Time, momentum and energy-resolved measurements of transient magnetism in quantum materials

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Workshop on Quantum Materials & Nanotechnology
University of Southampton, November 2019

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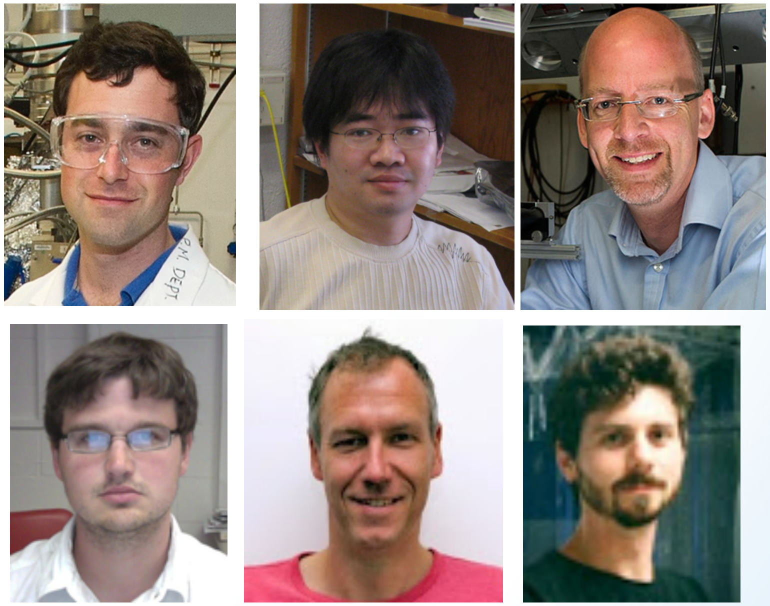
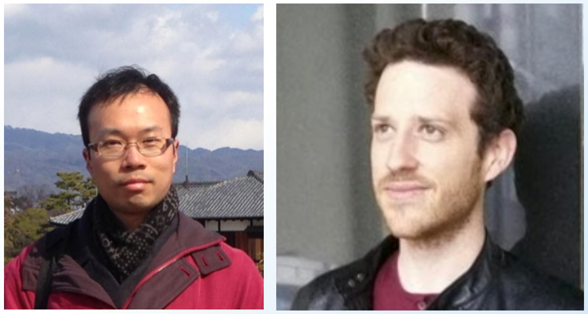




LCLS
D. Zhu

SACLA
S. Owada et al.

Advanced Photon Source
D. Casa, J.-H. Kim, M. Upton

And a few others still…

Outline

Why tr-RIXS?

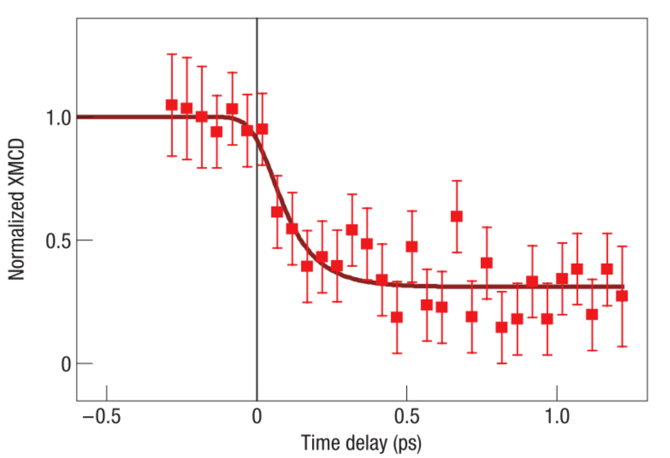
Ultra-fast RIXS measurements of magnetic correlations in Sr_2IrO_4

Extension to $Sr_3Ir_2O_7$

Measuring magnetism in transient states

Spin degree of freedom is often crucial

XMCD Ni



Stamm et al. Nat. Mat. (2007)

Possible to measure net magnetic moment:

- XMCD/ MOKE / Faraday effect Etc.

How can one go beyond this?

Energy/momentum space

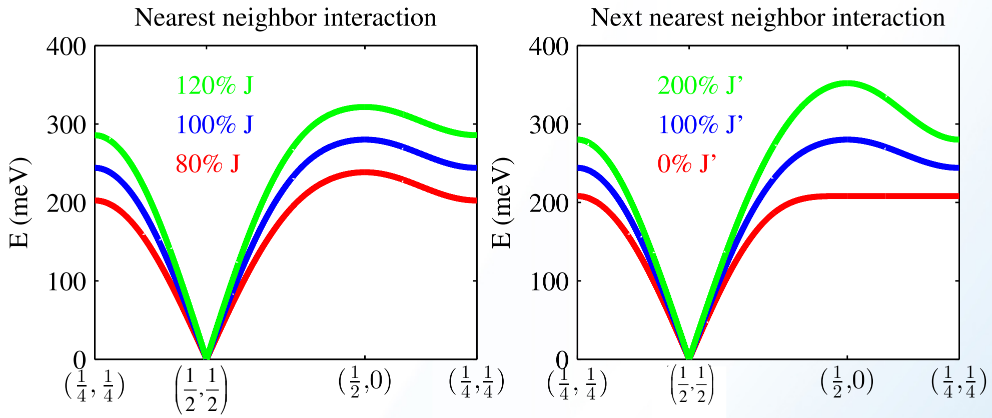
Magnetic excitation spectrum

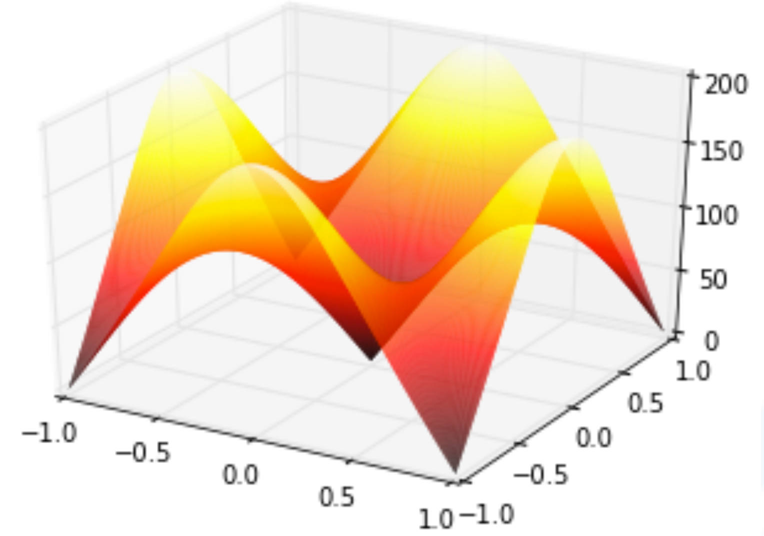
Inelastic neutron scattering (not time resolved)

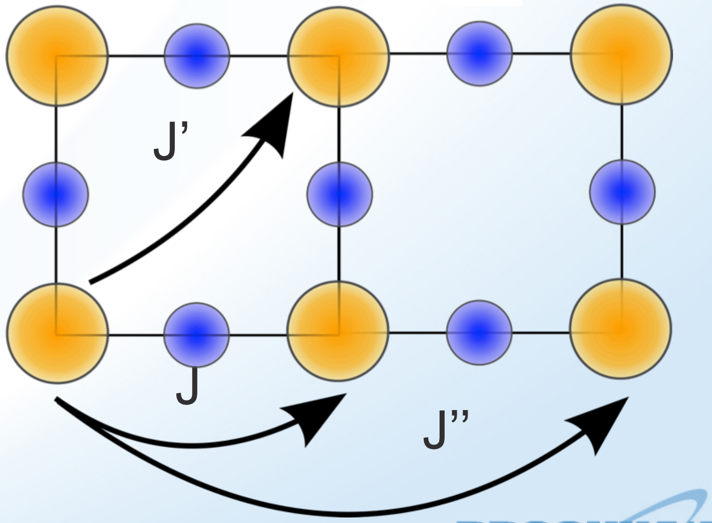
Resonant inelastic x-ray scattering

Sr_2IrO_4

2D AF Heisenberg model



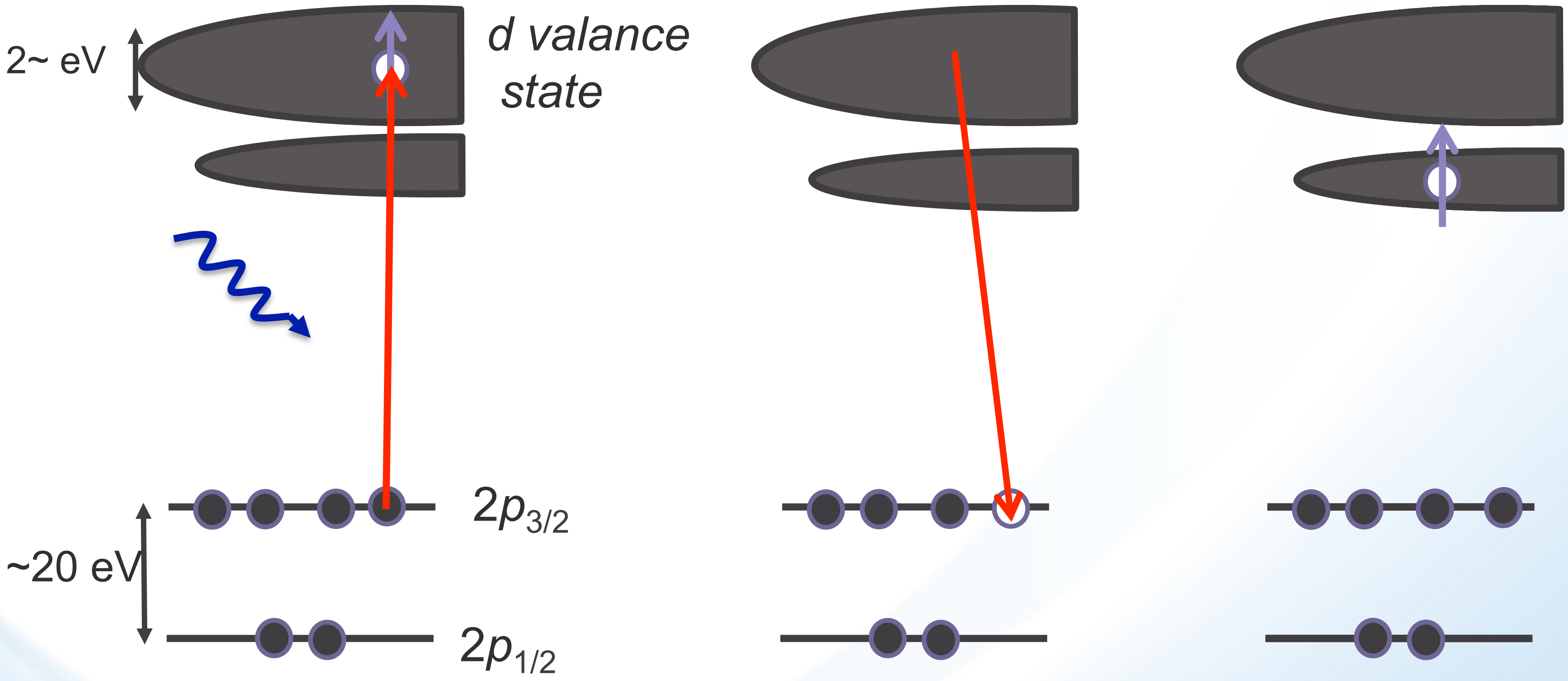




Jungho Kim et al., PRL (2012)

Resonant enhancement of sensitivity to valence electrons

L-edge RIXS

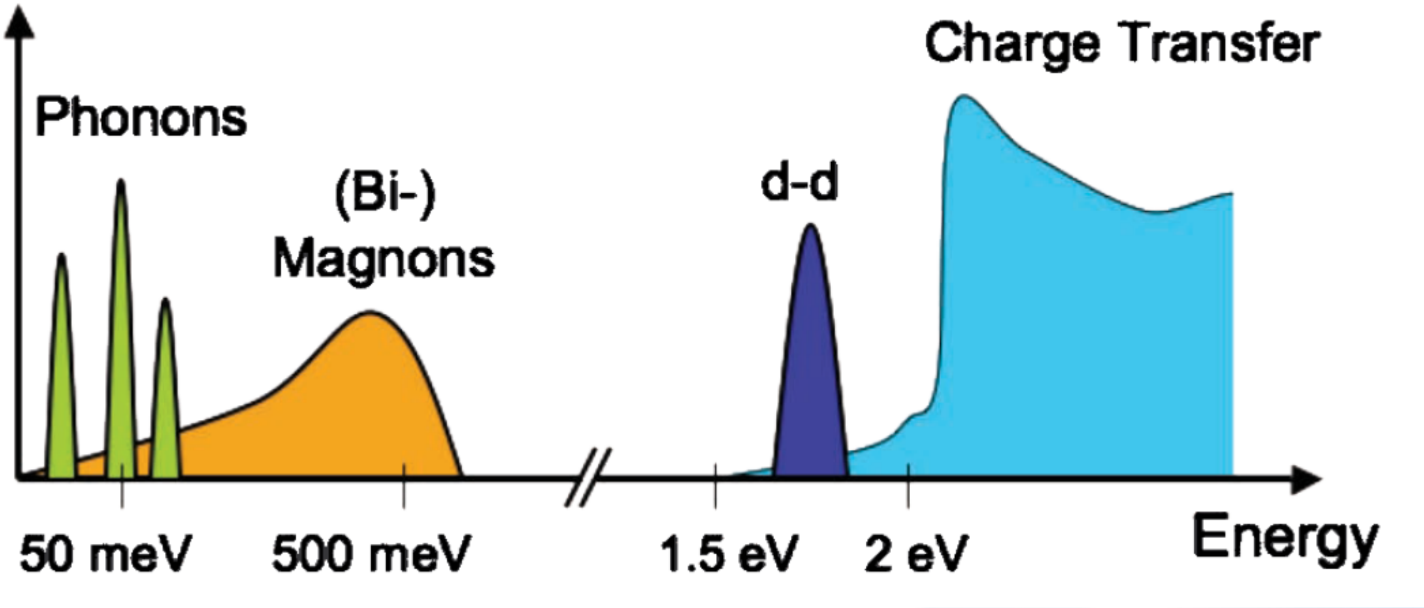


SO coupling

L. J. P. Ament et al., Rev. Mod. Phys 83, 705 (2011)
M. P. M. Dean JMMM 15, 3-13 (2015)

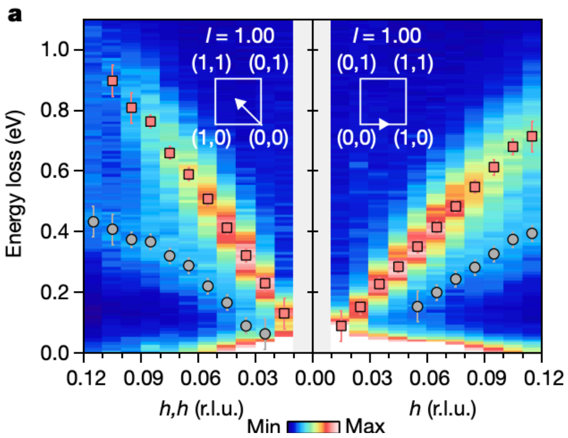
Excitation spectrum of a strongly correlated electron material

Mott insulator La_2CuO_4



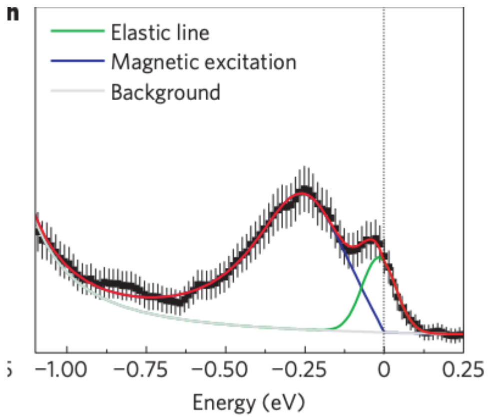
X-rays can access all these excitations providing very stringent tests for model Hamiltonians related to all these degrees of freedom.

Plasmons



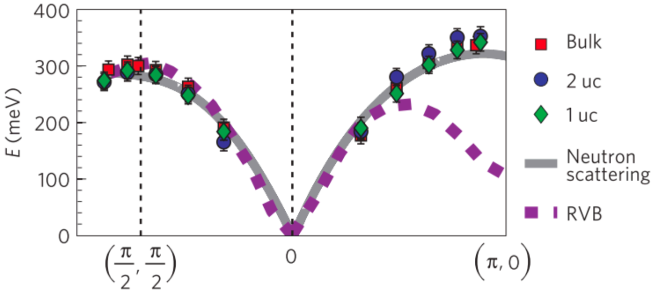
Hepting et al., Nature. (2018)

Paramagnons



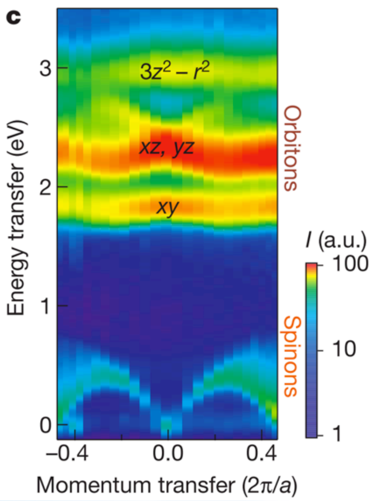
Le Tacon et al. Nat. Phys. (2011)
Dean et al., Nat. Mat. (2013)

Magnon dispersion in heterostructures



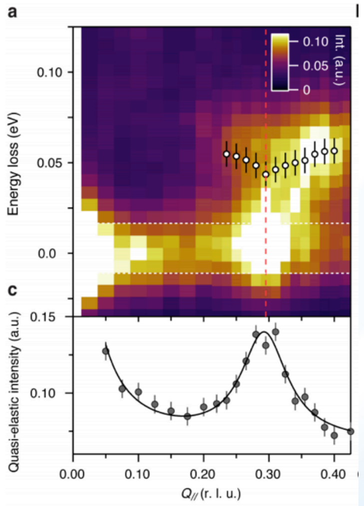
Dean et al., Nat. Mat. (2012)

Orbitons



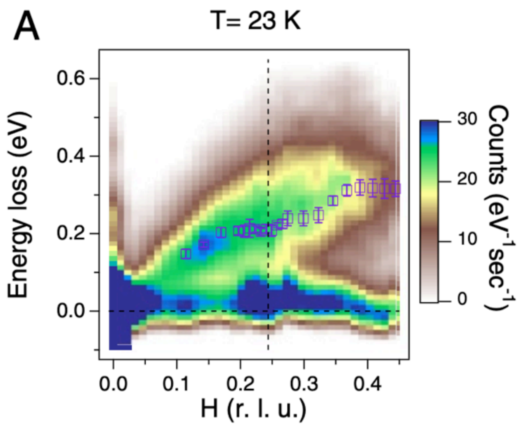
Schlappa et al., Nature (2012)
Kim et al., Nat. Comm. (2014)

Phonon coupled to a CDW



Chaix et al.,
Nat. Phys. (2017)

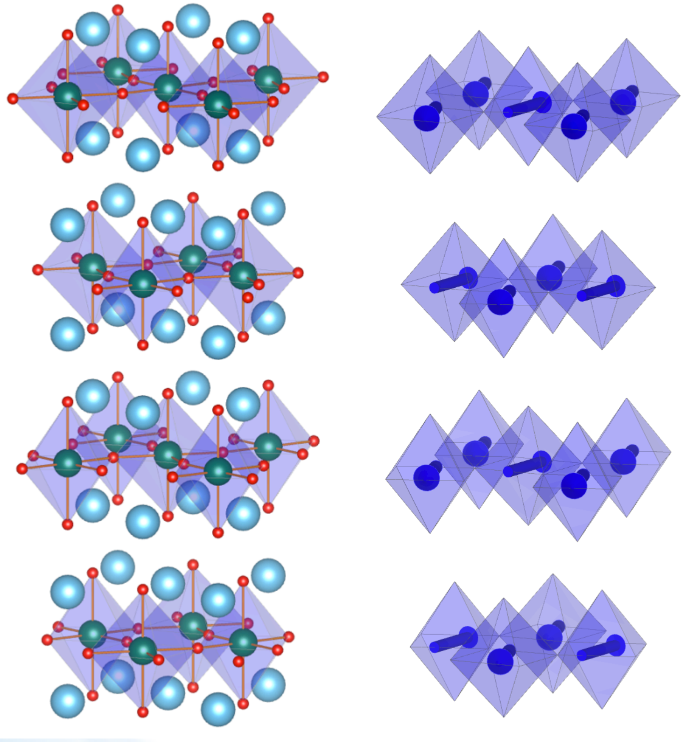
CDW modifying a magnon

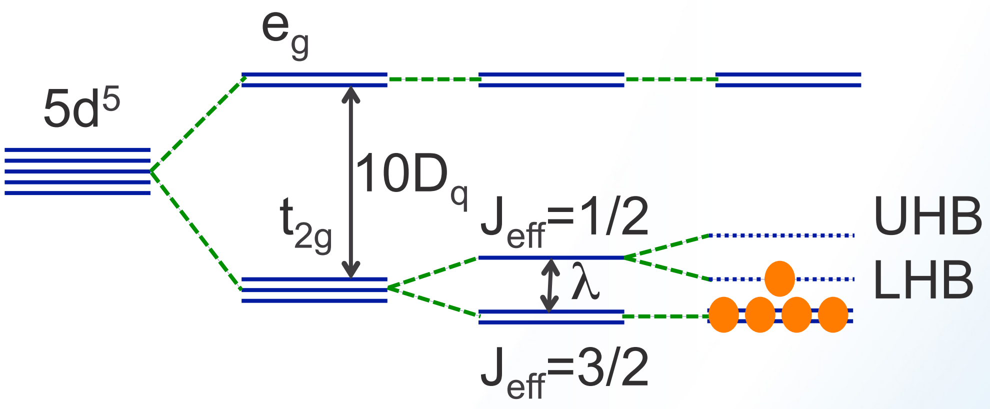


Miao et al., PNAS (2017)

Model system Sr_2IrO_4

T_N = 240 K
All data 110 K



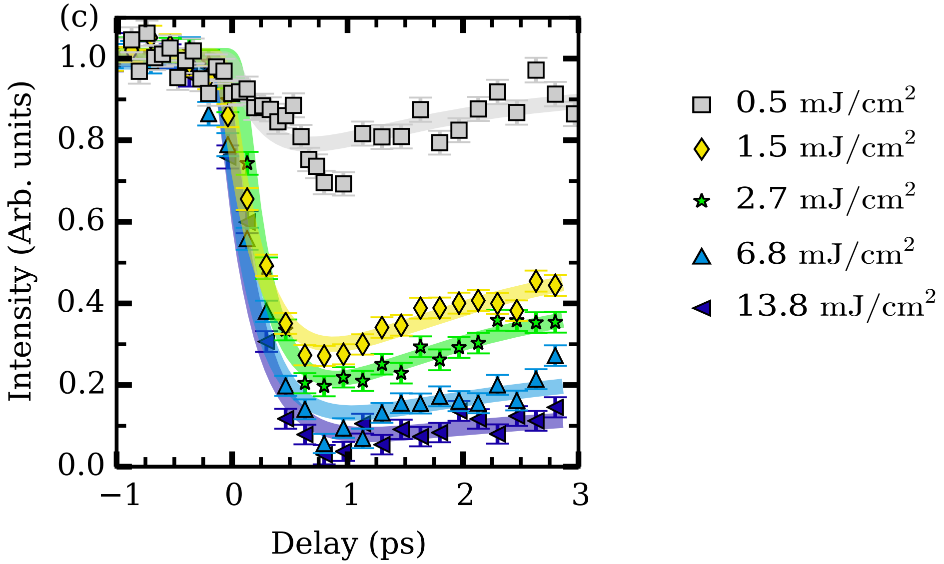


Forms a novel spin-orbit Mott insulator

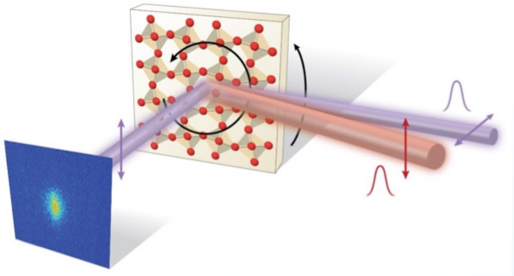
Close analogue of cuprate high temperature superconductors

S. Boseggia et al., J. Phys. Cond. Mat. (2013)

Destruction of long range magnetic order



Experiment at SACLA



3D long-range magnetic destroyed in < 300 fs

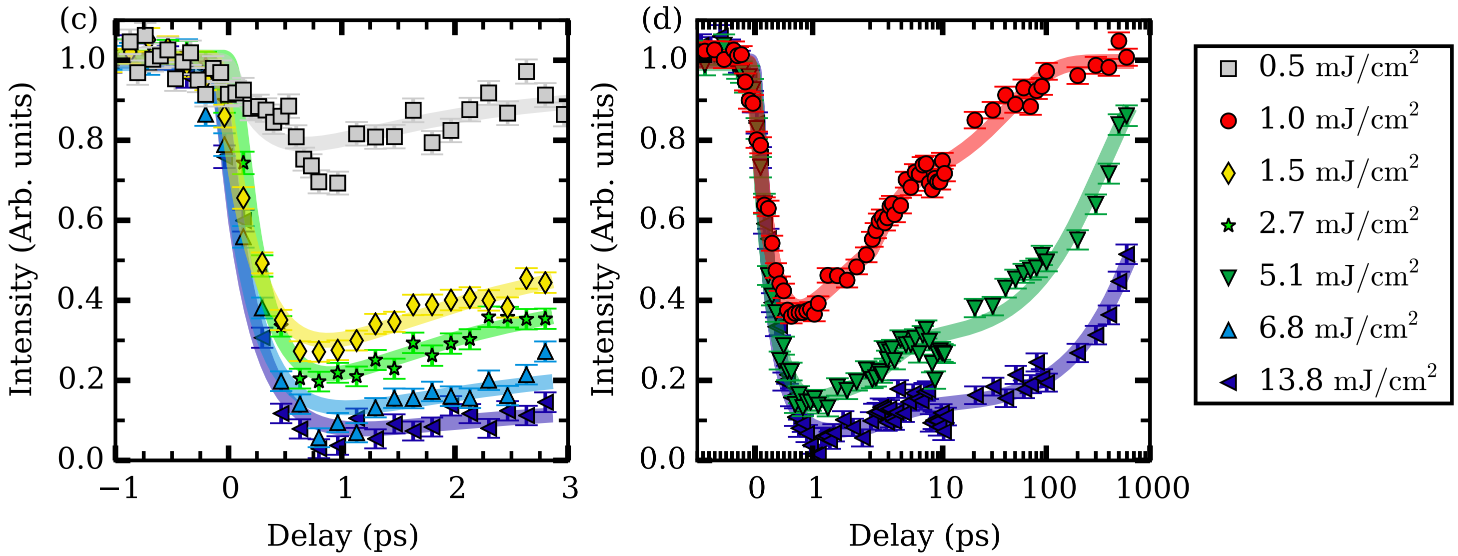
If fluence> 5 mJ/cm^2

Similar to $La_{0.5}Sr_{1.5}MnO_4$ and $La_{1.75}Sr_{0.25}NiO_4$

Ehrke et al., PRL (2011); Chuang et al, PRL (2013)

Magnetic recovery dynamics

M. P. M. Dean, et al. Nature Materials 15 601–605 (2016)

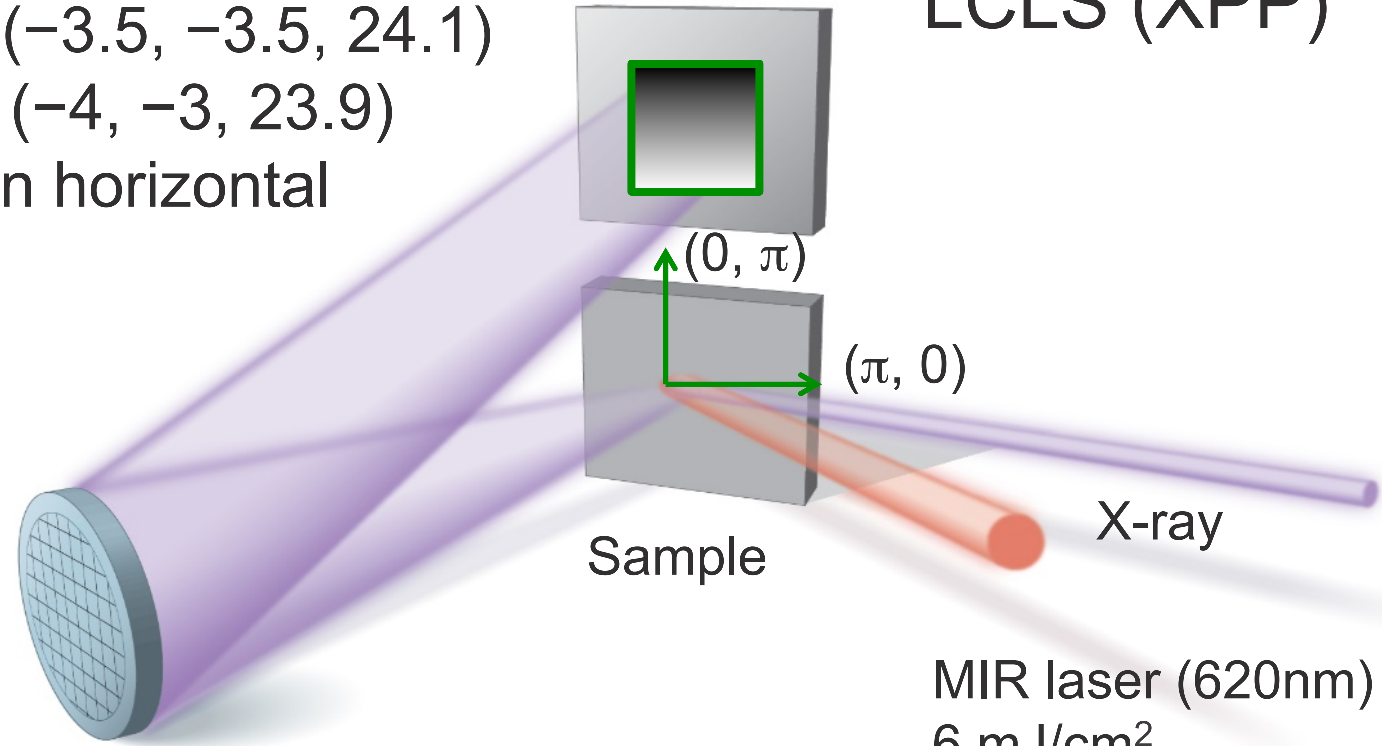


$$I(t) = I_0\Big(\exp(-t/\tau_{\mathrm{decay}}) + C\left[1 - \exp(-t/\tau_{\mathrm{2D}})\right] + (1-C)\left[1 - \exp(-t/\tau_{\mathrm{3D}})\right]\Big),$$

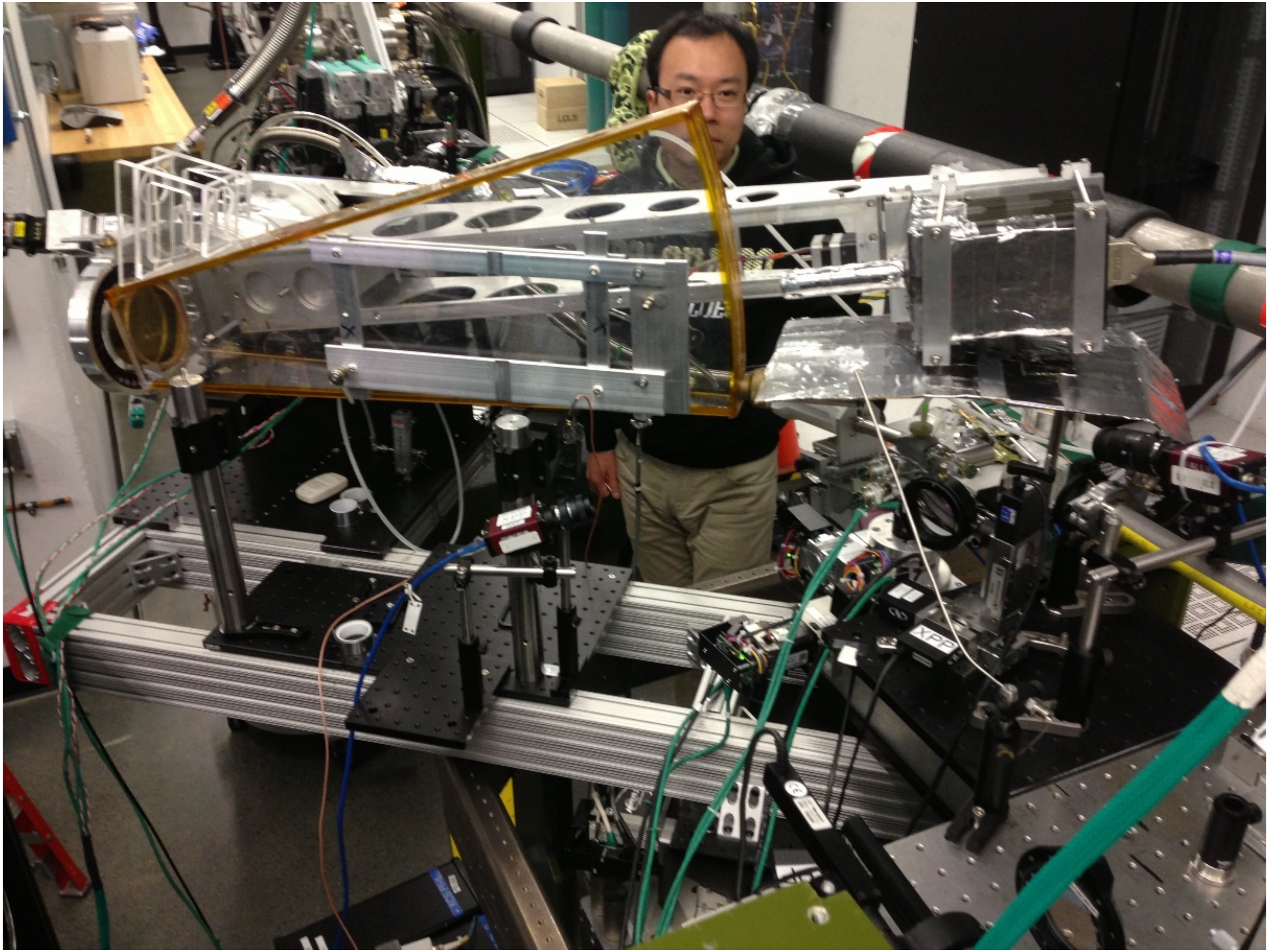
RIXS: Experimental schematic

E=11.215 keV (Ir L_3)
(π, 0) at (−3.5, −3.5, 24.1)
(π, π) at (−4, −3, 23.9)
2θ = 90 in horizontal

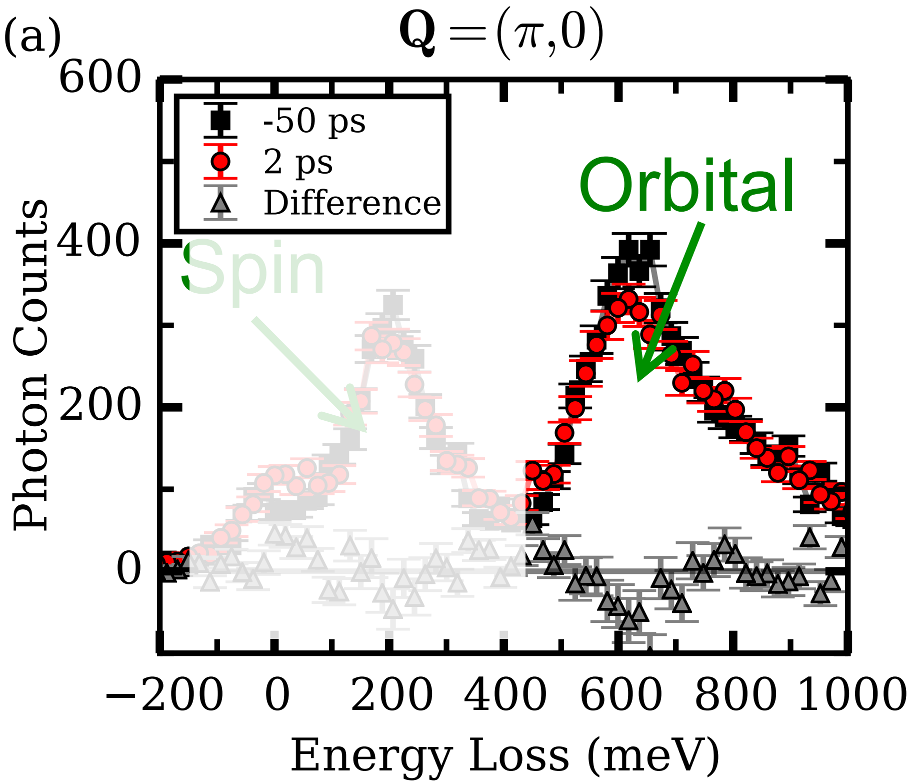
Experiment at LCLS (XPP)



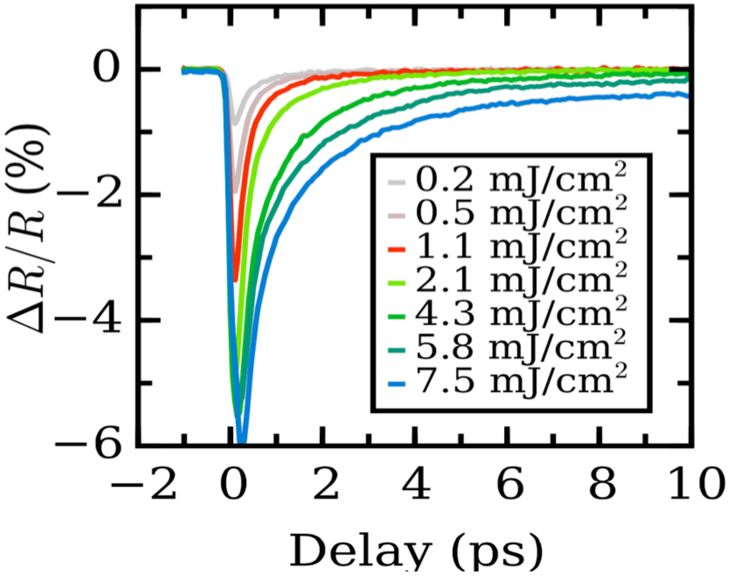
MIR laser (620nm)
6 mJ/cm^2
LHB -> UHB


LCLS
XPP
XPP

RIXS: Orbital transition



Optical reflectivity



Orbital excitation recovers in <2 ps

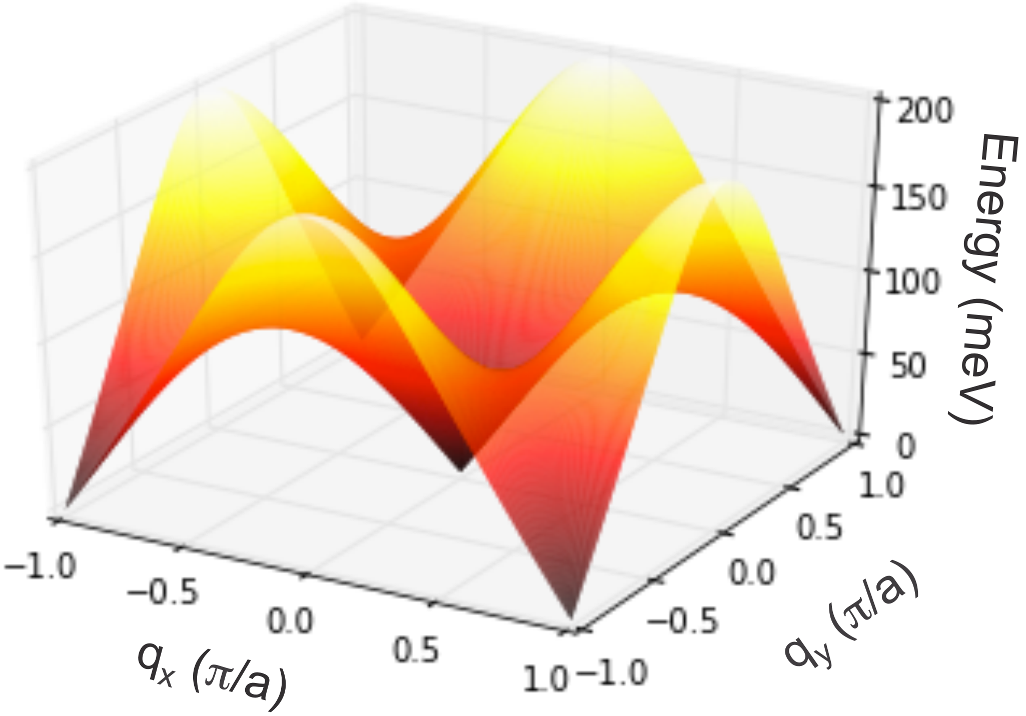
Charges have decayed out of the UHB at 2 ps

Equilibrium magnetic excitation spectrum

$$\left\langle S(r=0,t=0)\,S^{+}(r,t)\right\rangle$$

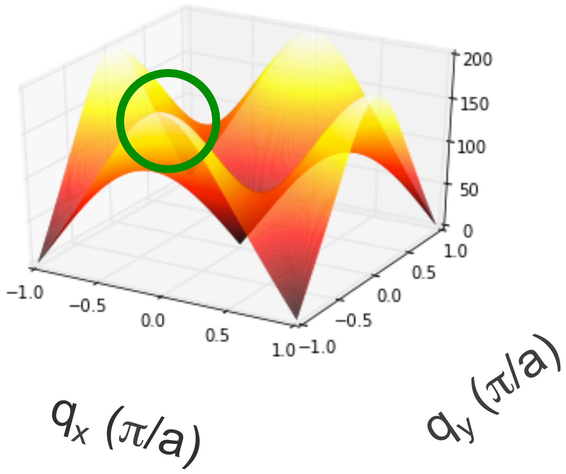
For Sr_2IrO_4

- Spin wave spectrum
- 2D excitations independent of q_z



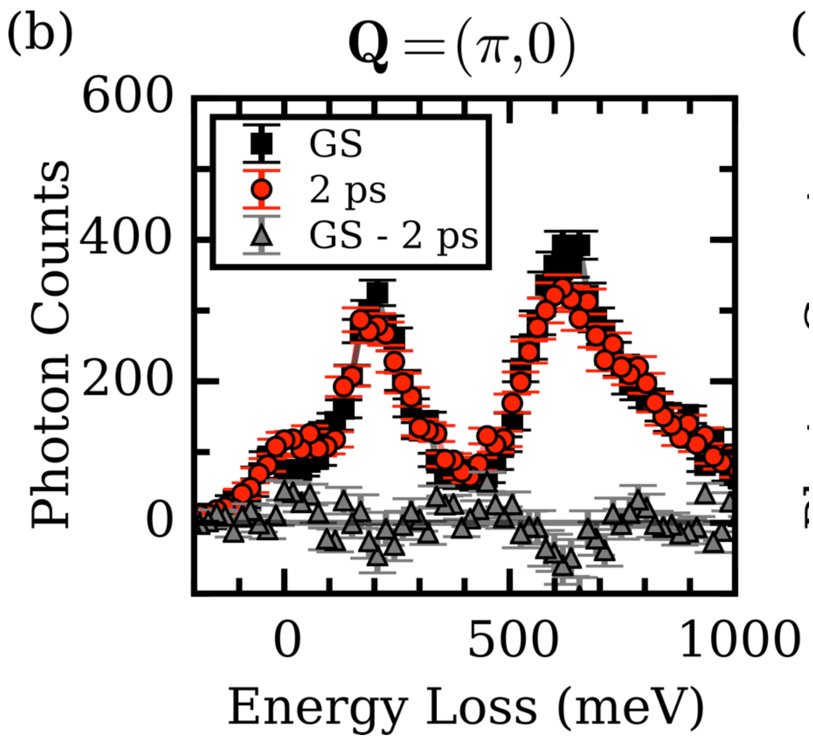
Jungho Kim et al. PRL 108, 177003 (2012)

RIXS: Magnetic correlations at (π,0)

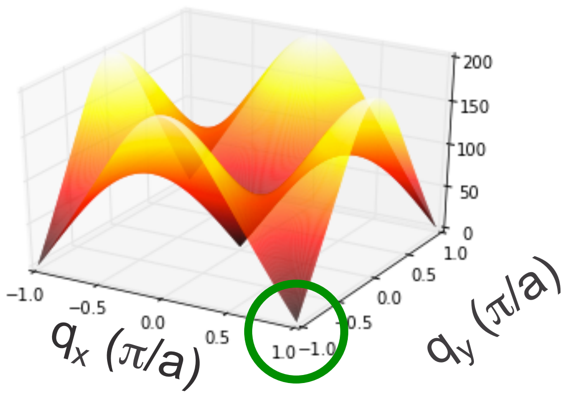


Proof of 2D correlations in melted state

(π,0) – nearest neighbour correlations recovered < 2 ps

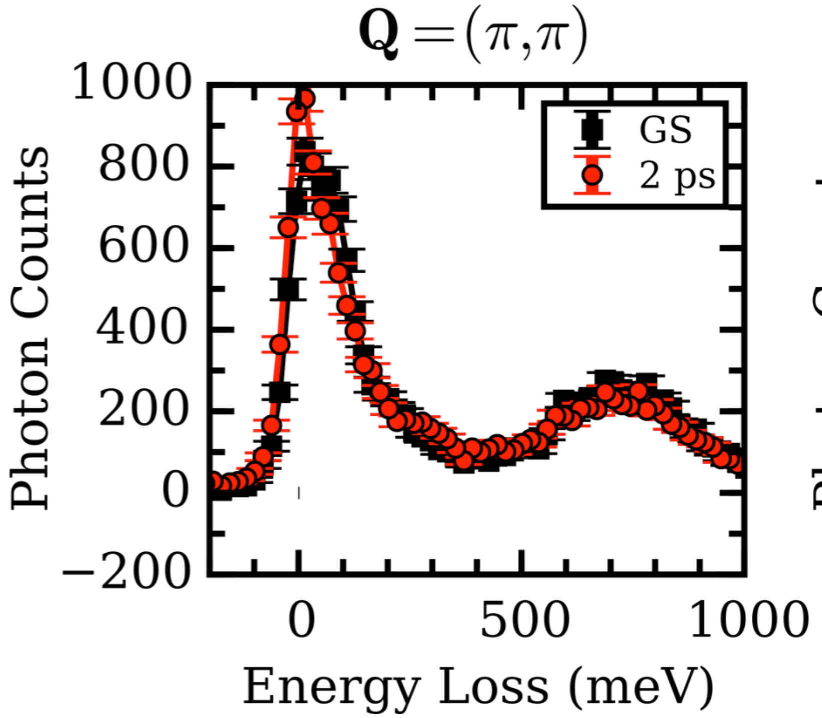


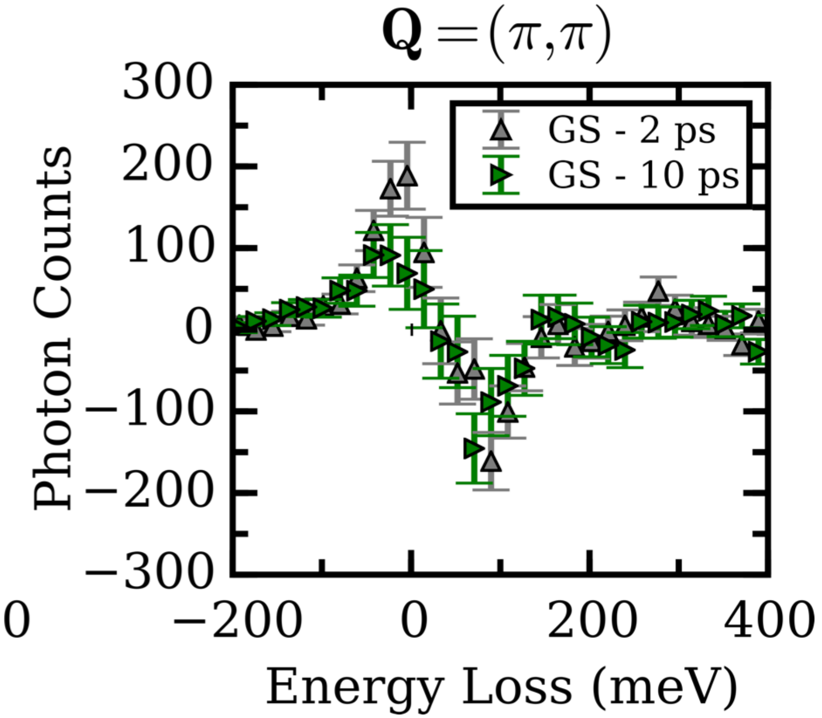
RIXS: Magnetic correlations at (π,π)



20% change in magnetic spectral weight (π,π)

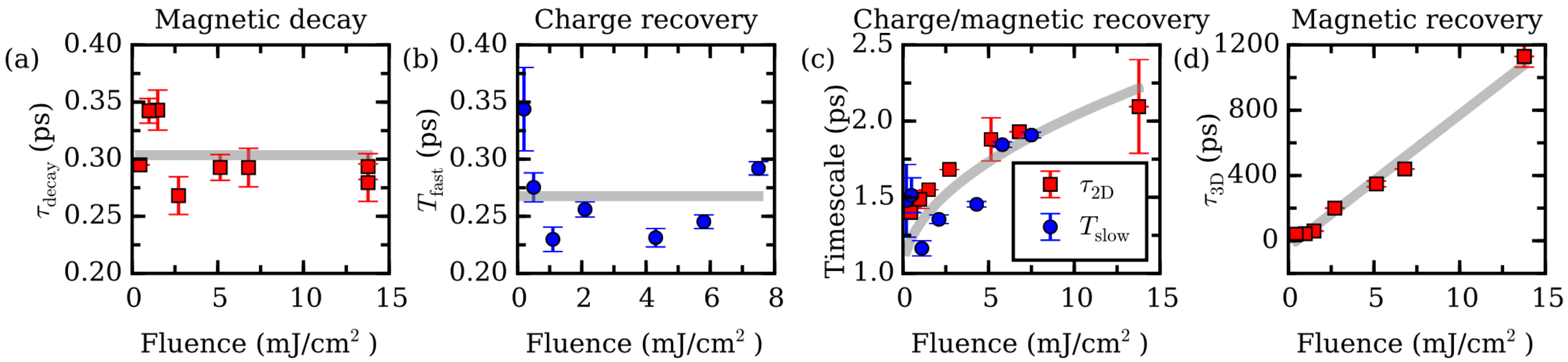
Not trivial thermal broadening





Magnetic versus charge evolution

M. P. M. Dean, et al. Nature Materials 15 601–605 (2016)



Spin channel

τ_{decay} <0.3 ps → τ_{2D} ~2 ps → τ_{3D} ~100-1000 ps

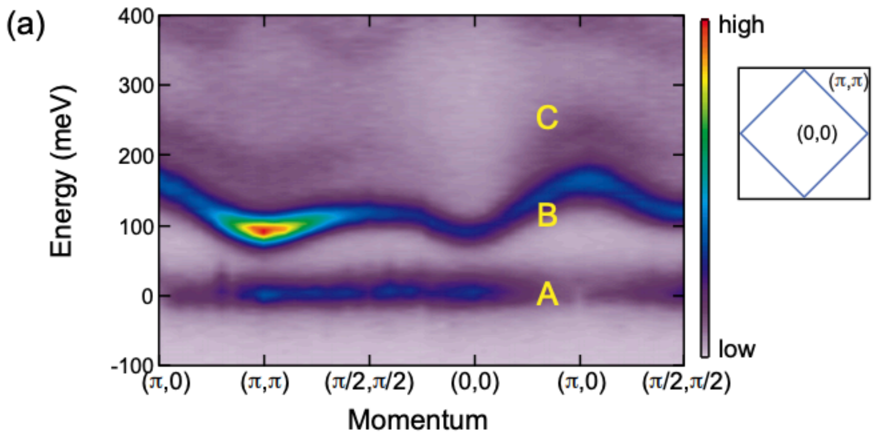
Charge channel: T_{fast} and T_{slow} from optical reflectivity

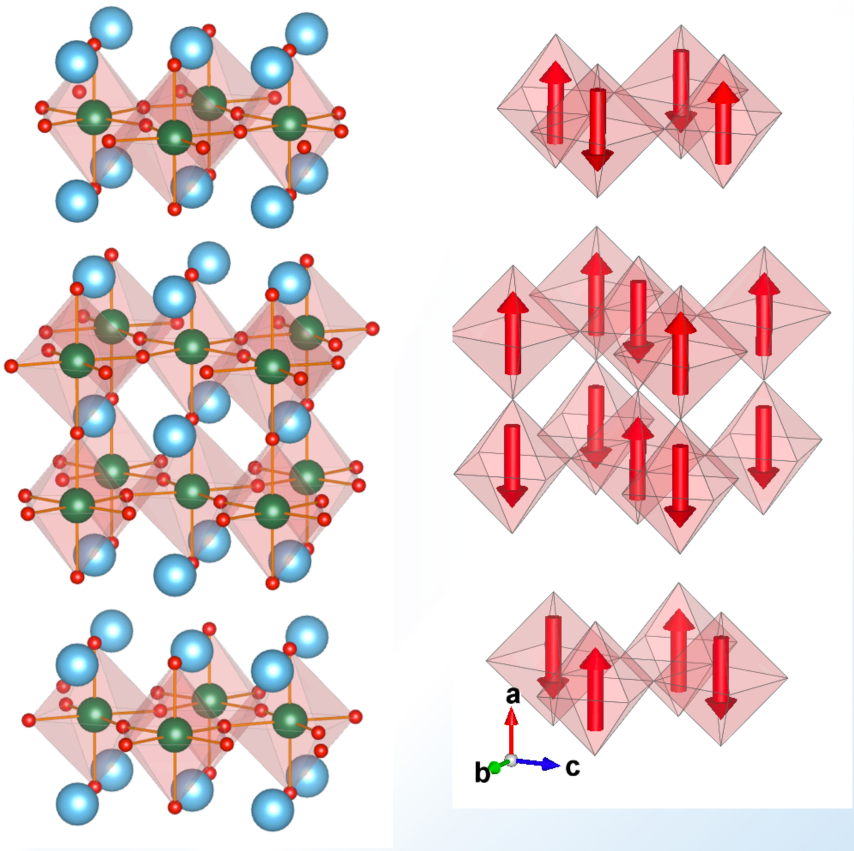
$Sr_3Ir_2O_7$

Bilayer AFM

Close to M-I transition

Big spin gap





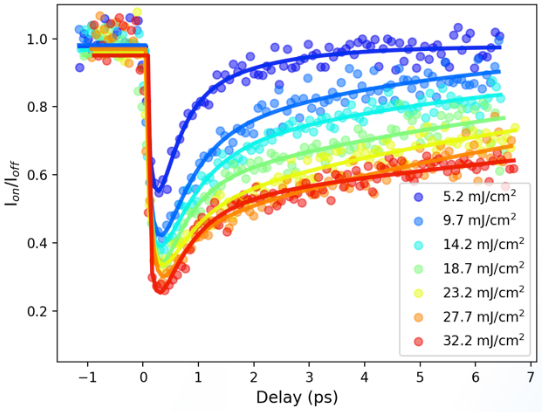
Jungho Kim et al., PRL (2012)

Data from $Sr_3Ir_2O_7$

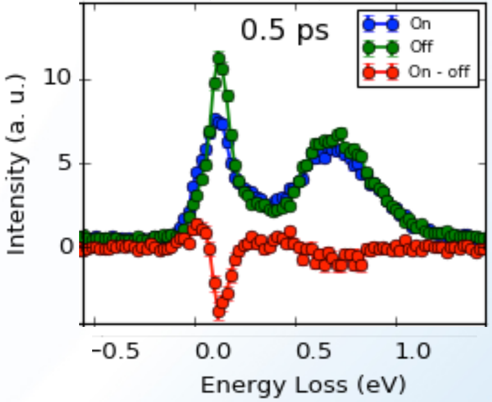
Persistent magnon in transient state at 500 fs persisting for several ps

Dynamics at 500 fs at both symmetry points persisting for several ps.

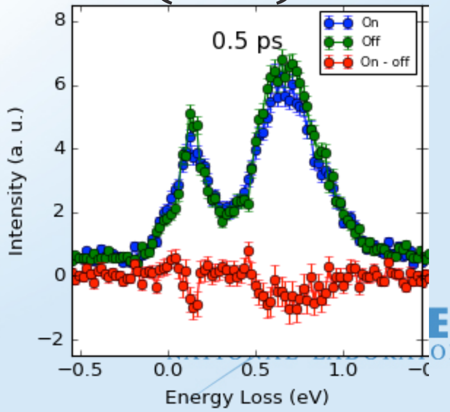
Spin-gap **bottleneck**



(π,π)

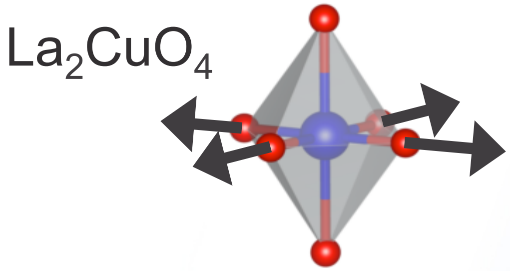


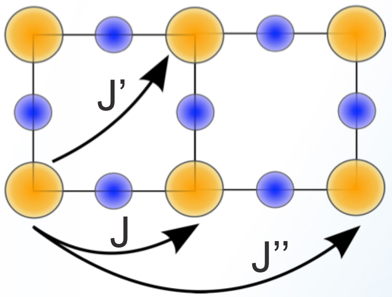
$(\pi,0)$

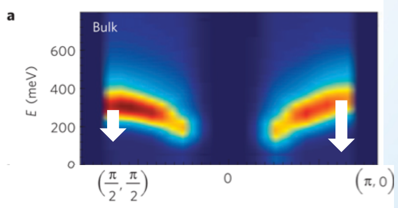


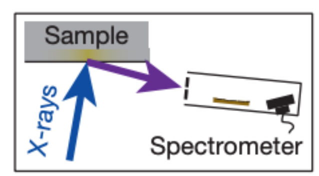
Going forwards

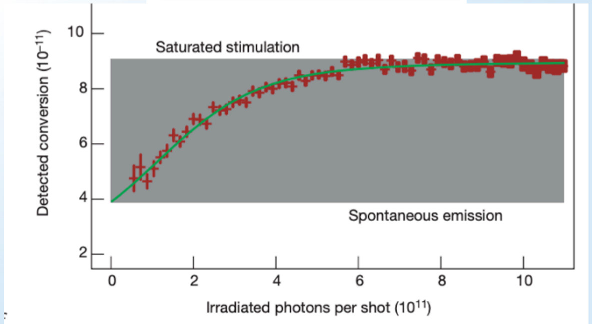
- New instruments
  - NEH 2.2 LCLS-II
  - hRIXS at XFEL
  - SwissFEL
  - UK XFEL
- New ideas
  - Photo-control of magnetic exchange
  - Quasi-correlated states
  - Anti-Stokes processes
  - Stimulated RIXS











Beye et al., Nature (2013)

Conclusions

- First glimpse of transient magnetic correlations in photodoped Sr_2IrO_4 , $Sr_3Ir_2O_7$ & their recovery

- Opportunities in for deeper understanding of magnetism in quantum materials out of equilibrium

M. P. M. Dean, et al. Nature Materials **15** 601–605 (2016)
Y. Cao et al., Phil. Trans. **377** 20170480 (2019)
D. Meyers et al., Sci Reports **9** 4263 (2019)
D. G. Mazzone et al., "Trapped Transient Magnons in the Gapped Antiferromagnet $Sr_3Ir_2O_7$", submitted (2019)