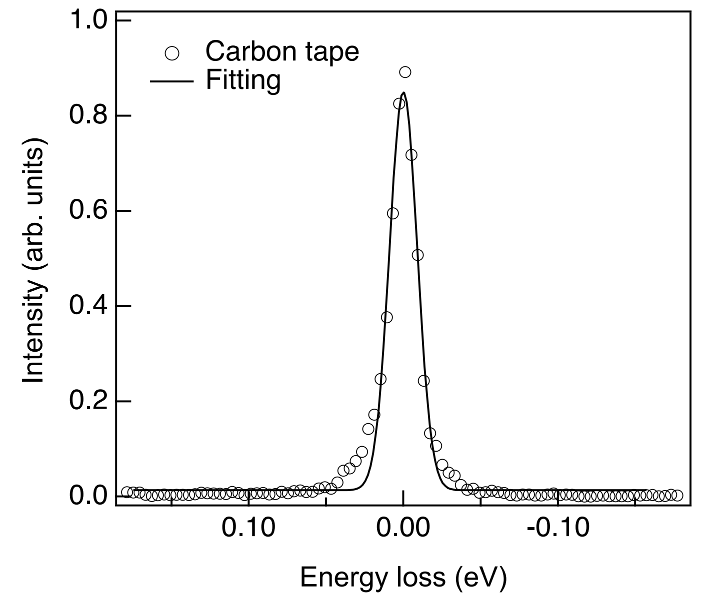
$$f_0(x) = c_0 + c_1 x + c_2 x^2 + c_3 x^3$$

$$f_0'(x) = c_1 + 2c_2 x + 3c_3 x^2$$

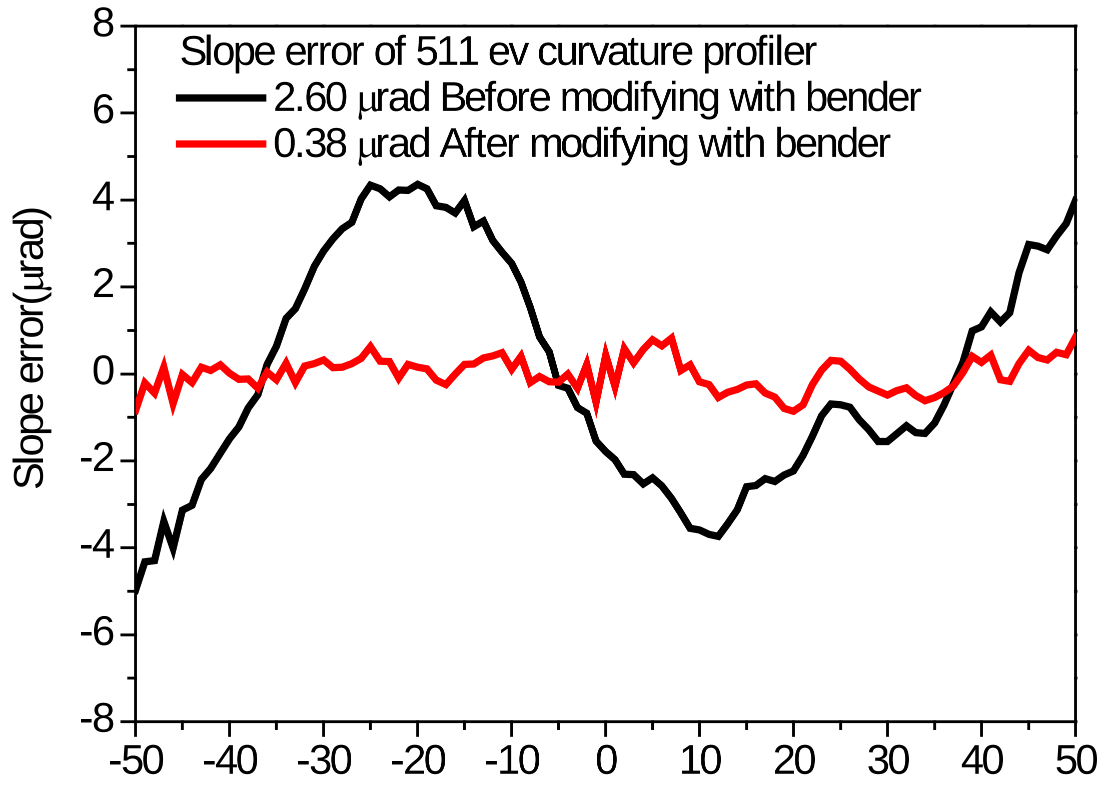
focusing (c_2) and comma (c_3) aberrations

Resolving power: 24000

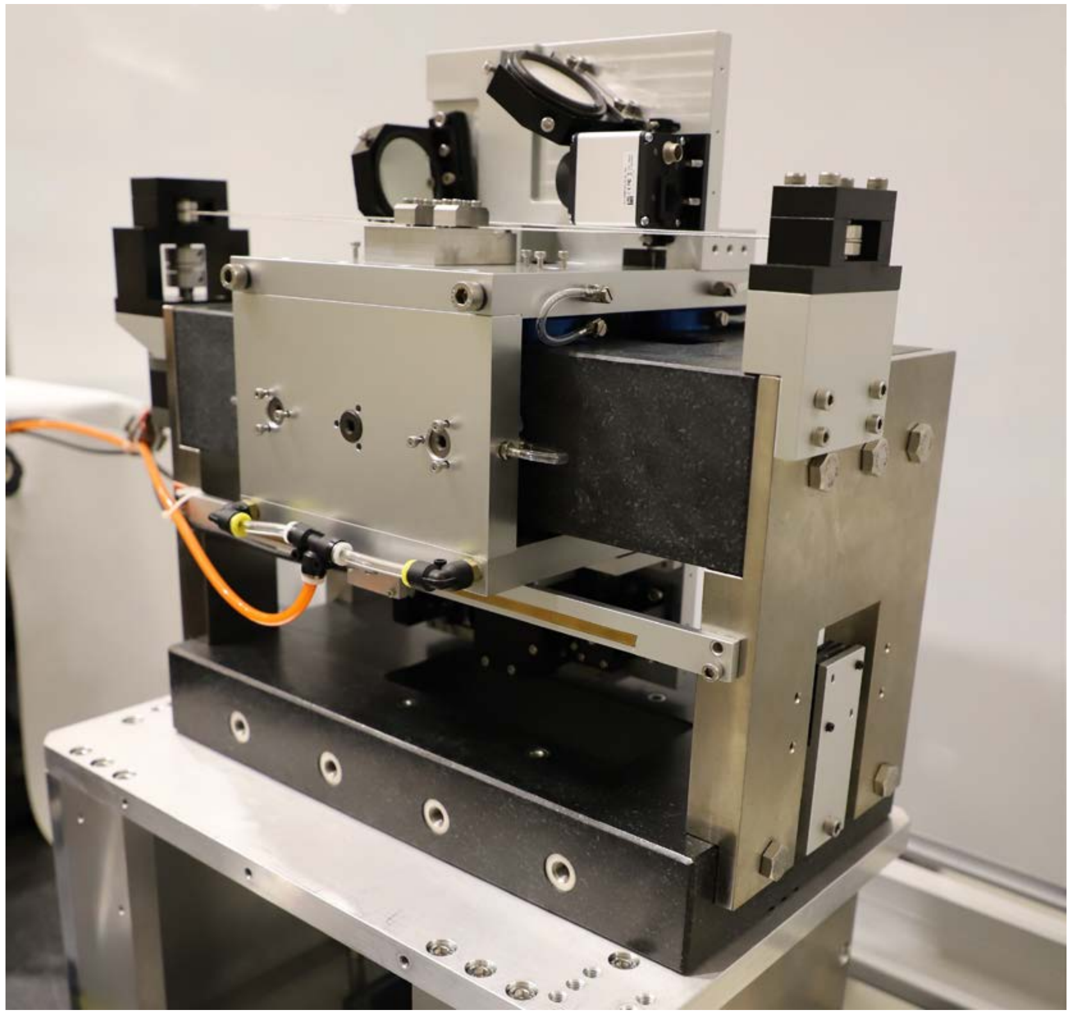
Energy resolution at 530 eV: 22 meV



$$\text{slope diff} = \text{LTP} - f_0'(x)$$



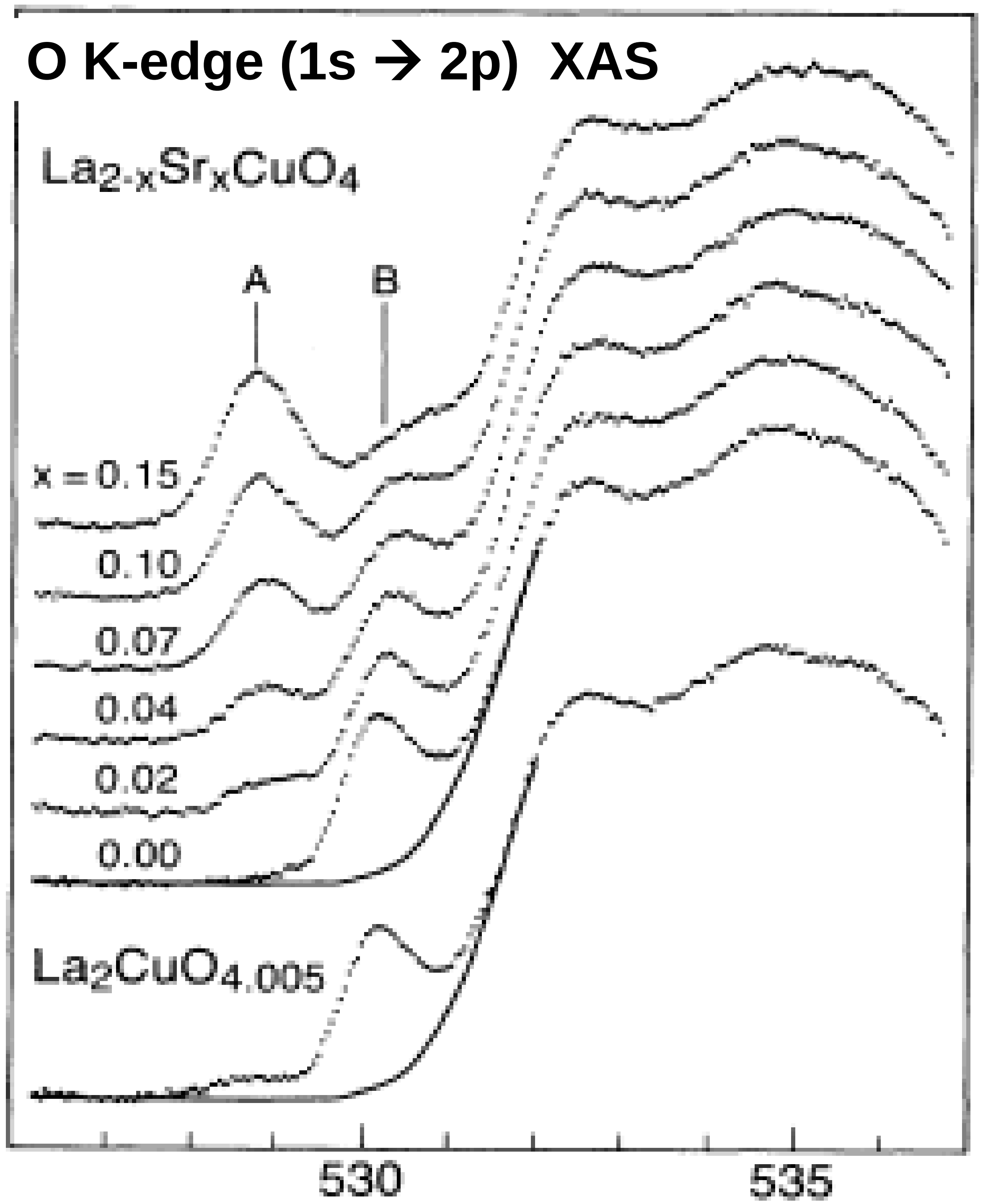
x (*mm*)



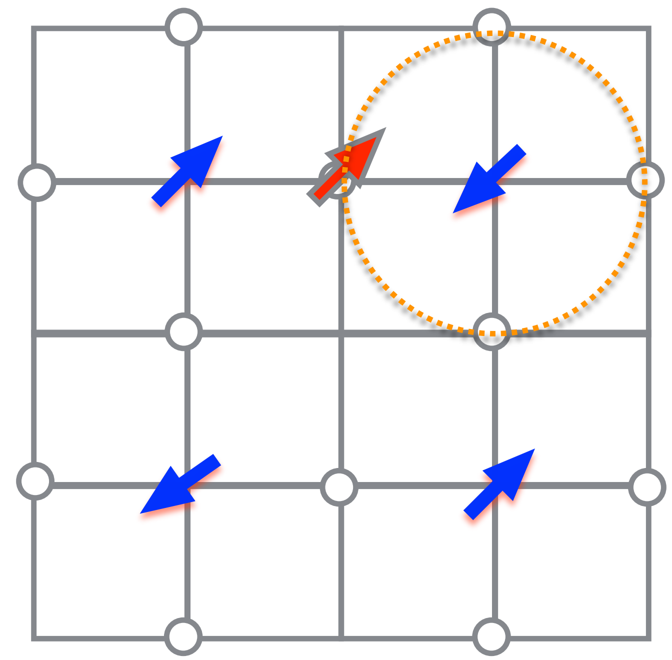
VOLUME 66, NUMBER 1 PHYSICAL REVIEW LETTERS 7 JANUARY 1991

Electronic States in $La_{2-x}Sr_xCuO_{4+\delta}$ Probed by Soft-X-Ray Absorption

C. T. Chen,[1] F. Sette,[1] Y. Ma,[1] M. S. Hybertsen,[1] E. B. Stechel,[2] W. M. C. Foulkes,[1] M. Schluter,[1] S-W. Cheong,[1] A. S. Cooper,[1] L. W. Rupp, Jr.,[1] B. Batlogg,[1] Y. L. Soo,[3] Z. H. Ming,[3] A. Krol,[3] and Y. H. Kao[3]



Zhang-Rice Singlet (ZRS)

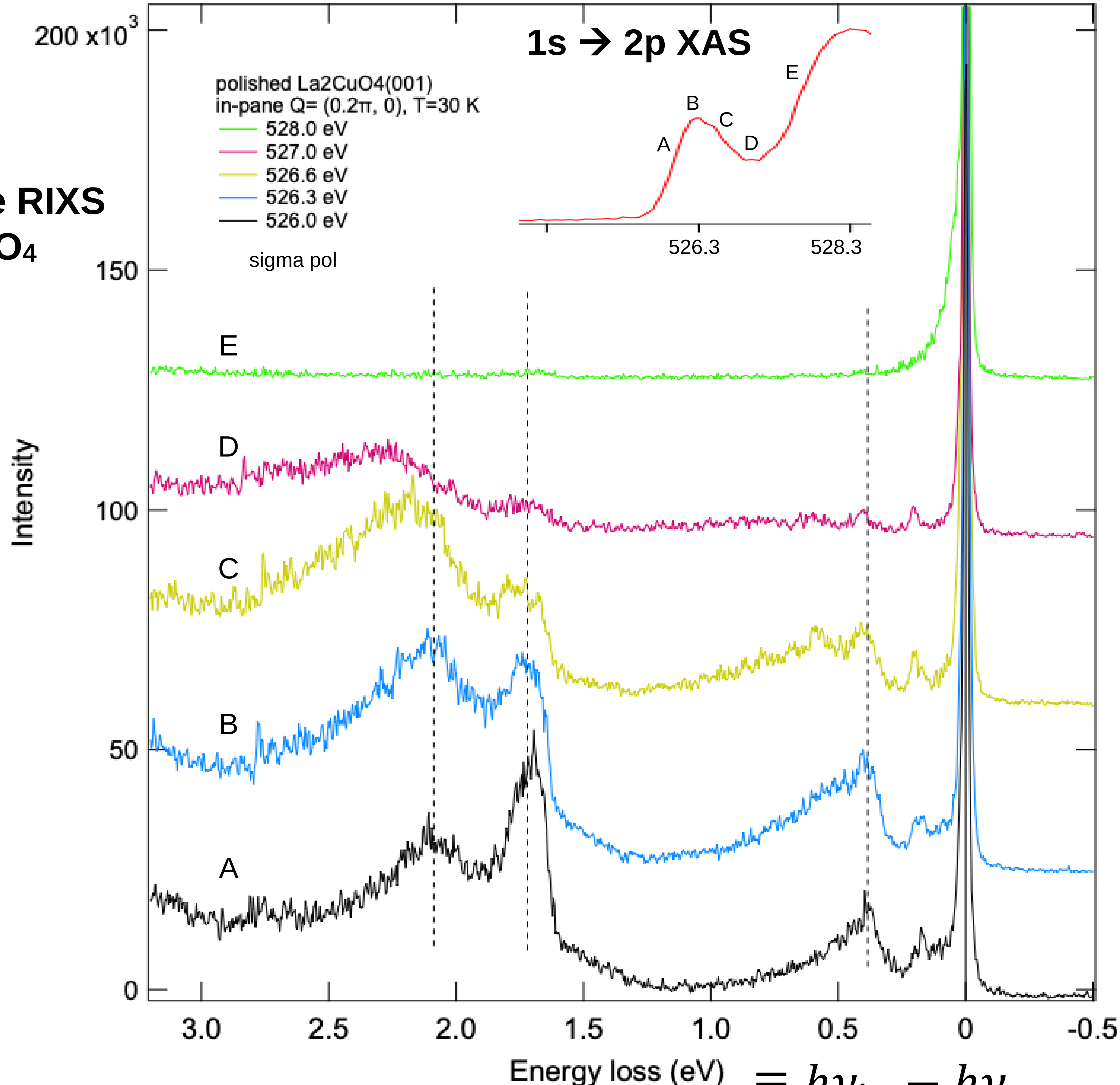


ground state of an undoped cuprate

: spin of **Cu 3*d* holes**

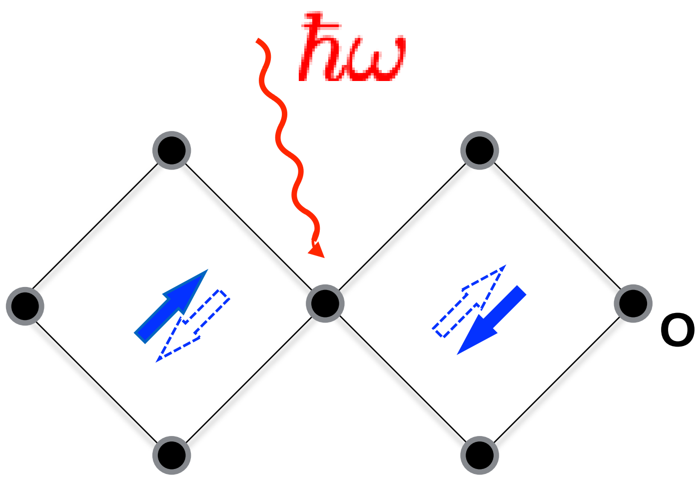
: spin of **O 2*p* holes**

O K-edge RIXS of La_2CuO_4



$\equiv h\nu_{\text{in}} - h\nu_{\text{out}}$

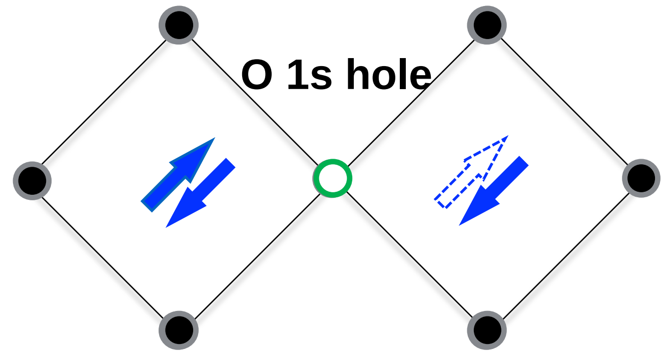
O K-edge RIXS of La_2CuO_4

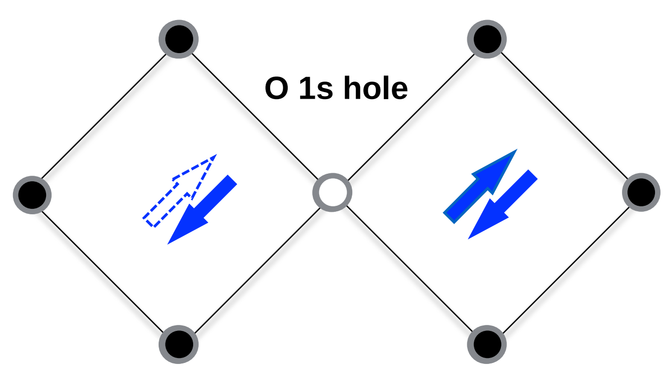


Cu^{2+} $3d_{x^2-y^2}$

ground state

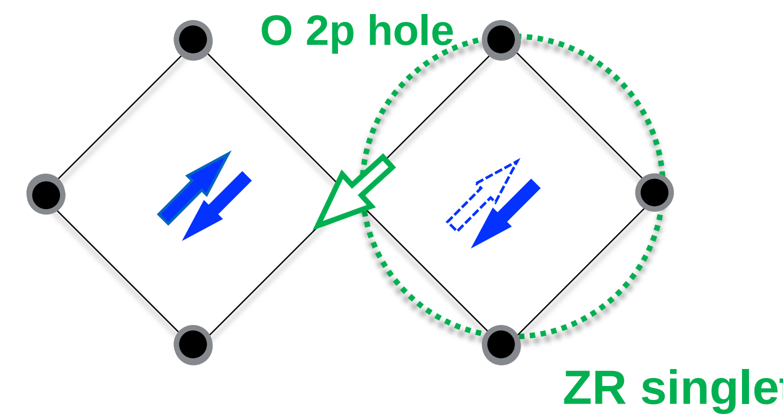
O 1s → O 2p / Cu 3d



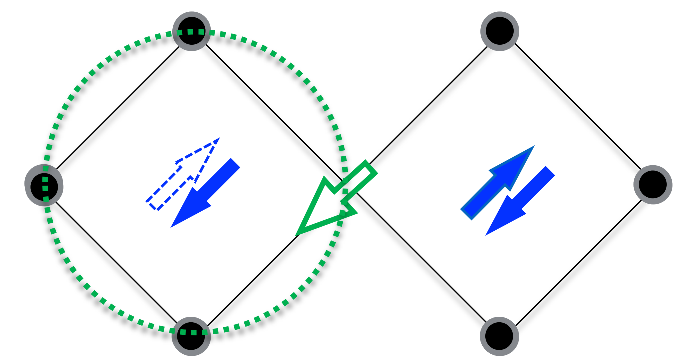


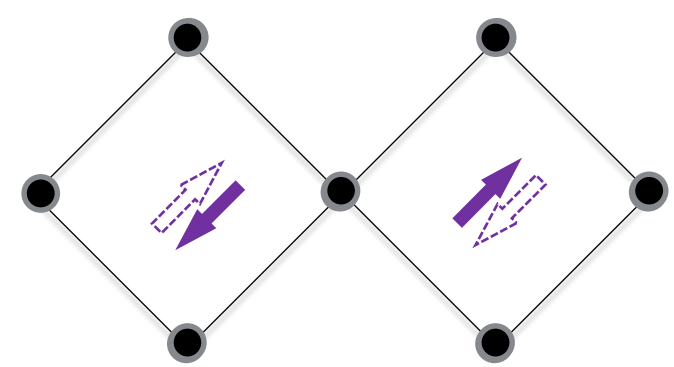
intermediate state

O 2p → O 1s



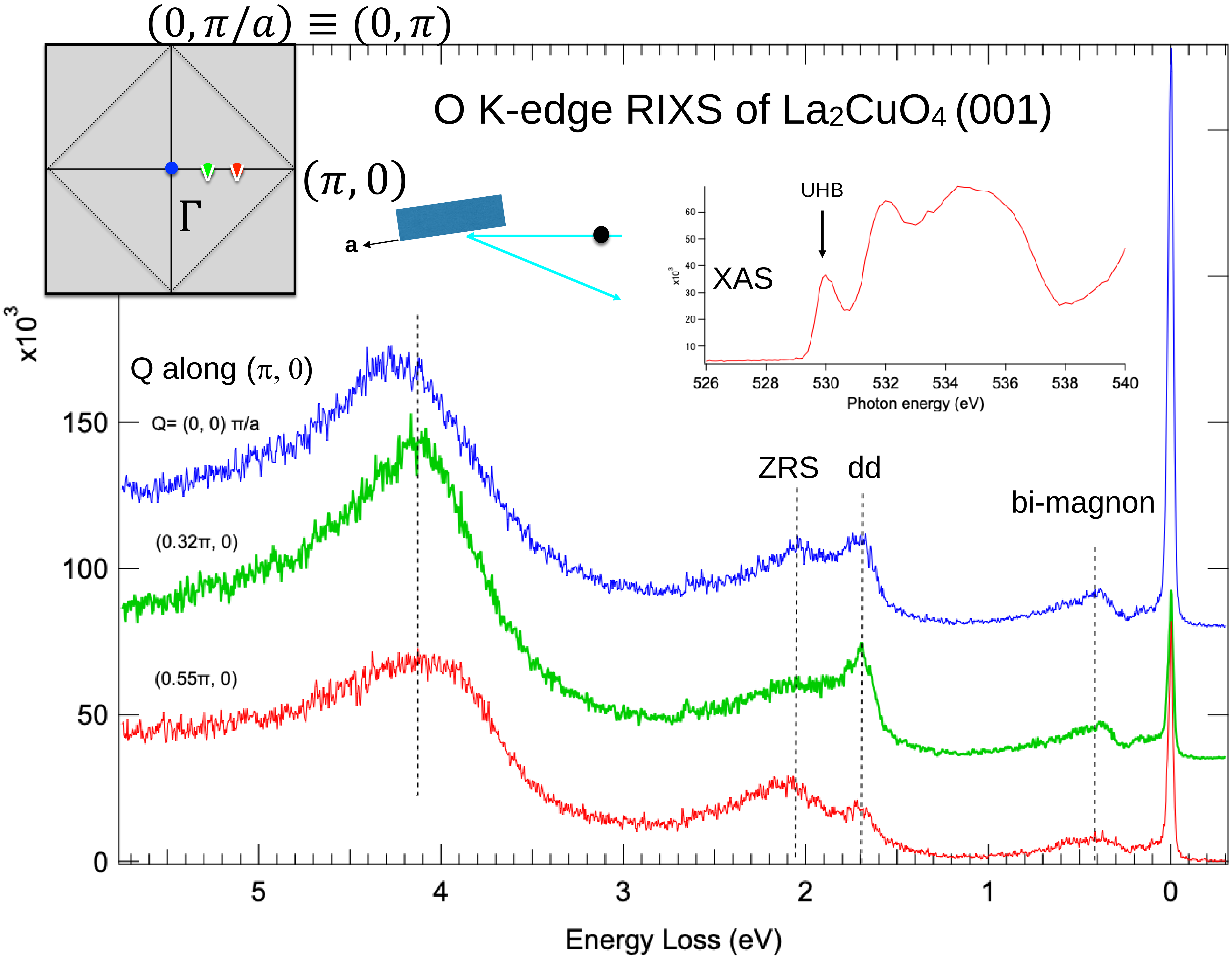
charge-transfer exciton





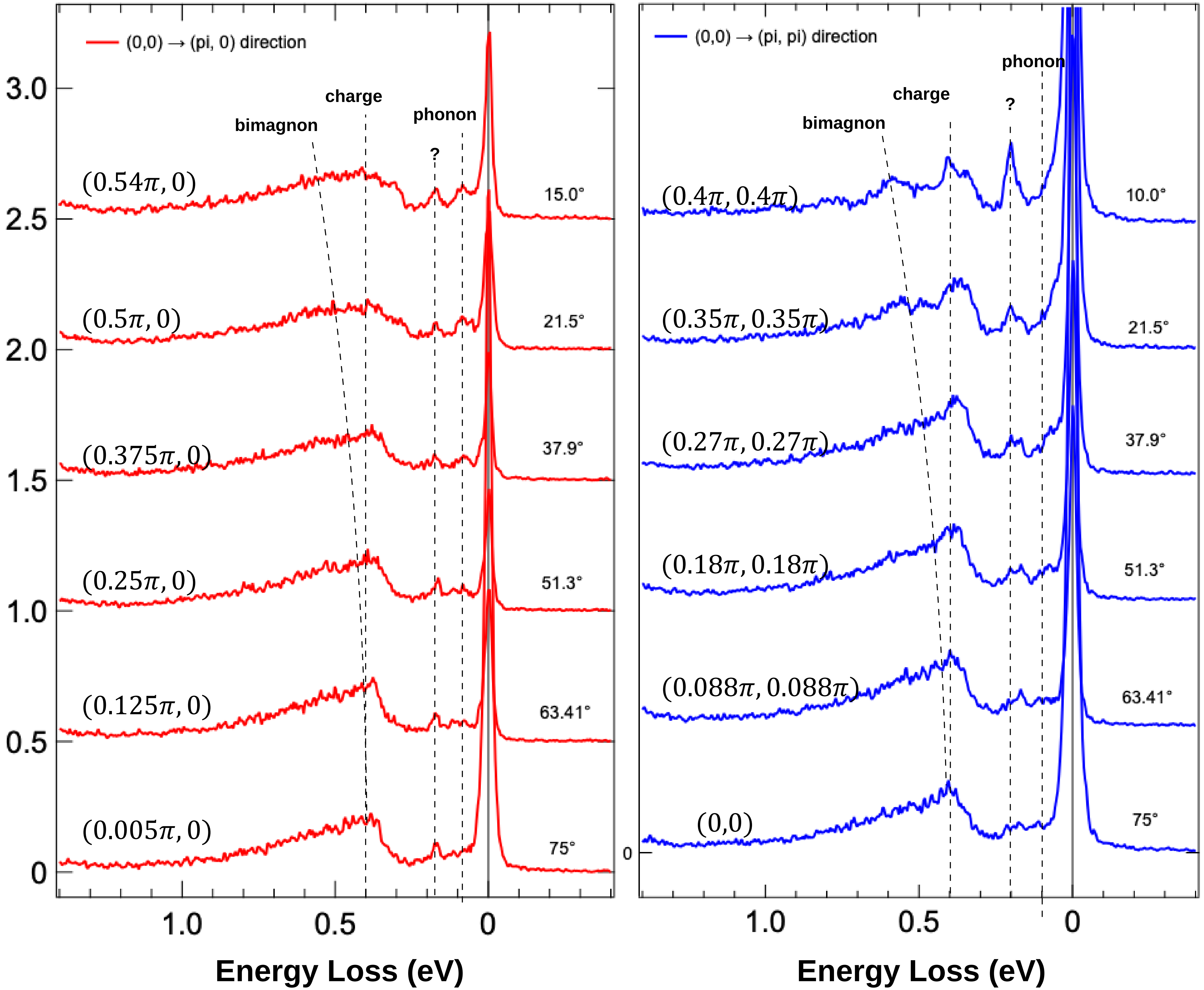
bi-magnon

final state

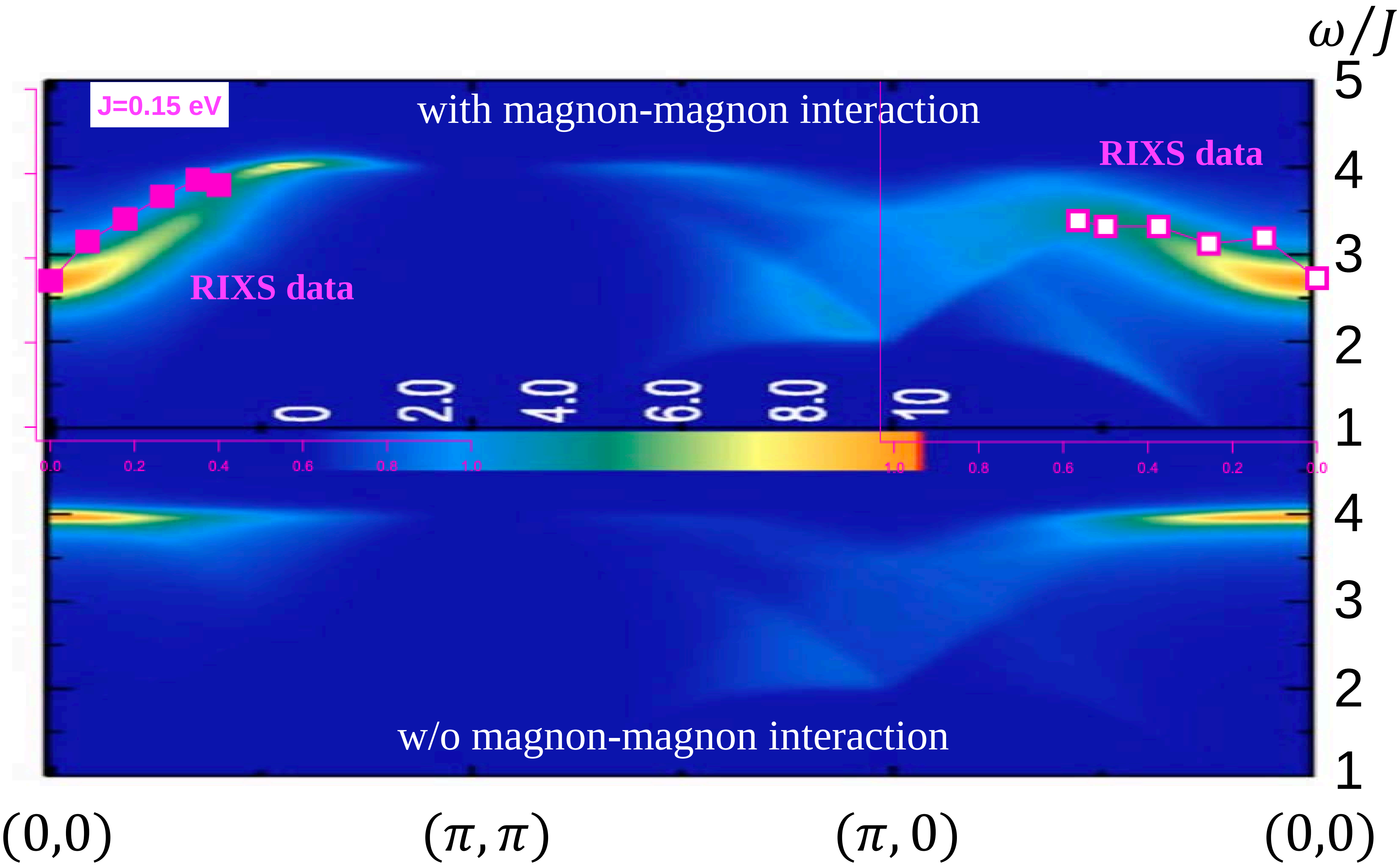


$(0,\pi/a) \equiv (0,\pi)$
$(\pi,0)$
Γ
a
O K-edge RIXS of La_2CuO_4 (001)
UHB
XAS
x10^3
60
50
40
30
20
10
526
528
530
532
534
536
538
540
Photon energy (eV)
x10^3
Q along (π, 0)
Q= (0, 0) π/a
(0.32π, 0)
(0.55π, 0)
ZRS
dd
bi-magnon
150
100
50
0
5
4
3
2
1
0
Energy Loss (eV)

O K-edge RIXS of La_2CuO_4



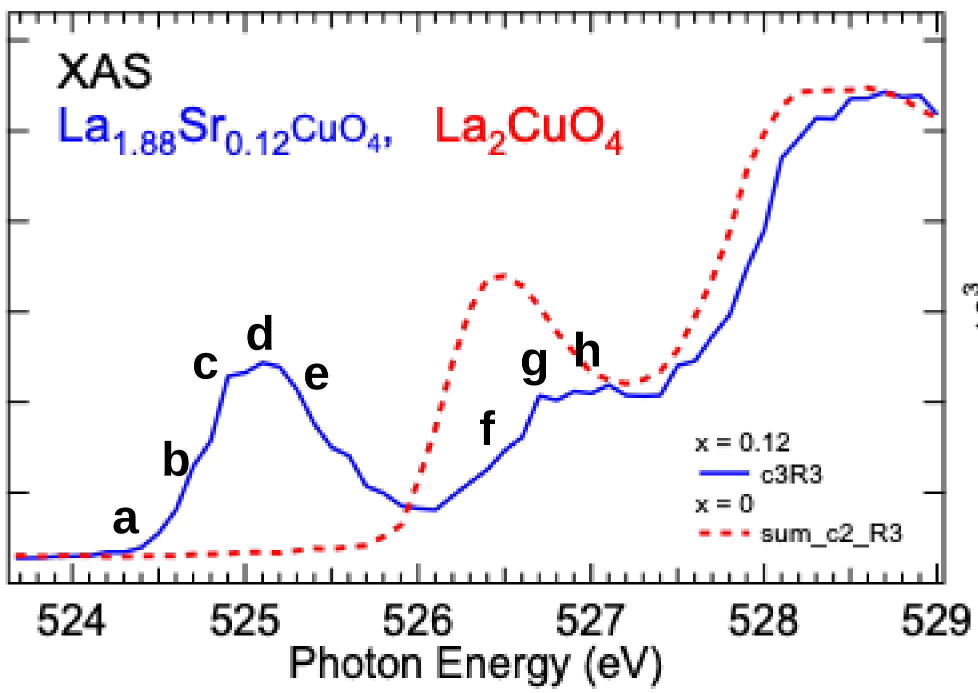
Bi-magnon dispersioon

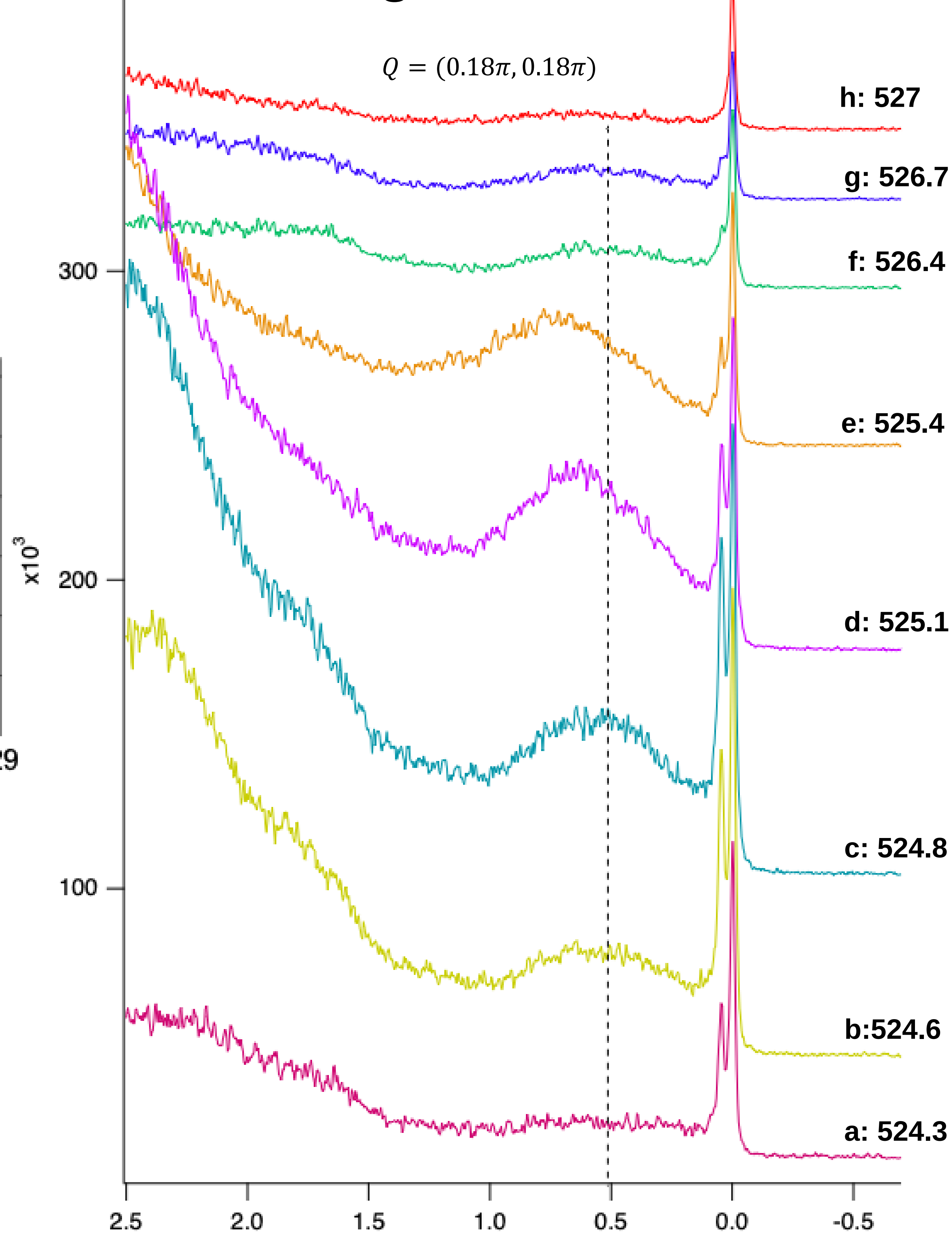


Vernay et al., PRB **75**, 020403(R) (2007)

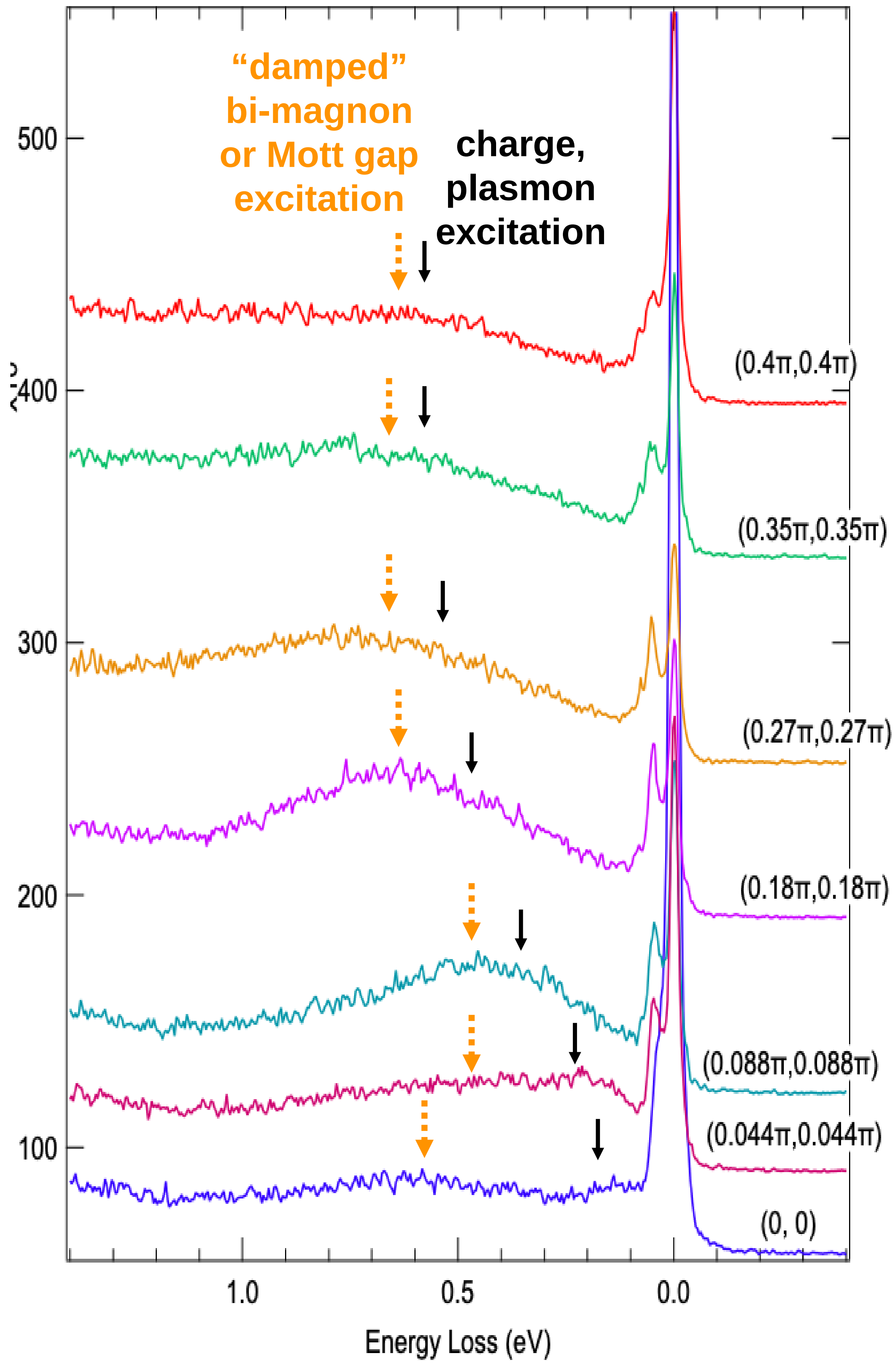
O K-edge RIXS of

$La_{1.88}Sr_{0.12}CuO_4$

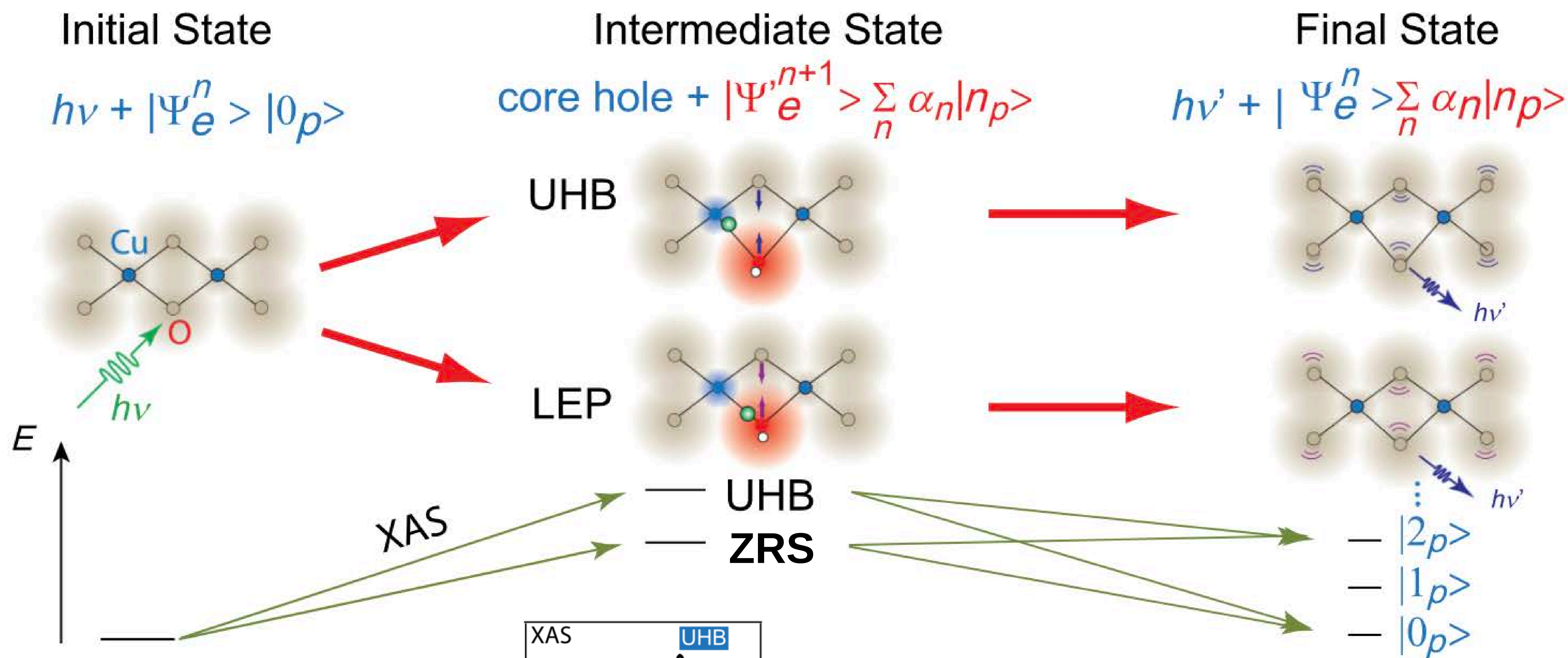




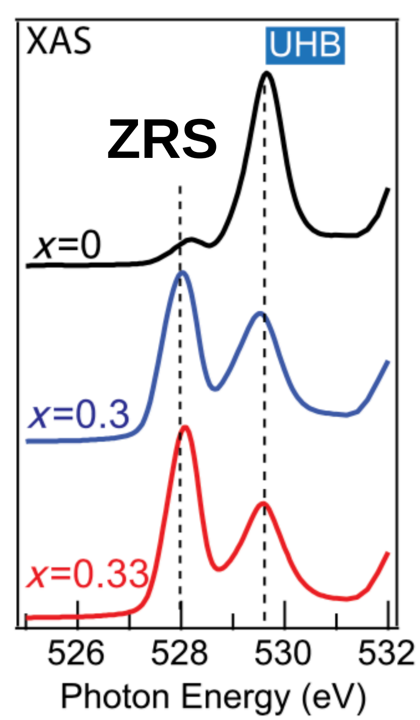
Q-dependent O K-edge RIXS of $La_{1.88}Sr_{0.12}CuO_4$



momentum resolved e-ph coupling



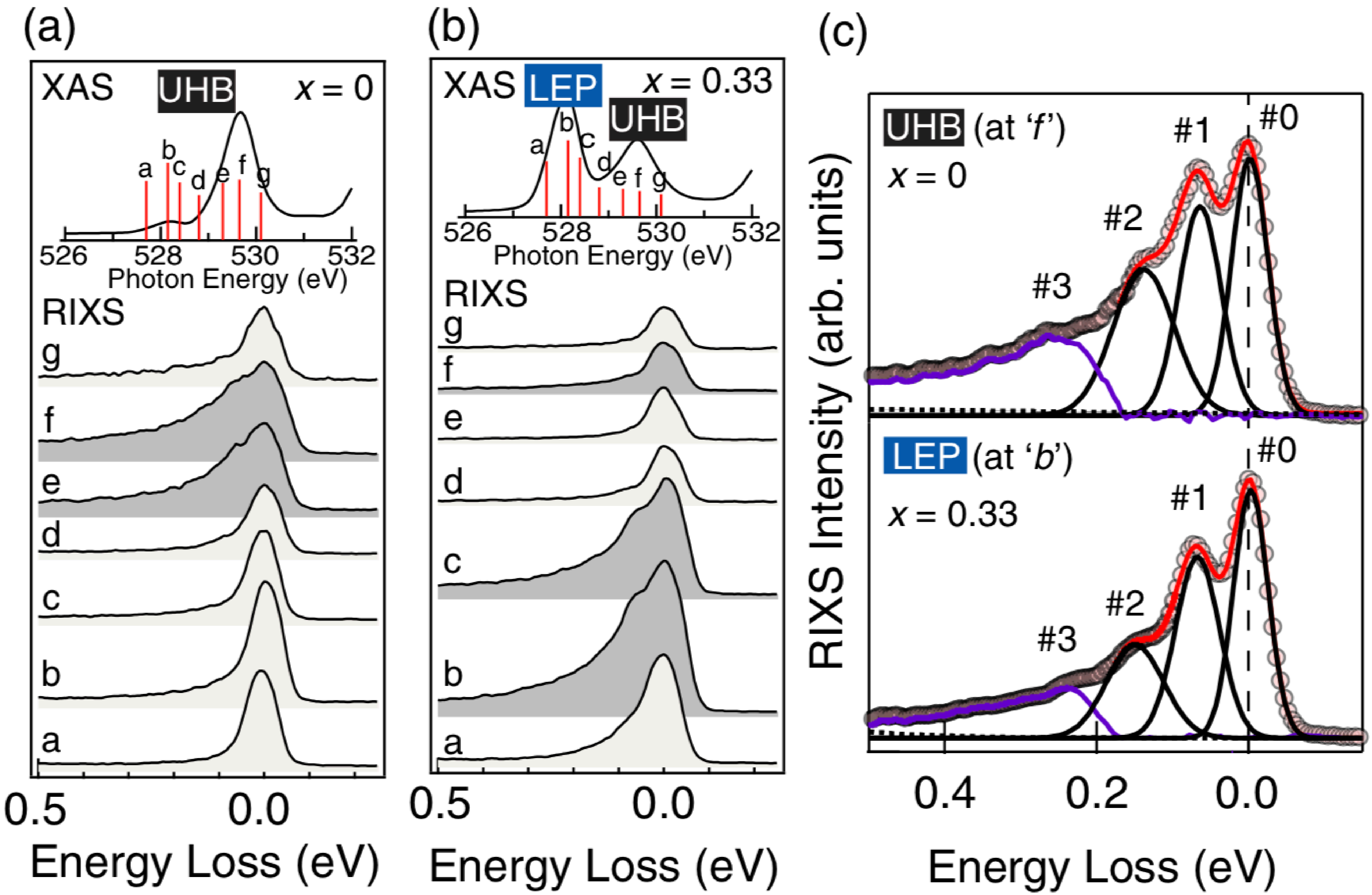
WS Lee et al., PRL (2013)



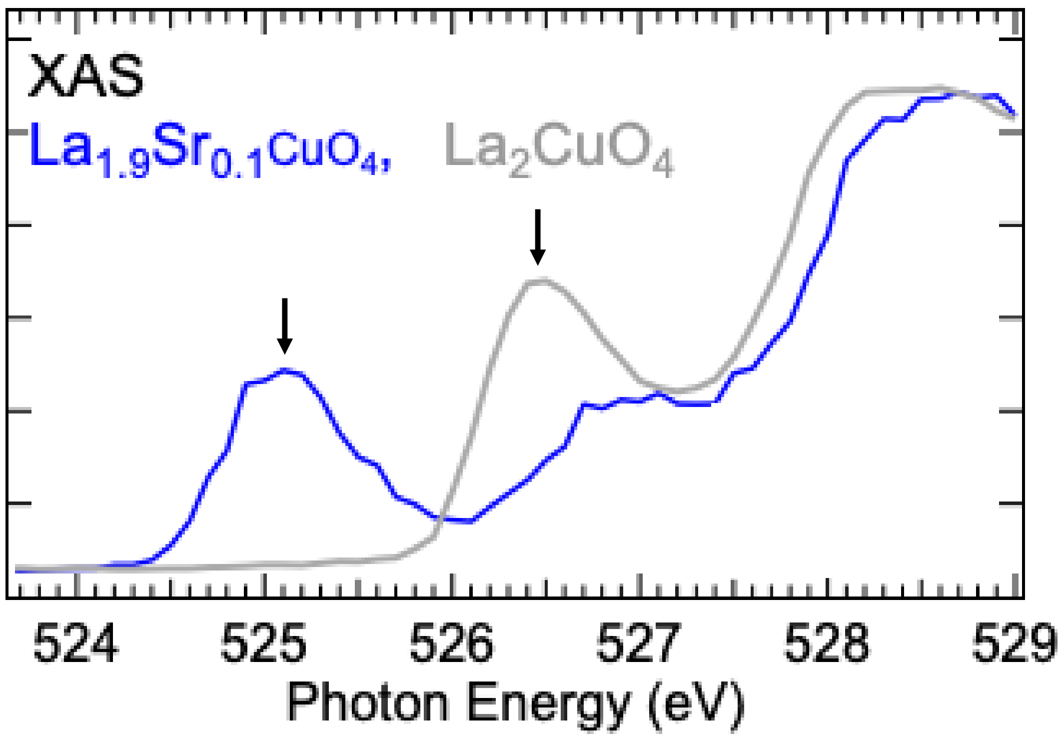
Phonons modulate the Cu-O bonds and the ZRS energy.

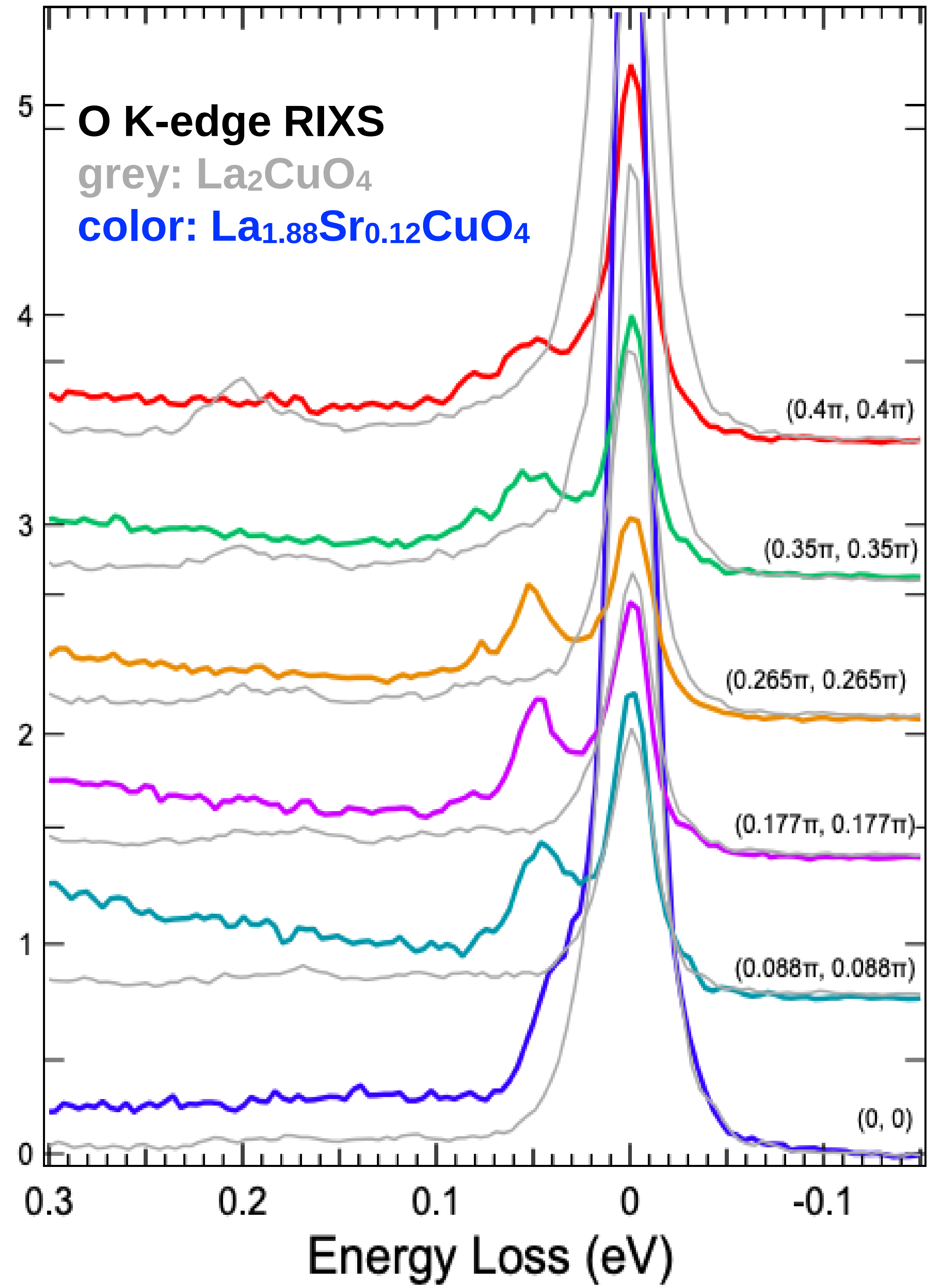
oxygen K edge for $Ca_{2+5x}Y_{2-5x}Cu_5O_{10}$

WS Lee et al., PRL **110,** 265502 (2013)



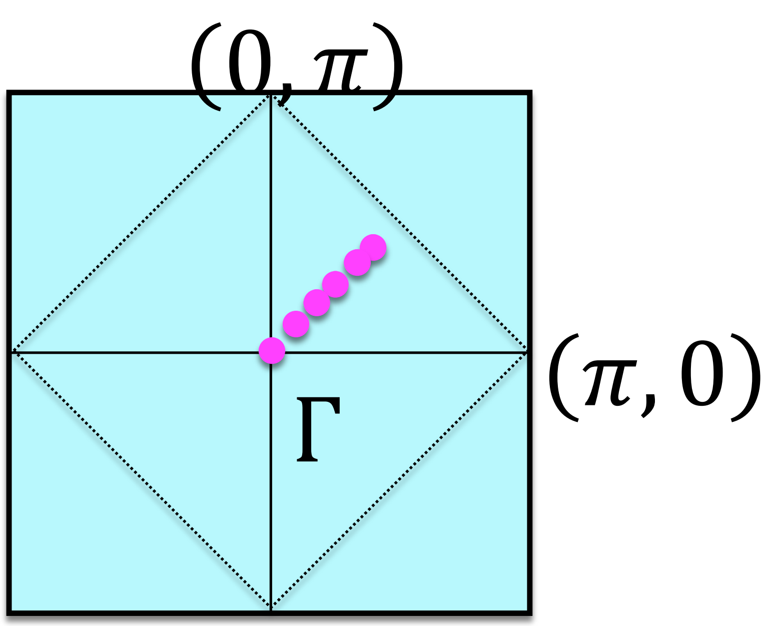
Electron-phonon coupling in $La_{2-x}Sr_xCuO_4$

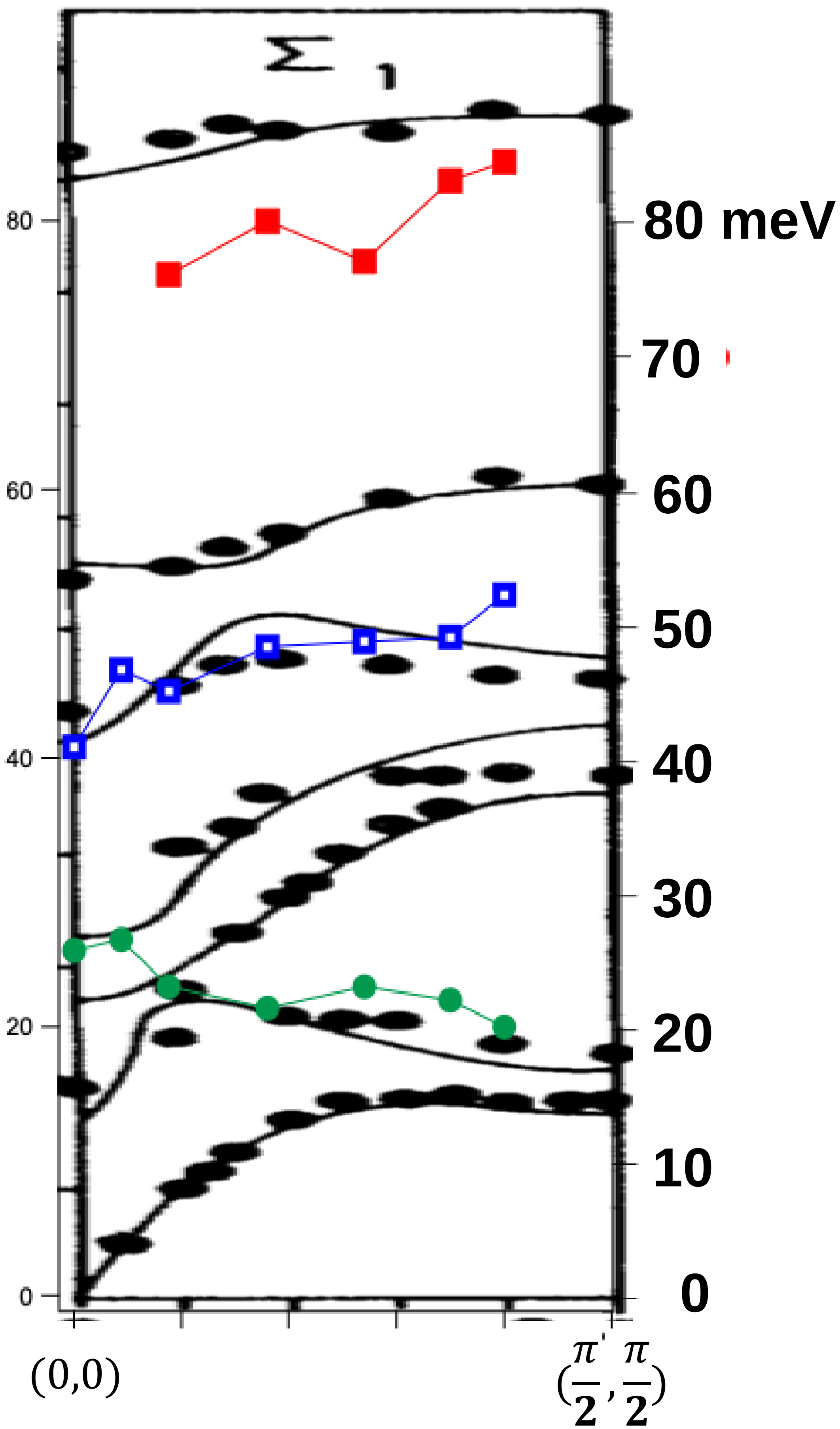


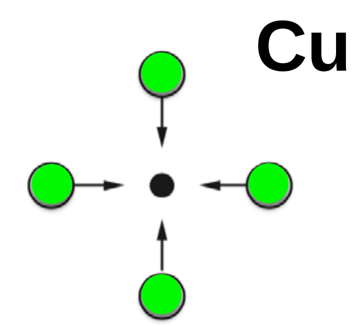


Comparison with the phonon dispersion from INS

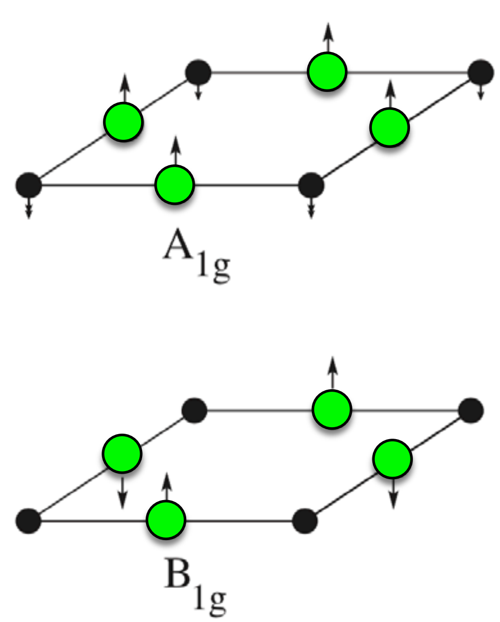
Chaplot et al.
PRB 52, 7230 (1995)
Fig. 3 ($La_{0.19}Sr_{0.1}CuO_4$)







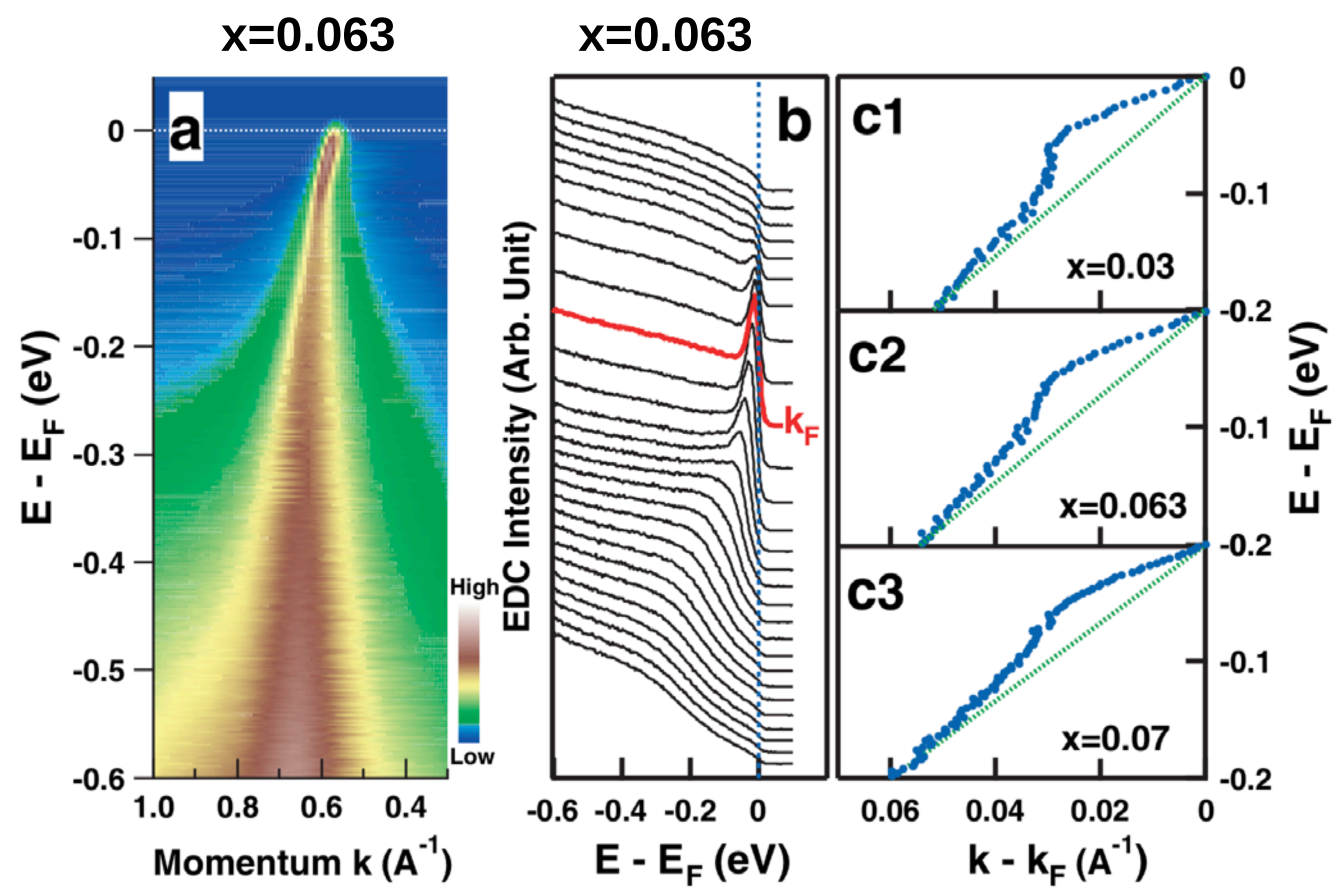
breathing



buckling

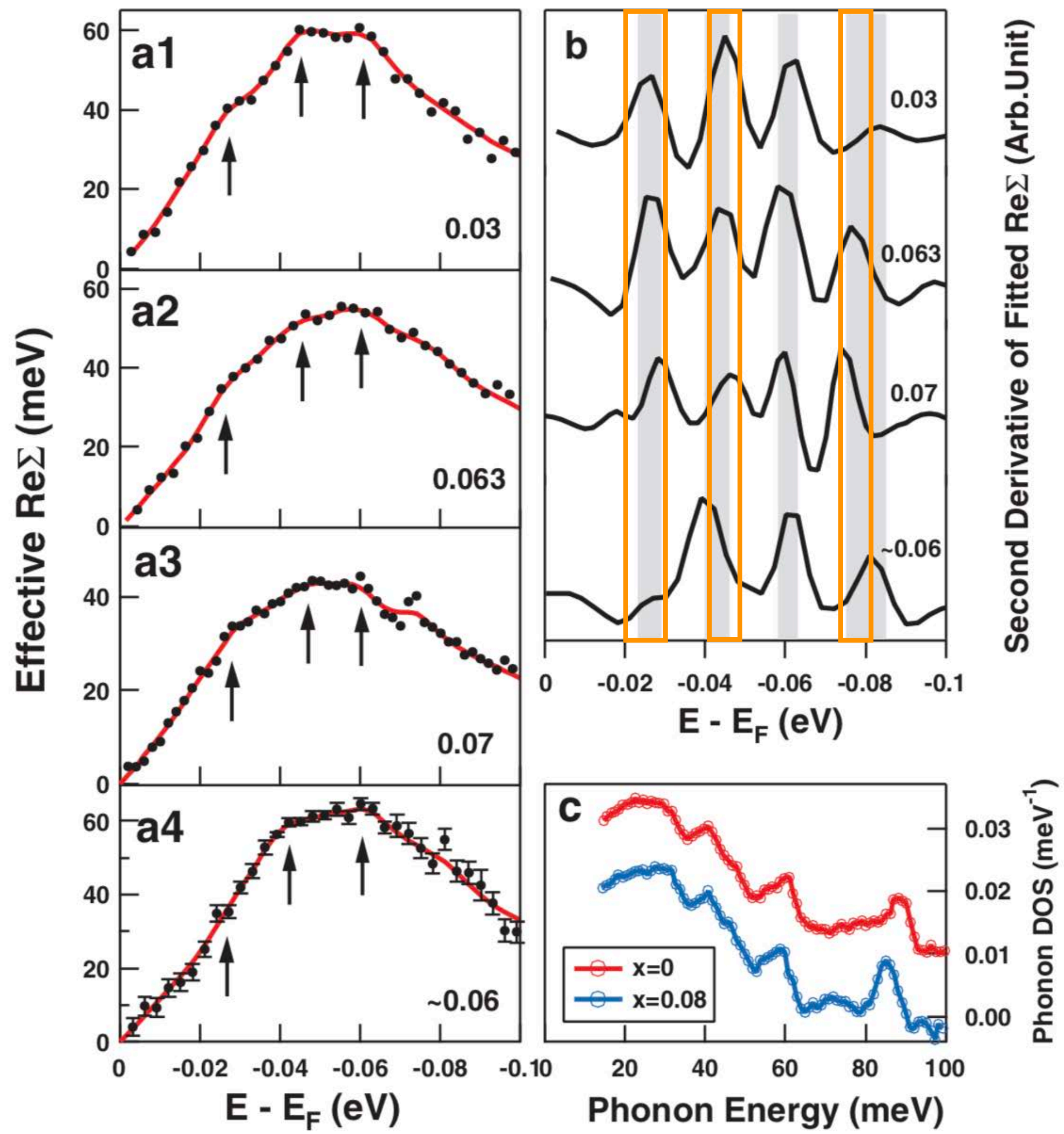
Comparison with ARPES and INS results

Zhou et al. PRL 95, 117001 (2005)



$$k_F = (0.44\pi/a, 0.44\pi/a)$$

RIXS



McQueeney et al.
PRL 87, 077001(2001)

ARPES kinks: integration of phonons of all Q's.

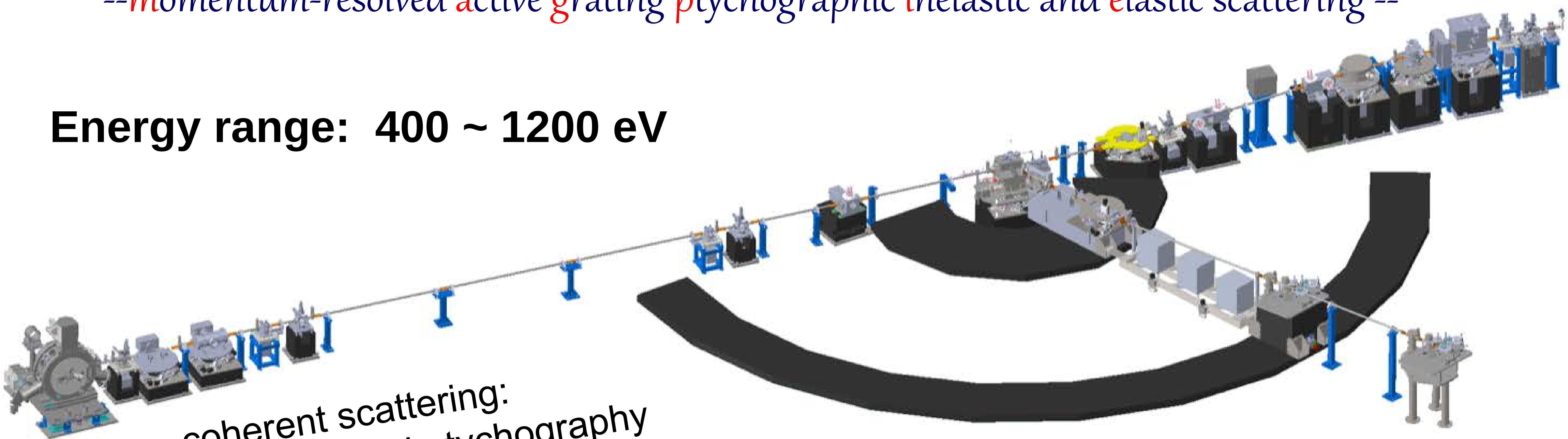
Summary

- The concept of energy compensation does work for RIXS
- High-resolution RIXS is powerful in revealing electronic excitations of oxides, particularly Q-resolved e-ph coupling.
- Charge-transfer excitons of ZRS existent in corner-sharing La_2CuO_4
- Magnon-magnon interactions in La_2CuO_4
- High-resolution IXS measurements on the electron-phonon anomaly in $La_{2-x}Sr_xCuO_4$ are on their way

TPS 41A *Blue Magpie*

--momentum-resolved active grating ptychographic inelastic and elastic scattering --

Energy range: 400 ~ 1200 eV



coherent scattering:
T-dependent CDI and ptychography

<u>at RIXS sample</u>

photon flux ≈ 7 x 10^{12}/sec @ 930 eV, BW=90 meV

beam size ≈ 5 μm (V) x 40 μm (H)

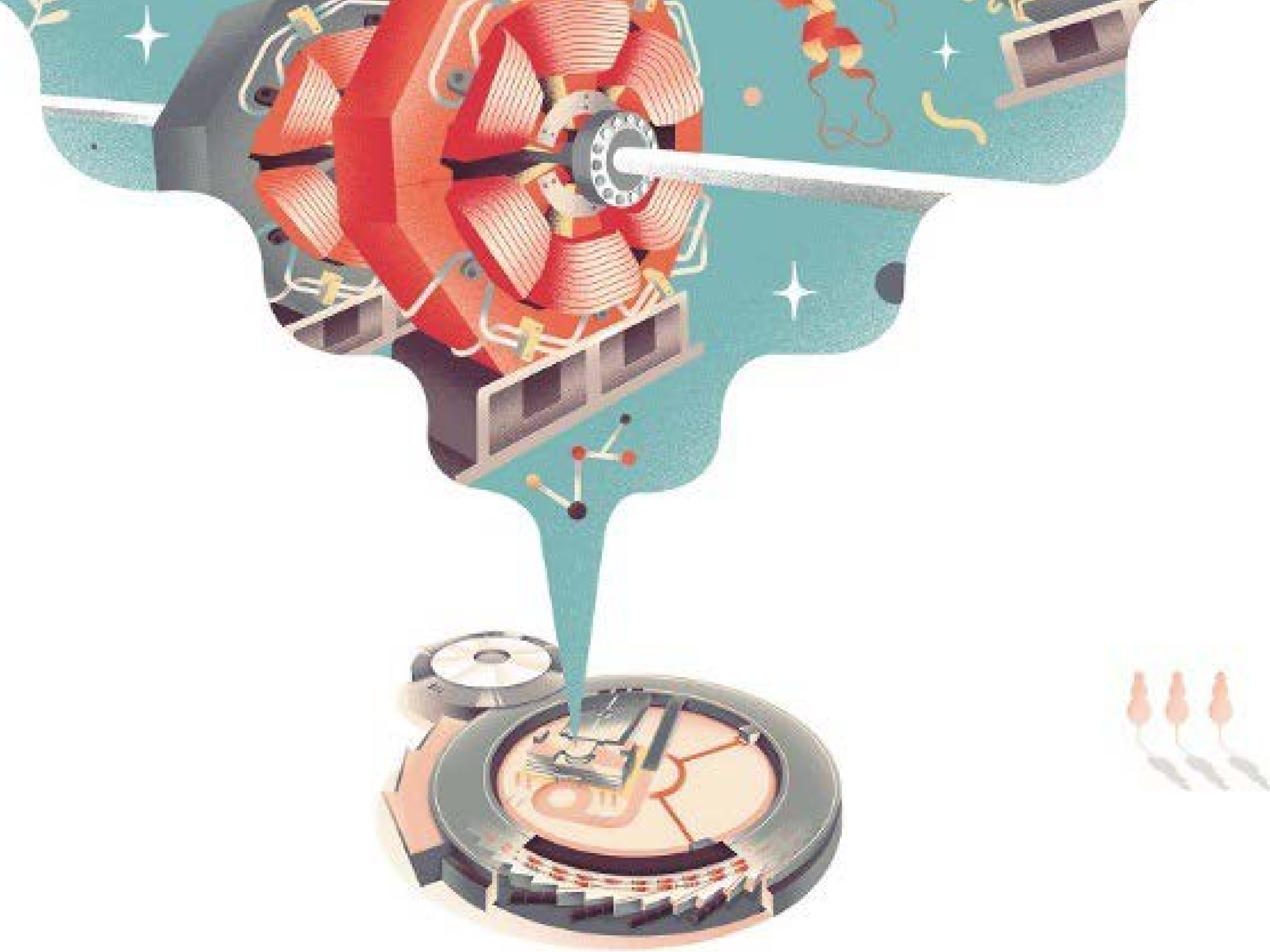
<u>RIXS resolving power</u>

active gratings (1200 lines/mm)+ in-vacuum LTP
+ 2-μm detector

⇨ **Goal: total resolving power of 100,000**

Taiwan blue magpie, a species of bird of the crow family

Thank you for
your attention.



Deadline for 2020-1 proposals: Sept 30, 2019

國家同步輻射研究中心
National Synchrotron Radiation Research Center