High-resolution momentum-resolved RIXS at Taiwan Photon Source

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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 $\rightarrow$ $\leftarrow$ count rate
Optical Concept of the AGM-AGS Beamline

Energy Compensation Principle

\[ \Delta E_{\text{total}} = \sqrt{\Delta E_{\text{mono}}^2 + \Delta E_{\text{spectr}}^2} \]

"large" exit slit

e.g. 50 or 100 μm

Test setup of AGM-AGS RIXS at the TLS

spin-state transition of LaCoO$_3$

diamagnetic insulator


rhombohedral unit cell

Co$^{3+}$, d$^6$

G. H. Jonker & J. H. van Santen (1953)
P. M. Raccah & J. B. Goodenough (1967)
LDA + U calculations:
Energy of IS state is lower than that of LS state above transition temperature.

Spin-state transition of a $d^6$ system

- Low $T$: large $10Dq$
- High $T$: small $10Dq$

(a) LS: $(t_{2g})^6$
- $e_g$
- $t_{2g}$

(b) $1A_{1g}$
- $3T_{2g}$
- $3T_{1g}$
- $1T_{1g}$

$S=0$ LS
$S=1$ IS
$S=2$ HS

Energy (eV) vs. $10Dq$ (eV)
The ground state is a LS state.
- At 100 K, mixture of LS & HS states
- At 500 K, close to a HS state.

\[ \sum_{i=1}^{3} V_i e^{-E_i / k_B T} \]
TPS 41A Soft X-ray Scattering

Photon energy: 450 eV – 1200 eV
Photon flux @900 eV: $5.6 \times 10^{12} \text{ Photons} \cdot \text{s}^{-1} \cdot (0.01\% \text{ BW})^{-1}

Tandem EPUs (3.5 m + 3.5 m)

Photon flux > $3 \times 10^{15}$

FWHM $\Sigma_x = 385 \mu\text{m}$
$\Sigma_y = 29 \mu\text{m}$
Momentum-resolved RIXS

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

- “active” (bendable) grating
- Varied-line-spacing (VLS) grating

\[ n(x) = n_0 + n_1 x + n_2 x^2 \]
\[ n_0 = 1200 \text{ mm}^{-1} \]

- 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.

\[ E_{\text{in}} = 530 \text{ eV} \]
“active” grating monochromator/spectrometer + LTP

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

0.05 $\mu$rad

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

$$f'_0(x) = c_1 + 2c_2 x + 3c_3 x^2$$

focusing ($c_2$) and comma ($c_3$) aberrations

slope diff = LTP − $f'_0(x)$

Resolving power: 24000
Energy resolution at 530 eV: 22 meV
Electronic States in La$_{2-x}$Sr$_x$CuO$_{4+\delta}$ Probed by Soft-X-Ray Absorption


O K-edge (1s $\rightarrow$ 2p) XAS

Zhang-Rice Singlet (ZRS)

ground state of an undoped cuprate

$\uparrow$ : spin of Cu 3$d$ holes

$\downarrow$ : spin of O 2$p$ holes
O K-edge RIXS of La$_2$CuO$_4$

1s $\to$ 2p XAS

polished La$_2$CuO$_4$(001)
in-plane $Q$ = (0.2$\pi$, 0), $T$=30 K
- 528.0 eV
- 527.0 eV
- 526.6 eV
- 526.3 eV
- 526.0 eV

$\sigma$ pol

Energy loss (eV) $\equiv h\nu_{\text{in}} - h\nu_{\text{out}}$
O K-edge RIXS of La$_2$CuO$_4$

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

ground state

$\hbar\omega$

intermediate state

O 1s $\rightarrow$ O 2p / Cu 3d

O 1s hole

O 2p hole

charge-transfer exciton

ZR singlet

final state

bi-magnon

O 1s hole

O 2p hole
\[(0, \pi/a) \equiv (0, \pi)\]

O K-edge RIXS of La$_2$CuO$_4$ (001)

\((\pi, 0)\) along (π, 0)

\((0.32\pi, 0)\) and \((0.55\pi, 0)\)

\(\Gamma\)

\(a\)

\(\Gamma\) along (0, 0), \(\pi/\alpha\) \(\equiv (0, \pi)\)

Energy Loss (eV)

\(x10^3\)

\(0\)

\(5\)

\(10\)

\(150\)

\(100\)

\(50\)

\(0\)

\(526\)

\(528\)

\(530\)

\(532\)

\(534\)

\(536\)

\(538\)

\(540\)

Photon energy (eV)

XAS

UHB

ZRS dd

bi-magnon
O K-edge RIXS of La$_2$CuO$_4$

Energy Loss (eV) vs. Energy Loss (eV)

- (0,0) → (π, 0) direction
  - (0.005π, 0)
    - 0.088π, 0.088π
      - 63.41°
      - 75°
    - (0.125π, 0)
      - 63.41°
      - 75°
    - (0.25π, 0)
      - 51.3°
      - 75°
    - (0.375π, 0)
      - 37.9°
      - 75°
    - (0.5π, 0)
      - 21.5°
      - 75°
    - (0.54π, 0)
      - 15.0°
      - 75°
- (0,0) → (π, π) direction
  - (0,0)
  - (0.088π, 0.088π)
    - 63.41°
    - 75°
  - (0.18π, 0.18π)
    - 51.3°
    - 75°
  - (0.27π, 0.27π)
    - 37.9°
    - 75°
  - (0.35π, 0.35π)
    - 21.5°
    - 75°
  - (0.4π, 0.4π)
    - 10.0°
    - 75°

Bimagnon
Charge
Phonon

?
Bi-magnon dispersion

with magnon-magnon interaction

w/o magnon-magnon interaction

J=0.15 eV

RIXS data

Vernay et al., PRB 75, 020403(R) (2007)
$La_{1.88}Sr_{0.12}CuO_4$
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 $\text{Ca}_{2+5x} \text{Y}_{2-5x} \text{Cu}_5 \text{O}_{10}$

WS Lee et al., PRL 110, 265502 (2013)
Electron-phonon coupling in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$

O K-edge RIXS
grey: $\text{La}_2\text{CuO}_4$
color: $\text{La}_{1.88}\text{Sr}_{0.12}\text{CuO}_4$

![Graph showing O K-edge RIXS spectra for different compositions of La$_{2-x}$Sr$_x$CuO$_4$.](image)
Comparison with the phonon dispersion from INS

Chaplot et al.
PRB 52, 7230 (1995)
Fig. 3 (La$_{0.19}$Sr$_{0.1}$CuO$_4$)

(0, $\pi$)

($\pi$, 0)

$\Gamma$

80 meV

breathing

buckling

(0,0)

$\frac{\pi}{2}$, $\frac{\pi}{2}$
Comparison with ARPES and INS results

Zhou et al. PRL 95, 117001 (2005)

$\mathbf{k}_F = \left( 0.44\pi/a, 0.44\pi/a \right)$

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

McQueeney et al. PRL 87, 077001 (2001)
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$_2$CuO$_4$
- Magnon-magnon interactions in La$_2$CuO$_4$
- High-resolution IXS measurements on the electron-phonon anomaly in La$_{2-x}$Sr$_x$CuO$_4$ are on their way
Energy range: 400 ~ 1200 eV

coherent scattering: T-dependent CDI and ptychography

at RIXS sample
photon flux ≈ 7 x 10^{12}/sec @ 930 eV, BW=90 meV
beam size ≈ 5 μm (V) x 40 μm (H)

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

Goal: total resolving power of 100,000
Thank you for your attention.

Deadline for 2020-1 proposals: Sept 30, 2019