Current status of resonant inelastic X-ray scattering spectroscopy (RIXS) and the need for nano-RIXS

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• Introduction.
• Compare different spectroscopic probes.
• Advantages and limitation of RIXS.
• Which edge to choose? TM L or M edges?
• Few examples to argue the need of nano-RIXS.
• Conclusions.
DOE BES five grand challenges –

- Control materials and processes at the level of electrons.
- Synthesize materials with tailored properties.
- Exploring emergent phenomena from complex correlations.
- Master energy/information flow at nanoscale with capability rivaling living things.
- Understand materials far from equilibrium states.

The goal is achieve the material control

Understanding complex phenomena requires sharper and sharper tools
Selection of experimental tools

**Physical/Chemical properties**

- Length: 10mm, 1mm, 100Å, 10Å, 1Å
- Frequency (Hz): 10^-17 to 10^14
- Energy (eV): 10^-17 to 10^1

- bulk properties
- absorptions polarizability resistivity specific heat magnetization ...etc.
- dynamics of glass and liquids
- proton transfer
- chemical bond breaking
- magnetic excitations
- orbital excitations
- phonons
- protein folding

**Experimental probes**

- Length: 10mm, 1mm, 100Å, 10Å, 1Å
- Frequency (Hz): 10^-17 to 10^14
- Energy (eV): 10^-17 to 10^1

- Hard X-ray
- Soft X-ray
- Deep UV (inelastic) scattering
- Neutrons
- Inelastic Scattering
- ARPES

**Microscopies**

- IXS
- APRES
- Neutrons

IXS, APRES and Neutrons are the ideal tools
Comparison between different probes

**Spectral Function**

**Single particle -**

**Charge:** Angle-Resolved Photoemission Spectroscopy (ARPES): \( I(k,\omega) \sim A(k,\omega) \)

**Two particles -**

**Spin:** Inelastic Neutron Scattering (INS): spin-spin correlation \( S(q,\omega) \)

**Charge:** Inelastic X-ray Scattering (IXS): charge-charge correlation \( S(q,\omega) \)

**Relative cross-section**

<table>
<thead>
<tr>
<th>Probe</th>
<th>Relative cross-section</th>
<th>Resolving power (E/(\Delta E))</th>
<th>Energy resolution ((\Delta E))</th>
<th>Energy range (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>ARPES (electron)</td>
<td>&gt;10^4</td>
<td>10eV/1meV (\sim 10^5)</td>
<td>1meV</td>
<td>(\sim h\nu (100\text{eV}))</td>
</tr>
<tr>
<td>INS (neutron)</td>
<td>&lt;&lt;1</td>
<td>100meV/0.1meV (\sim 10^3)</td>
<td>0.1meV</td>
<td>(\sim 1\text{eV})</td>
</tr>
<tr>
<td>RIXS (photon)</td>
<td>1</td>
<td>1-10keV/100meV (\sim 10^4-5)</td>
<td>100meV ((\sim 10\text{meV}))</td>
<td>(\sim h\nu (\text{keV}))</td>
</tr>
</tbody>
</table>

RIXS is a powerful probe, but needs improvements -
- Higher throughput
- Better resolution
- More capabilities

*J. Stohr*
RIXS spectroscopy

Advantages of RIXS

- Directly coupled to charge degree of freedom.
- Bulk sensitive – study interface properties; in-situ studies.
- Charge neutral process – works with presence of electric/magnetic field.
- Not limited by optical transition rule – dd excitations.
- Finite momentum-transfer – study indirect bandgap and exciton dispersions.
- Resonance effect - resonance enhancement of the electronic contribution and elemental selectivity.
- Symmetry selective – polarization dependence.
- Not limited by core-hole lifetime – $\Delta E$ can be greatly improved.
- Fast dynamics – internal clock set by the core-hole when incident energy is detuned away from elemental edges.

... but RIXS has certain limitations (resolution and throughput)
RIXS at different edges

<table>
<thead>
<tr>
<th>Edge</th>
<th>Energy range</th>
<th>Resolving power for 10meV</th>
<th>pros</th>
<th>cons</th>
</tr>
</thead>
<tbody>
<tr>
<td>L</td>
<td>500eV ~ 1keV</td>
<td>1keV/10meV = 10^5</td>
<td>large Δq small elastic peak on resonance fewer branching...</td>
<td>large machine lower throughput (with reasonable OE)</td>
</tr>
<tr>
<td>M</td>
<td>50~100eV</td>
<td>100eV/10meV = 10^4</td>
<td>small Δq smaller machine larger throughput</td>
<td>strong elastic peak more branching</td>
</tr>
</tbody>
</table>

S.G. Chiuzbaian et al. PRL 95, 197402 (2005)
L. Braicovich et al, PRL 102, 167401 (2009)
J. Schlappa et al., PRL 103, 047401 (2009)
G. Ghiringhelli et al. PRL 102, 027401 (2009)
magnetic excitation (double-triplon) in Sr_{14}Cu_{24}O_{41}

Finite momentum transfer can be helpful in exploring the dispersion of excitations; however, could be an issue as well...

Bi-magnon dispersion in layered cuprate (?)

Single magnon dispersion probed by IXS and neutron
Cu K-edge RIXS spectrum (neglecting polarization, 4p)

$q_{in}=(0,0), q_{out}=(0,0), \Delta q =0$

$q_{in}=(\pi,0), q_{out}=(\pi,0), \Delta q=0$

RIXS is probing the wave functions of the involving states

$$I(\omega_i, \Omega = \omega_i - \omega_f) \propto \sum_f \left| \sum_i \sum_{4p} \langle \psi_f | \sum_l p_{4p}^{1} s_l e^{i q_{out} \cdot r_1} | \psi_{ci} \rangle \langle \psi_{ci} | \sum_j p_{4p} s_j^{1} e^{i q_{in} \cdot r_1} | \psi_{0} \rangle \right|^2 \times \delta (E_{f} - E_{i} - \hbar \Omega)$$
Scientific case for nano-RIXS

Crystal field effect

\[ L\text{-edge, } \Delta E \approx 100\text{meV} \]

- Crystal field excitation \((dd)\) + Coulomb repulsion play important roles in RIXS spectra
- Sensitive to intra-atomic distance (bond length)
- Sensitive to TM valency
- Sensitive to symmetry

A powerful probe for local electronic environment – important for determining the electronic structures of nanoparticles

Co nanoparticles

- Surfactant: Oleic Acid, $C_{18}H_{34}O_2 \left( CH_3 (CH_2)_7 CH:CH(CH_2)_7 CO_2 H \right)$
- Solvent: Dichlorobenzene, $C_6H_4Cl_2$

Charge transfer @ nanoscale


Courtesy of Jinghua Guo
Bandgap engineering

J. Guo et al. PRB 61, 9140 (2000)

Courtesy of Jinghua Guo
Elementary excitations in correlated systems

Magnon in 2D spin $\frac{1}{2}$ Sr$_2$CuO$_2$Cl$_2$

M. Guarise et al. PRL 105, 157006 (2010)
C. Ulrich et al., PRL 103, 107205 (2009)
Strong phase separation makes the identification of the origin of excitations much harder.

- Strong memory effect
- Kinetic arrest due to lattice distortion

Strong phase separation in correlated systems makes the identification of the origin of excitations much harder.

Conclusion

- Nano-RIXS is a powerful probe for studying local electronic structures – if we have the energy resolution and/or flux (near elastic peak and around crystal field excitations).
- Limitation on throughput and energy resolution will determine the scientific scope.
- Potential impact to energy science – especially in the *in-situ* research.
- Important for understanding correlated electron system when electronic phase separation dominates.

Thank you