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U.S. DEPARTMENT OF ENERGY

Resonant elastic and inelastic scattering

Mark P. M. Dean
Brookhaven National Laboratory

July 18th, 2022

National School on Neutron and X-Ray Scattering

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Outline

Resonant x-ray scattering (RXS)

- What is it?
- Why should you care?
- X-ray resonances
- Some theory

Instrumentation

- Hard vs. soft x-rays
- Elastic vs. inelastic

Examples

- Kitaev magnetism in $RuCl_3$
- Oxygen states in superconducting nickelates

Extended versions

- Imaging
- Ultrafast

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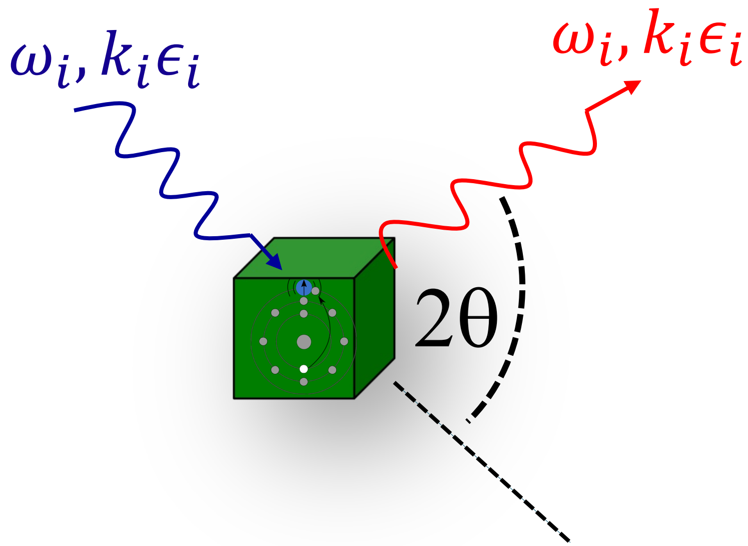
What is resonant scattering?

What is resonant x-ray scattering in one sentence?



$$Q = k_f - k_i = \frac{4\pi}{\lambda}\sin(\theta)$$
$$\omega = \omega_f - \omega_i$$

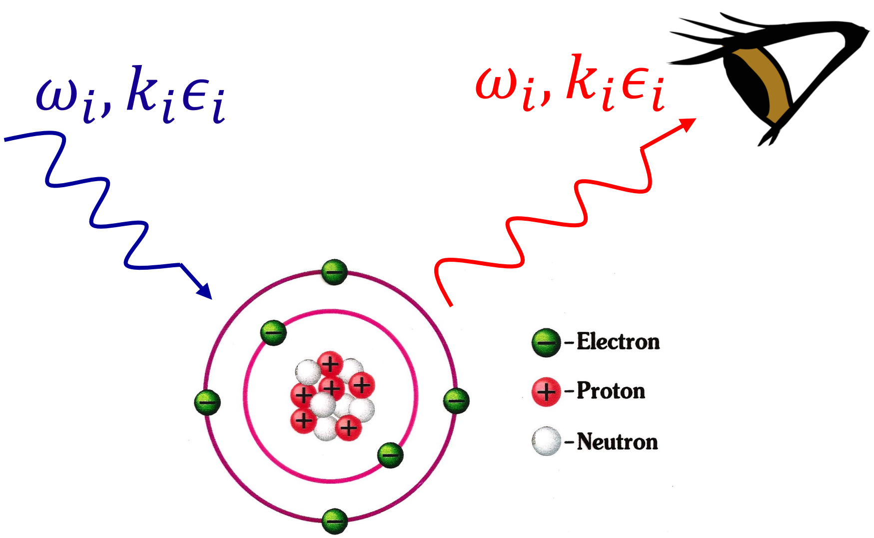
Family of photon-in photon-out techniques that use x-ray resonances to access

(i) Element & valence charge specificity or

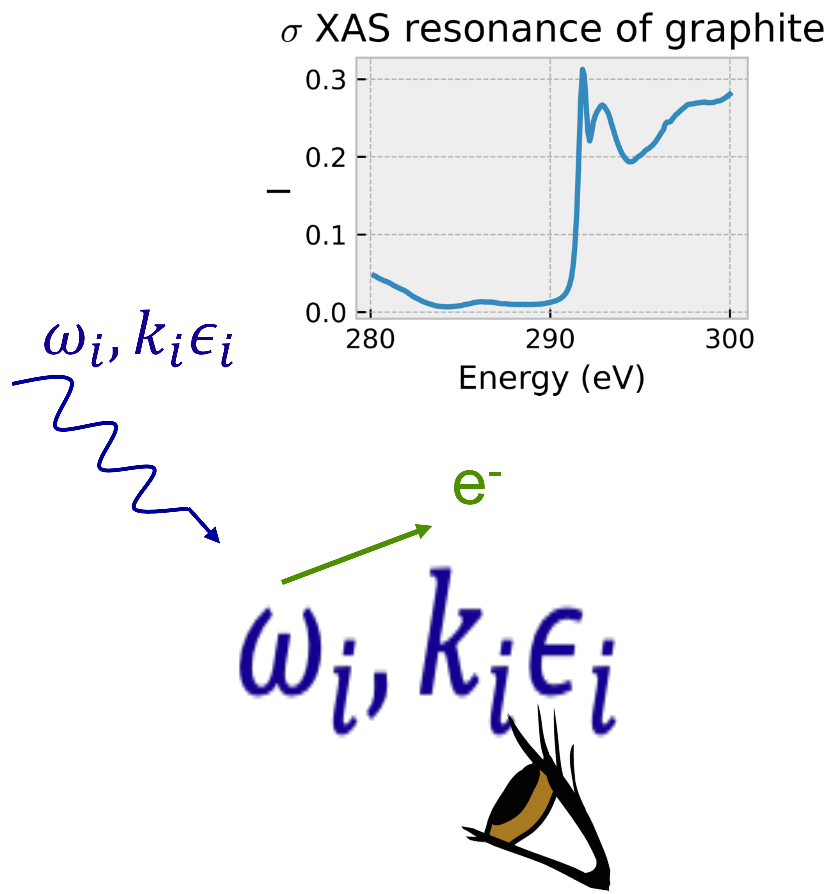
(ii) Electronic information: Magnetism, valence charge, electronic orbitals, etc.

Reminder of x-ray scattering & x-ray absorption

Standard (i.e. non-resonant) X-ray scattering



X-ray absorption



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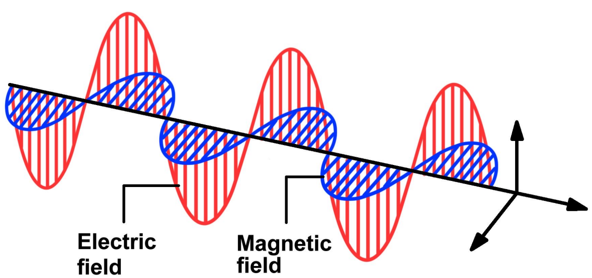
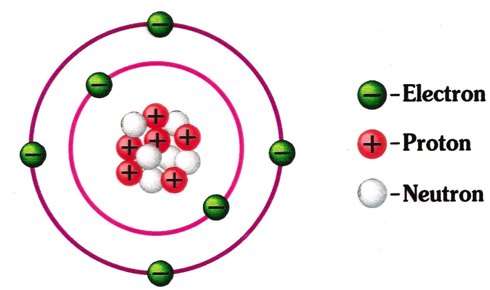
X-ray-matter interaction for non-resonant x-ray scattering

Classical wave cartoon of x-ray beam

Classical cartoon of atom



Thompson interaction



X-ray scatter from all electrons and see atomic positions

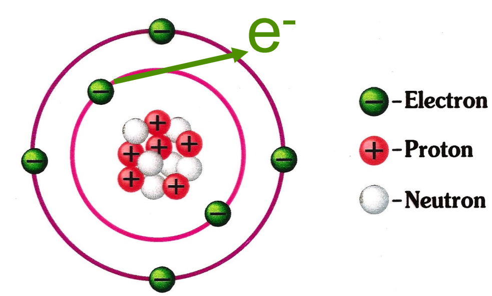
X-ray-matter interaction for x-ray absorption

Particle cartoon of x-ray beam

Classical cartoon of atom

photon

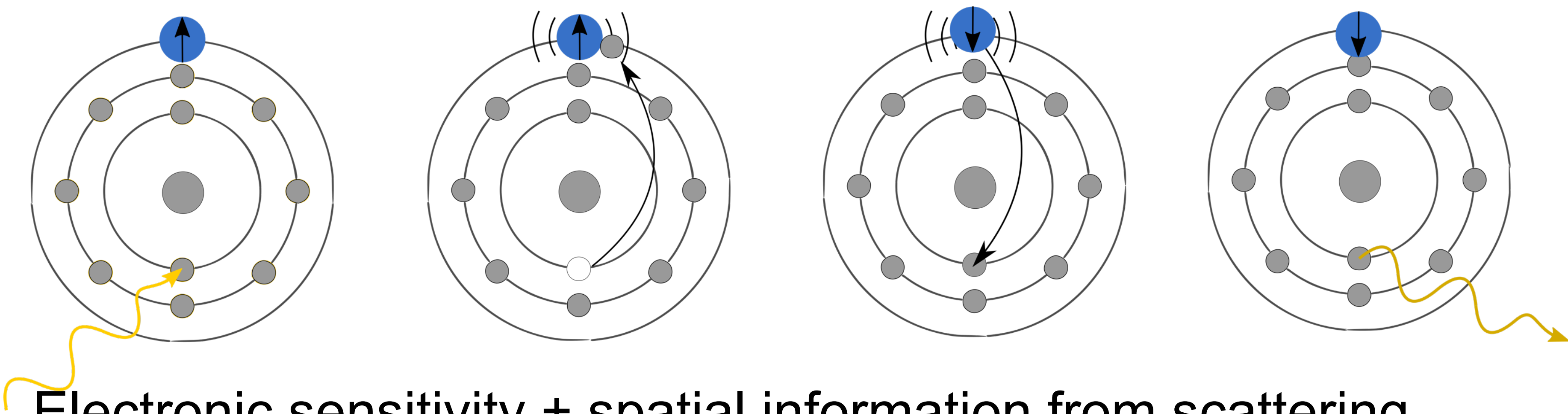
X-ray absorption



Observe transitions of electrons from core states to valence states

- Sensitive to magnetism
- Losses the information you had in scattering

Resonant x-ray scattering



Electronic sensitivity + spatial information from scattering

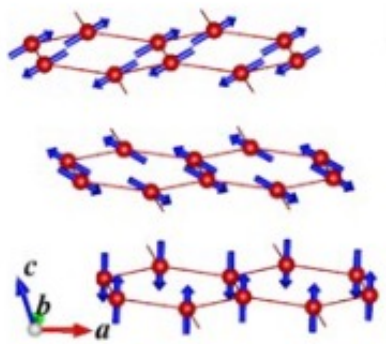
$$\frac{d^2\sigma}{d\Omega d\omega} \text{ or } \frac{d\sigma}{d\Omega}$$

N.b. Coherent quantum mechanical process

Why should you care about resonant scattering?

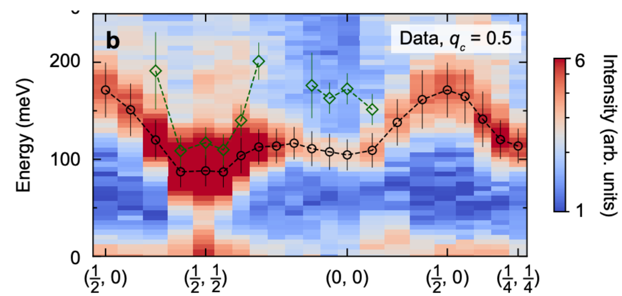
Resonant x-rays

Magnetic order



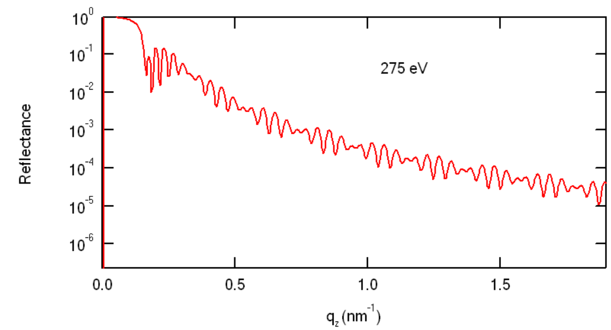
J. Sears et al., Nature Physics (2020)

Magnetic interactions



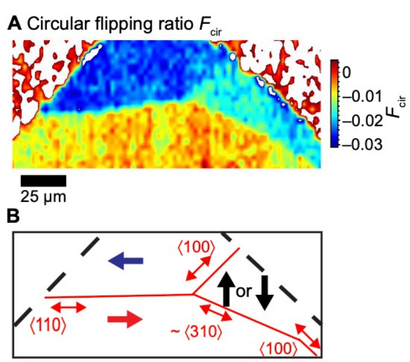
D. Mazzone et al., Nature Comm. (2022)

Orbital contrast



C. Wang et al., APL (2005)

Magnetic Imaging



P. Evans et al., Sci. Advances (2022)

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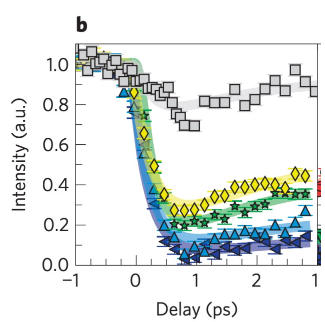
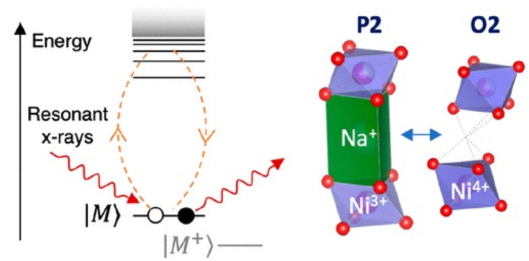
Ultrafast magnetism



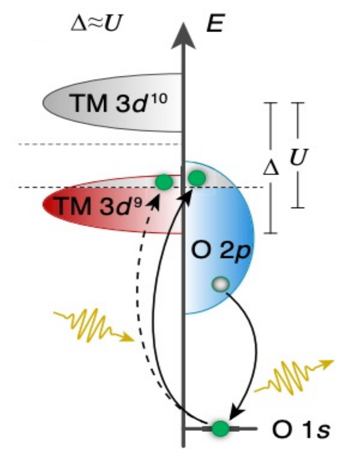
M. Dean et al., Nature Materials (2016)

Operando



D. Weinstock et al., ACS Energy (2022)

Electronic states



Y. Shen et al., PRX (2022)

Pros and cons

Pros

- Highly selective: species orbital
- Versatile access to many degrees of freedom: charge, spin, lattice, orbital
- Applicable in extreme situations: small volumes, in-operando, ultrafast
- Extendable to imaging

Cons

- Challenging experiments
- Complex cross section

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X-ray resonances

Energy levels of hydrogen (1)

Balmer series of emission lines from hydrogen

$$\frac{1}{\lambda} = R\left(\frac{1}{n_1^2} - \frac{1}{n_1^2}\right). \qquad n = 1,2,3,\ldots, n_2 > n_1$$

Discovery predates quantum mechanics

Energy levels of hydrogen (2)

$$\psi_{nlm}(r,\theta,\phi) = R_{nl}(r)Y_{lm}(\theta,\phi)$$

$n = 1,2,\ldots.$ $\quad l = 0,1,\ldots,n-1$ $\quad m = -l,-l+1,\ldots,l-1,l$

n : Principal quantum number

l : Angular momentum quantum number

m: Magnetic quantum number

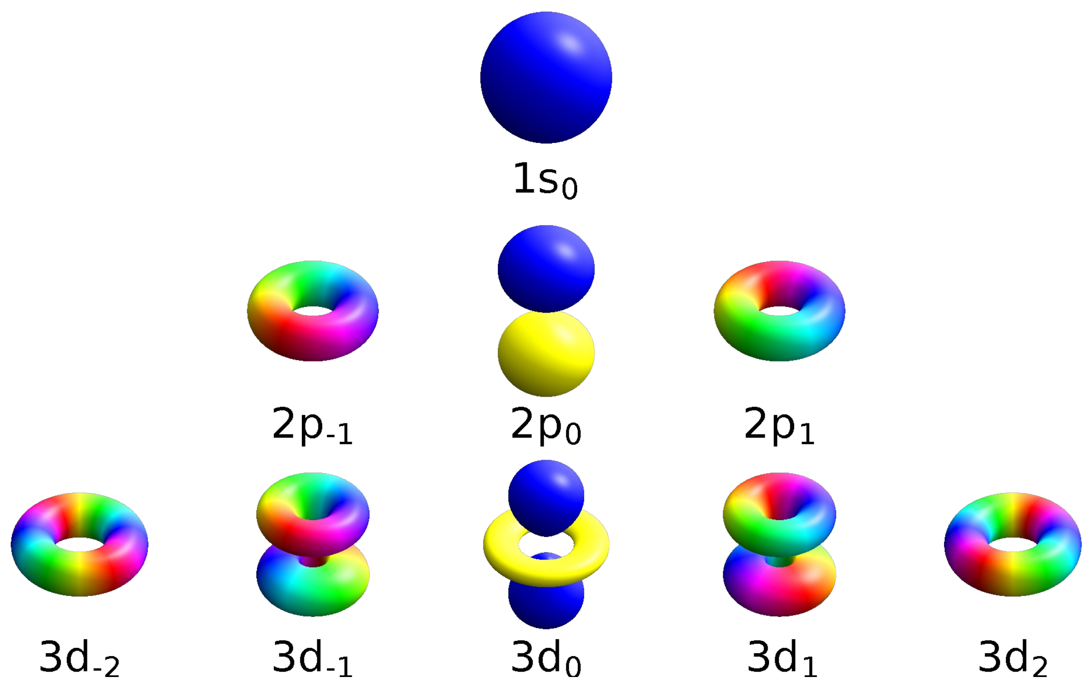
$R_{nl}(r) \sim \frac{1}{n^2}$

$l = 0$ s

$l = 1$ p

$l = 2$ d

$l = 3$ f



Onto x-ray resonances

Spin orbit coupling

$$J = L + S$$

$|L - S|, |L - S|$+1, …, $|L + S|$

Core hole notation

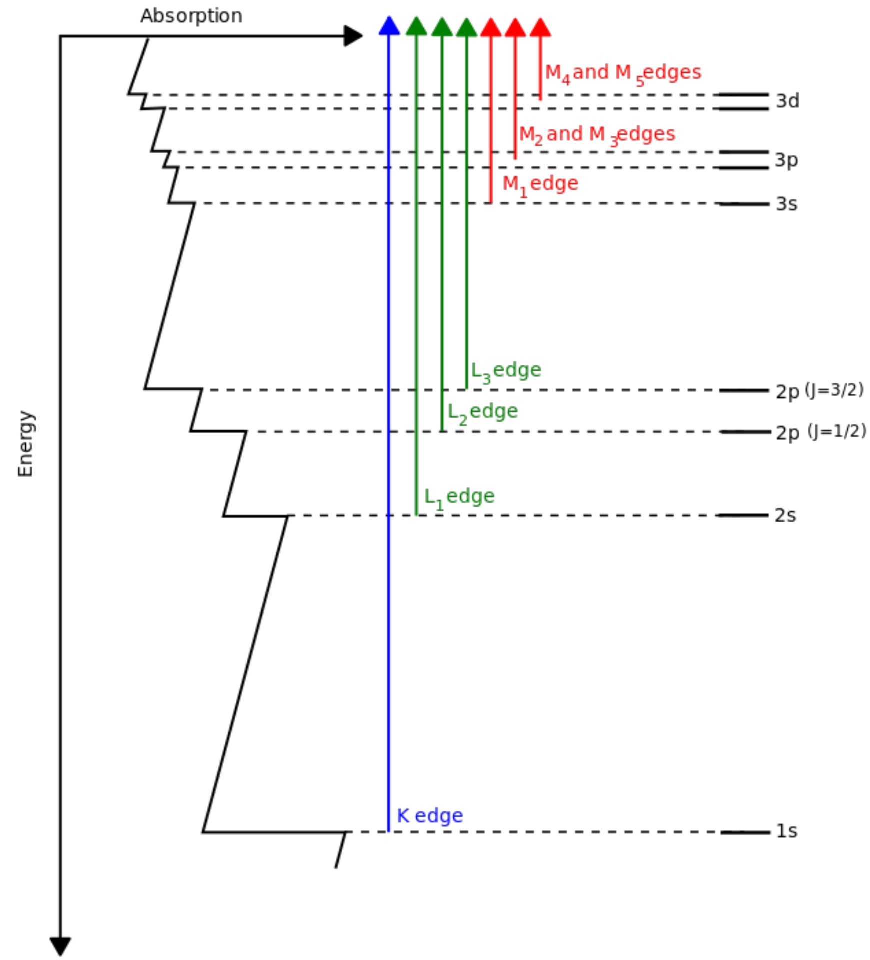
$$2p_{3/2}$$

X-ray spectroscopic notation

$$\mathrm{M}_4$$

n=1, 2, 3 as K, L, M, …

Index low L to high L, then low J to high J



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Types of transition

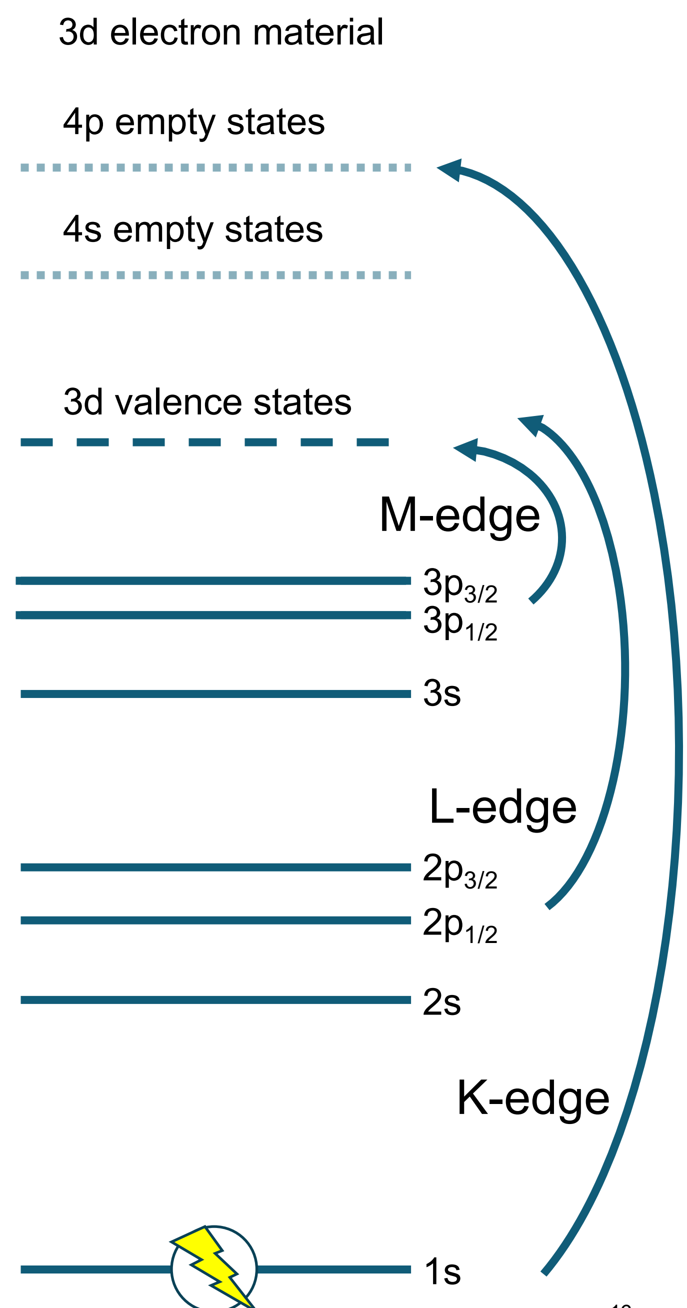
Dipole $\Delta L = \pm 1$ called E1

Quadrupole $\Delta L = \pm 2$ called E2

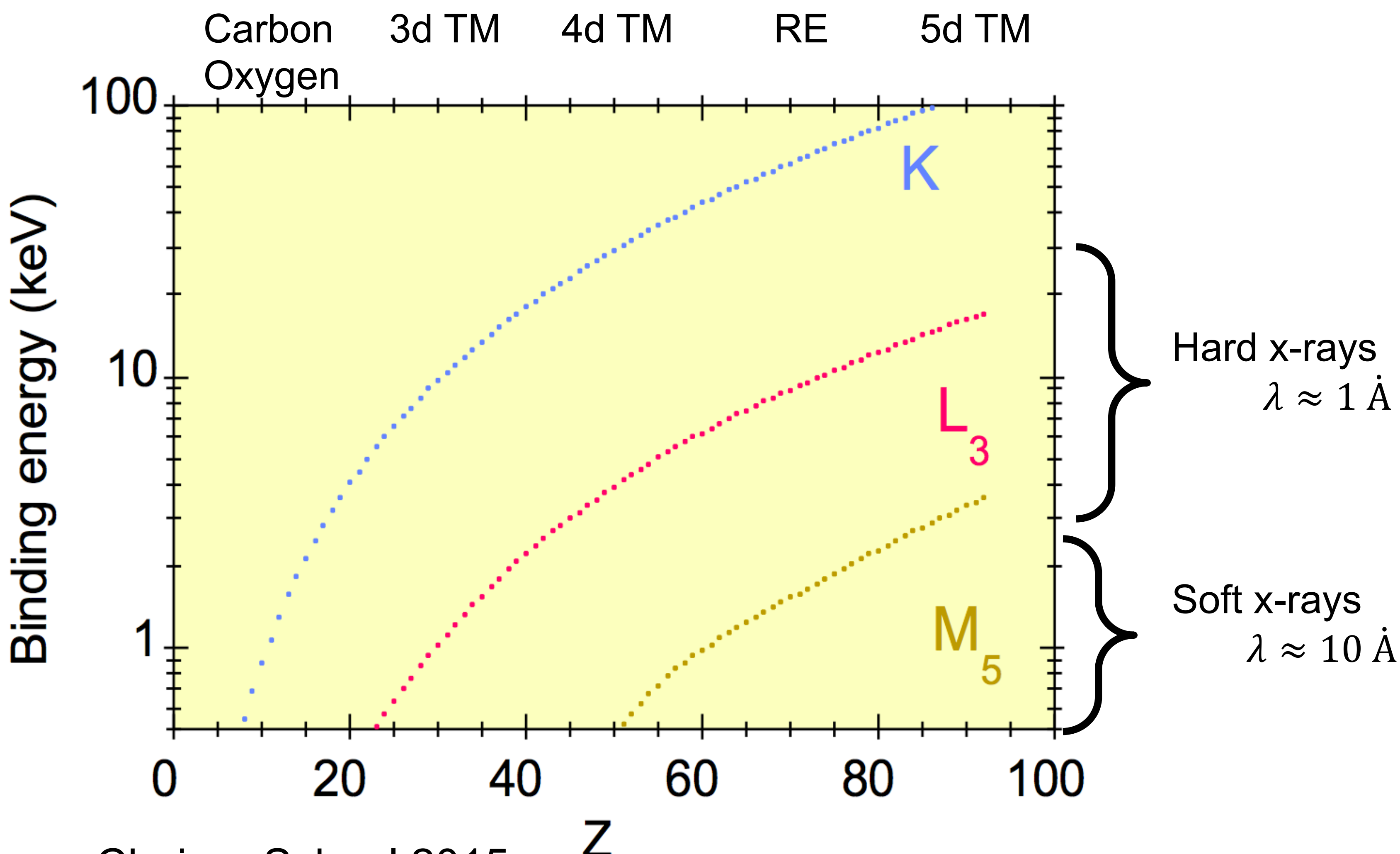
Etc.

Direct or "operator" RIXS

Indirect or "shakeup" RIXS



Energies of different resonances



Fons, Cheiron School 2015

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A few words for the theorists

Which parts of the x-ray-matter interactions are involved here?

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A theorist's perspective (1) Photon matter interactions

$$H_0 = H_\mathrm{e} + H_\mathrm{rad}$$

$$\boldsymbol{p} \to \boldsymbol{p} + eA \qquad E = -\frac{\partial A}{\partial t}$$

$$H = \frac{(\boldsymbol{p} + e\boldsymbol{A})^2}{2m} + H_\mathrm{rad}$$

$$H = \frac{p^2}{2m} + \frac{e\boldsymbol{A}\cdot\boldsymbol{p}}{m} + \frac{e^2A^2}{2m} + H_\mathrm{rad}$$

$$H_\mathrm{int} = \frac{e\boldsymbol{A}\cdot\boldsymbol{p}}{m} + \frac{e^2A^2}{2m}$$

absorption scattering

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Ament et al., Rev. Mod. Phys. 83, 705 (2011)

A theorist's perspective (2) Kramers-Heisenberg equation

$$\frac{d^2\sigma}{d\Omega \mathrm{d}\omega} \propto \left| \langle f|H_{\mathrm{int}}|i\rangle + \sum_n \frac{\langle f|H_{\mathrm{int}}|n\rangle\langle n|H_{\mathrm{int}}|i\rangle}{E_i - E_n + \hbar\omega_i + i\Gamma} \right|^2$$

1st order | 2nd order

$$H_{\mathrm{int}} = \frac{e\boldsymbol{A}\cdot\boldsymbol{p}}{m} + \frac{e^2A^2}{2m}$$

absorption | scattering

1 photon | 2 photon

Ament et al., Rev. Mod. Phys. 83, 705 (2011)

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Connection between scattering and absorption (I)

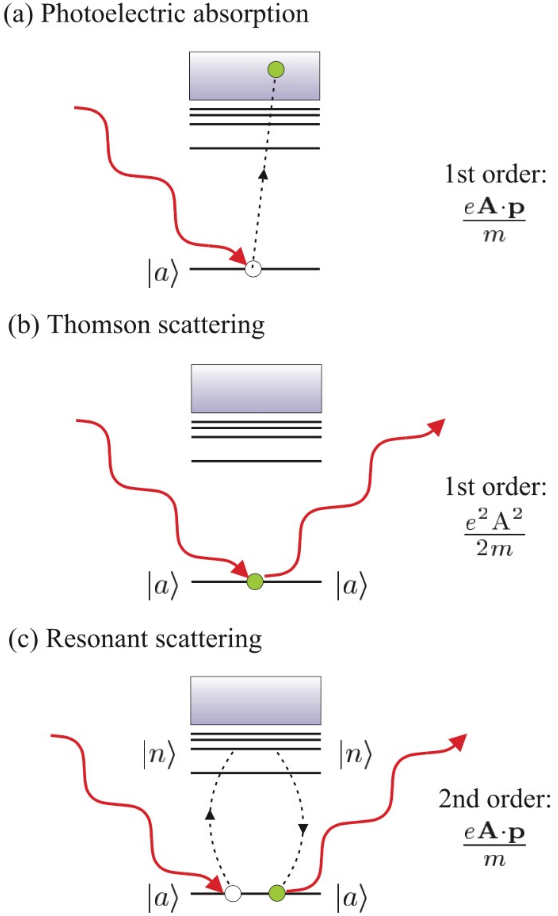
$$\frac{d^2\sigma}{d\Omega d\omega} \propto \left| \langle f|H_{\text{int}}|i\rangle + \sum_n \frac{\langle f|H_{\text{int}}|n\rangle\langle n|H_{\text{int}}|i\rangle}{E_i - E_n + \hbar\omega_i + i\Gamma} \right|^2$$

$$H_{\text{int}} = \frac{e\boldsymbol{A}\cdot\boldsymbol{p}}{m} + \frac{e^2A^2}{2m}$$

Resonant elastic scattering

$$\frac{d\sigma}{d\Omega} \propto \left| \langle i|H_{\text{int}}|i\rangle + \sum_n \frac{\langle i|H_{\text{int}}|n\rangle\langle n|H_{\text{int}}|i\rangle}{E_i - E_n + \hbar\omega_i + i\Gamma} \right|^2$$

$$f(Q, E_i) = f^0(Q) + f'(E_i) + if''(Ei)$$



Elements of modern x-ray physics by Jens Als-Nielsen and Des McMorrow

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Connection between scattering and absorption (II)

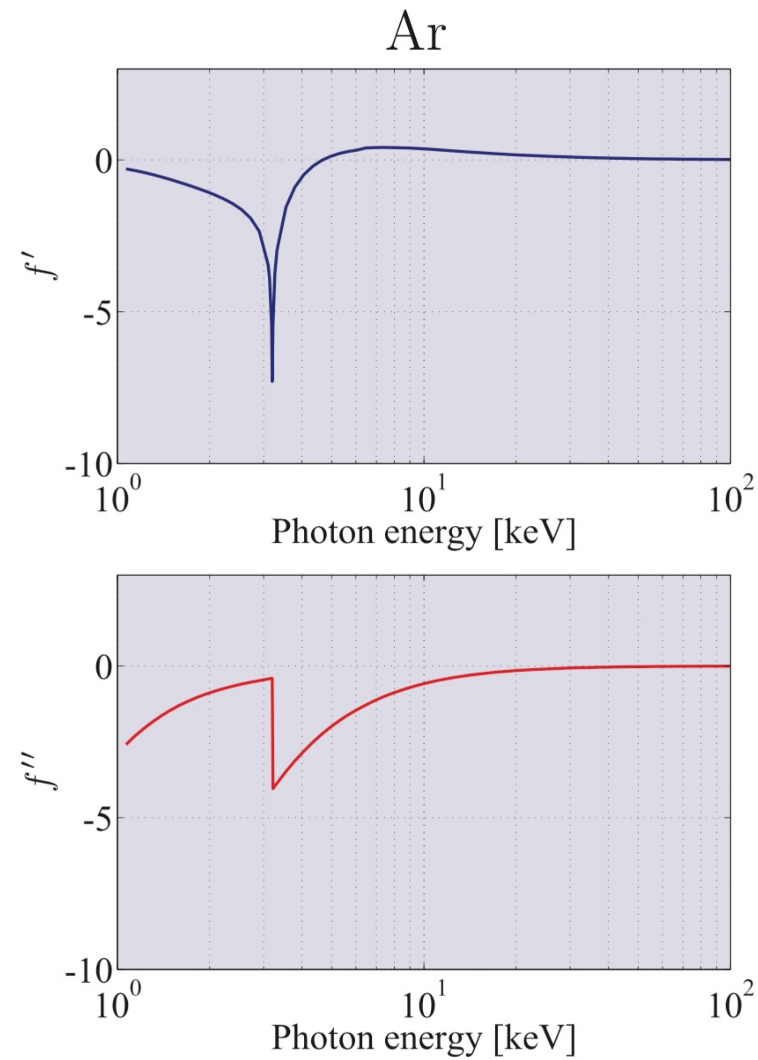
$$f(Q, E_i) = f^0(Q) + f'(E_i) + if''(E_i)$$

$f^0(Q)$ Thompson scattering from all electrons

$f'(E_i)$ Resonant scattering

$f''(E_i)$ Resonant absorption

f' and f'' are related to one-another



Elements of modern x-ray physics by Jens Als-Nielsen and Des McMorrow

Instrumentation

Hard x-rays vs. soft x-rays

Elastic vs. inelastic

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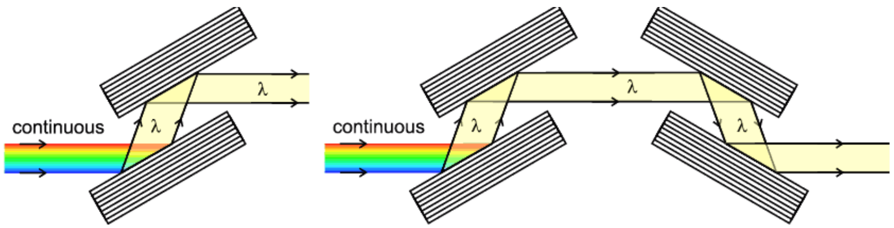
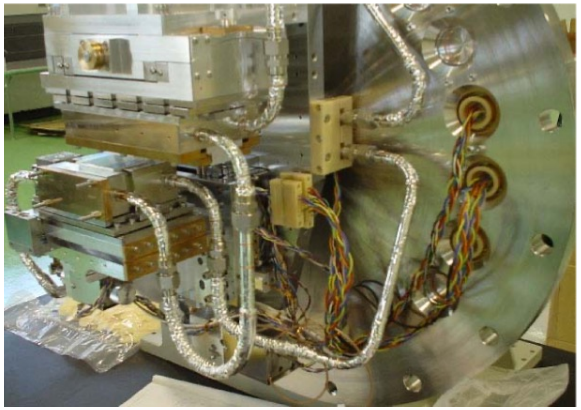
Soft x-ray vs. hard x-ray

Hard x-rays have $\lambda \sim d$ in Si

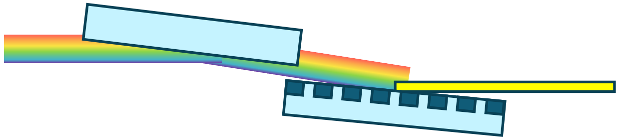
Bragg's law $n\lambda = 2d \sin\theta$

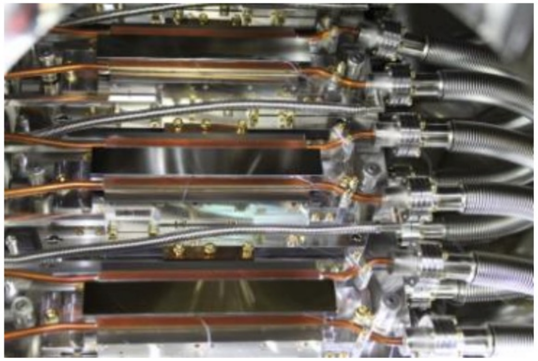




Soft x-rays have $\lambda \gg d$ in suitable crystals

Grating equation $m\lambda = d_0(\sin\alpha + \sin\beta)$



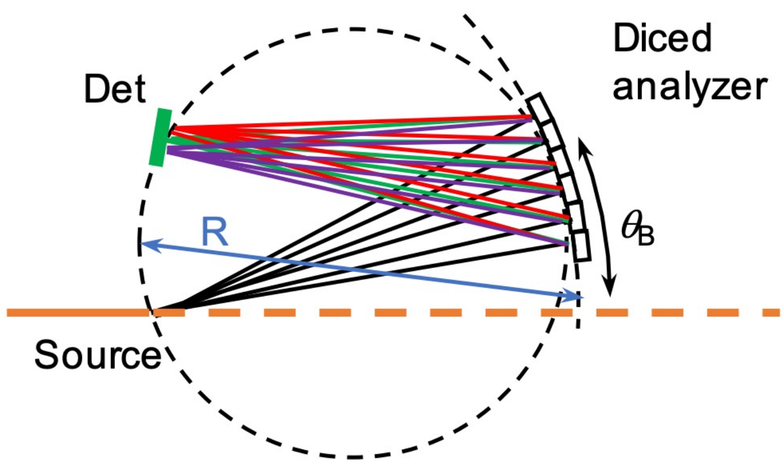


Images: T. Matsushita, Photon Factory

Inelastic scattering (I) hard x-rays

Key issue: angular acceptance

Diced crystal analyzer



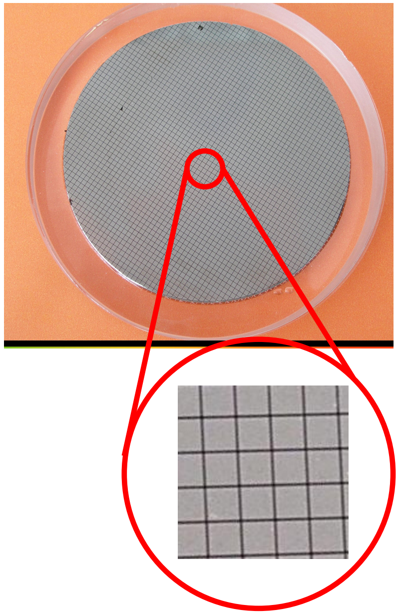


Image: Y. Cao et al., Philos. Trans. R. Soc. A 377, 20170480 (2019)

Inelastic scattering (II) soft x-rays

Spherical variable line spacing grating

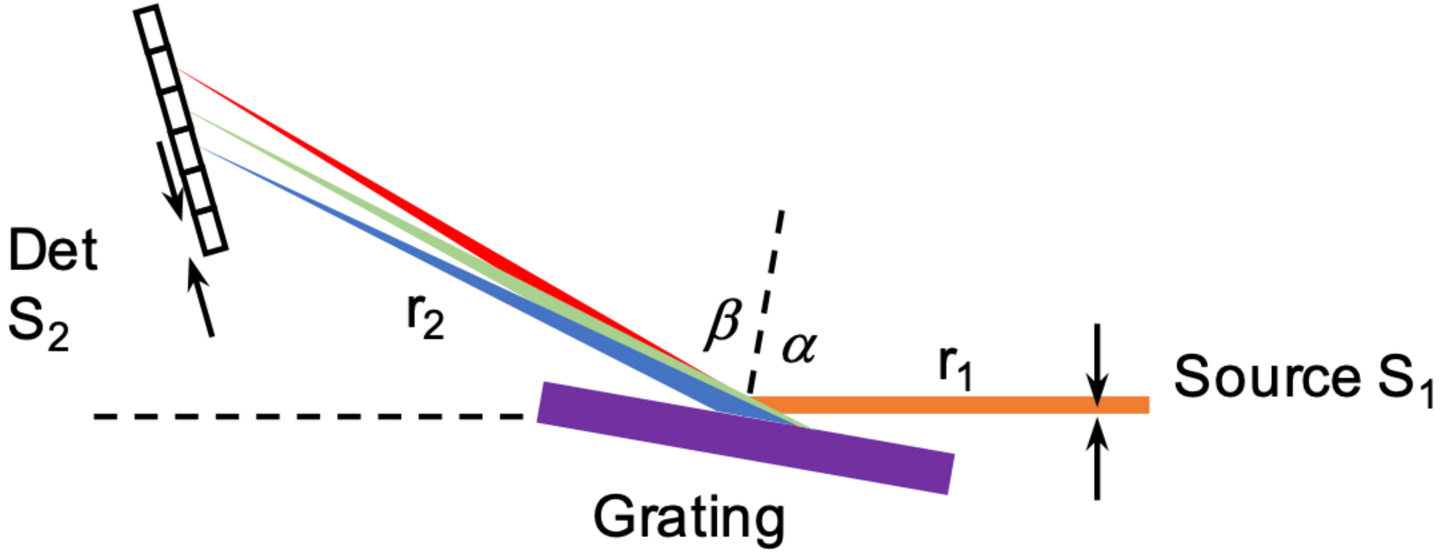


Image: Y. Cao et al., Philos. Trans. R. Soc. A 377, 20170480 (2019)

ISR NSLS-II

Hard x-ray REXS

Some other beamlines

P09 DESY
Sector 6IDB APS
I16 DLS

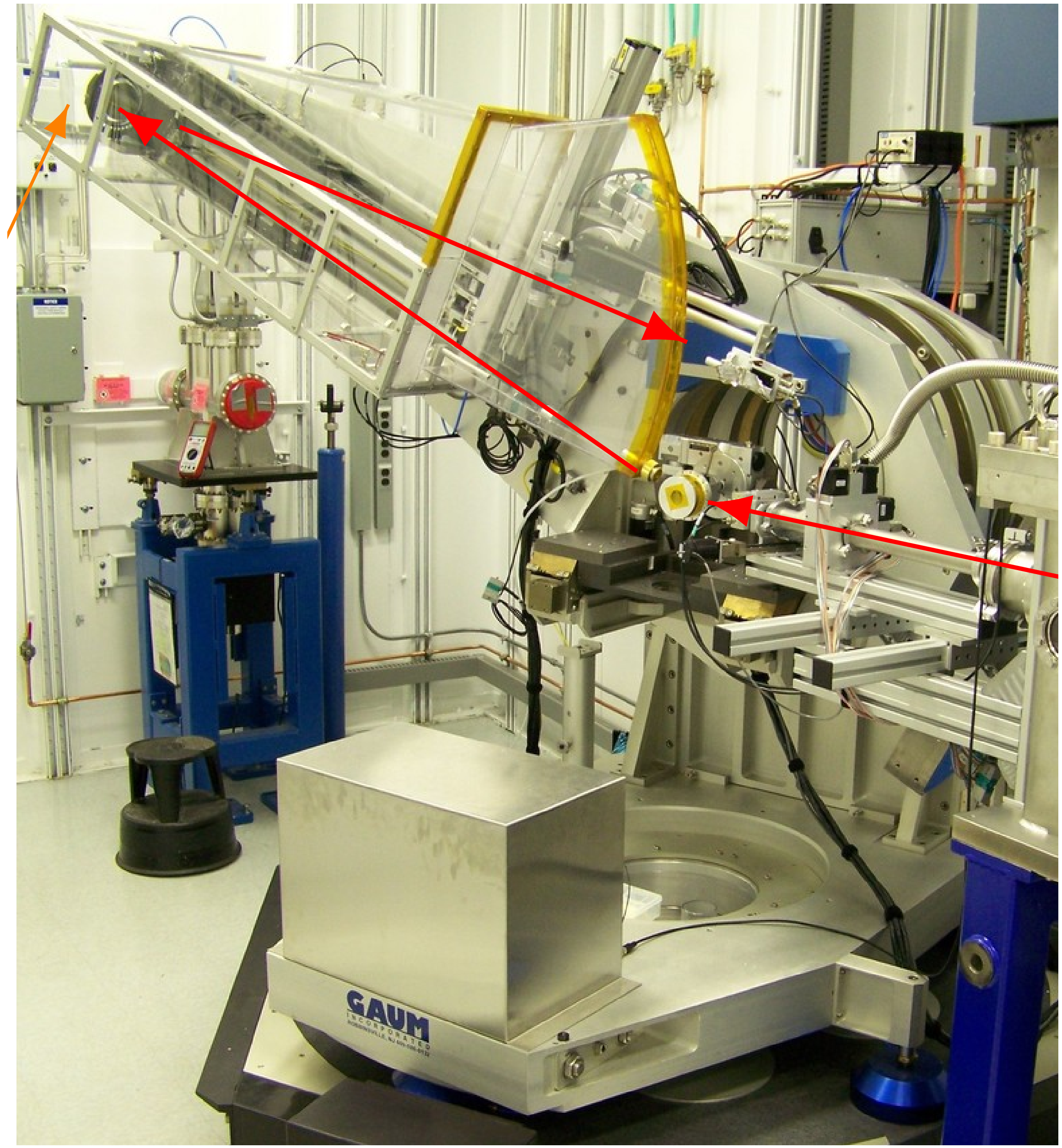
Example 1 was done with P09

Hard x-ray RIXS

Some other beamlines

ID20 ESRF

Sector 27 APS



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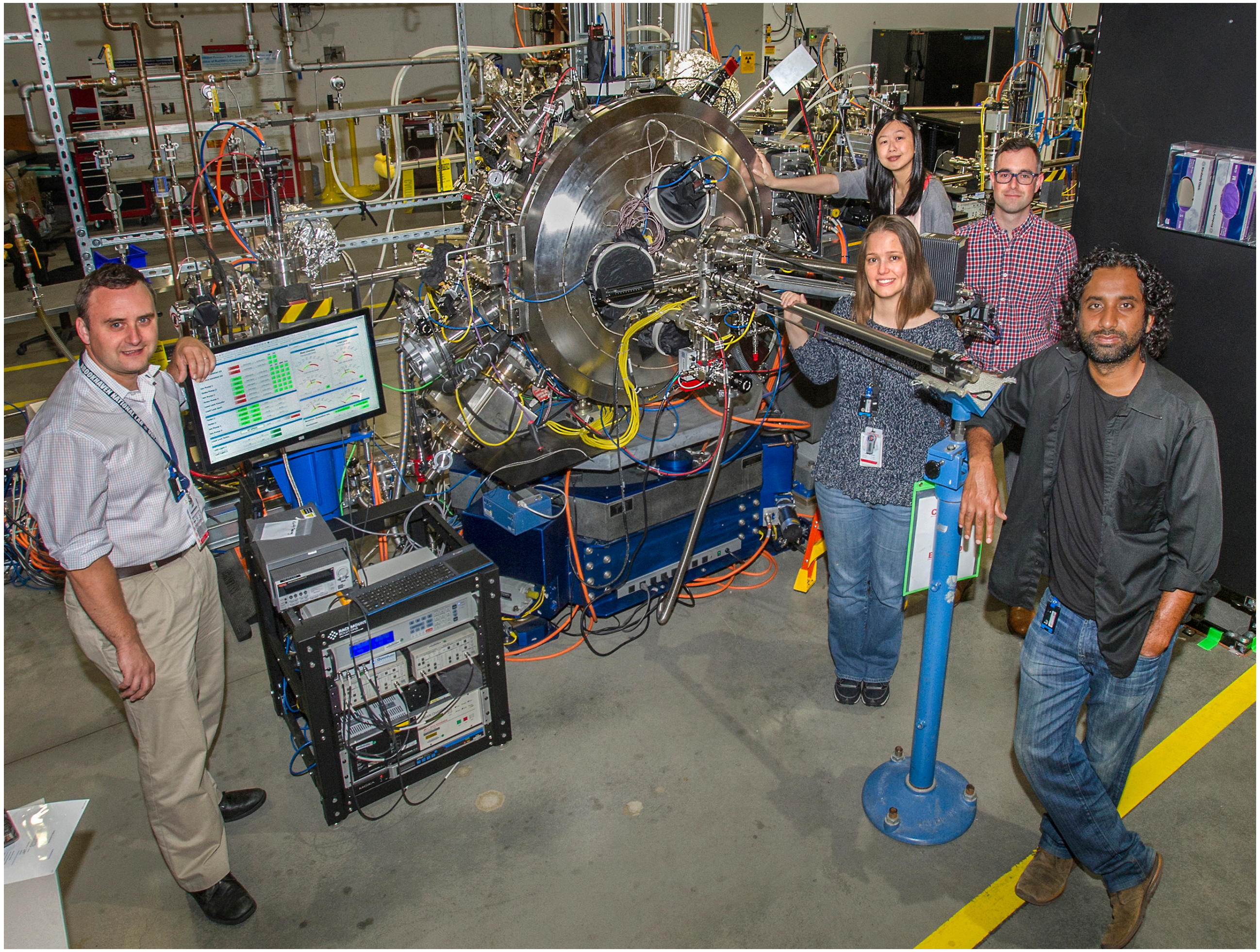
Soft x-ray REXS

CSX NSLS-II

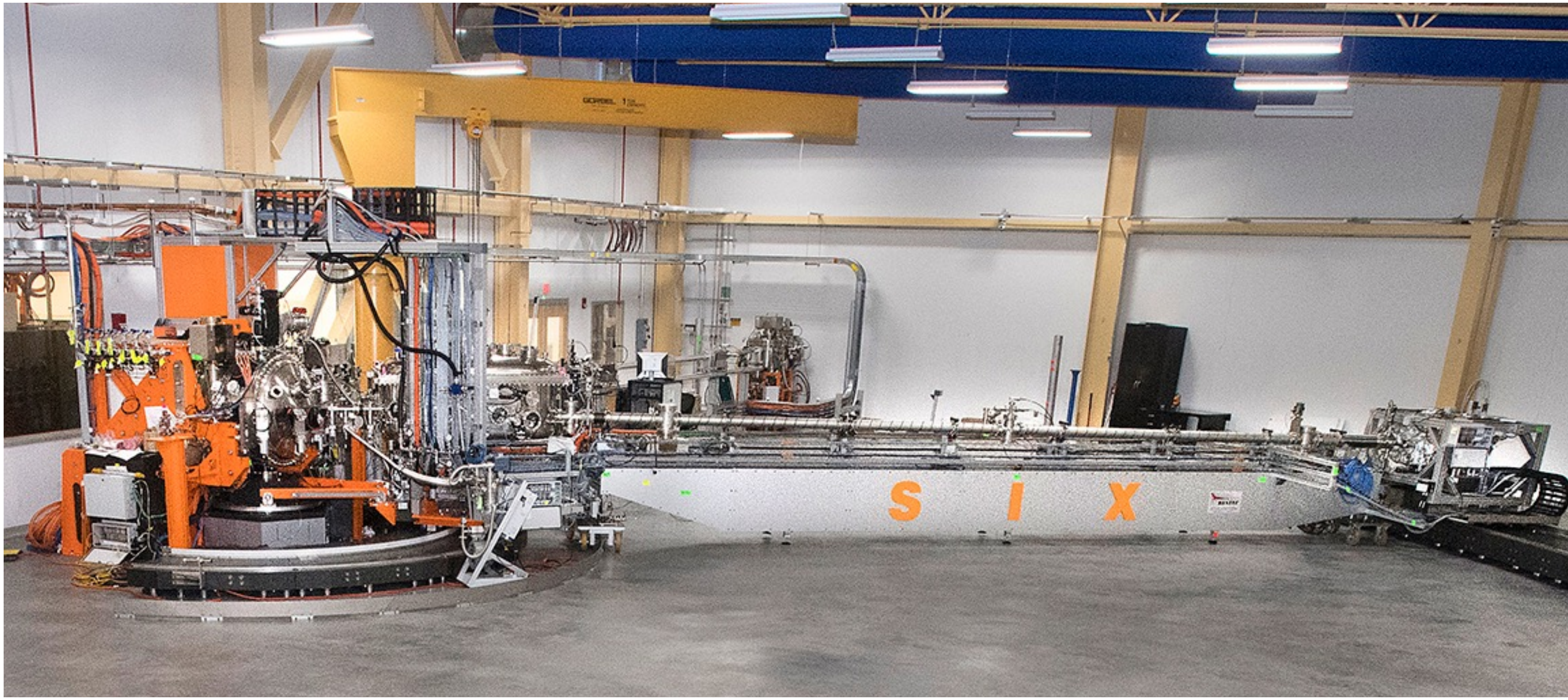


Some other beamlines

REIXS CLS
Sector 29 APS
BL 11 ALS
I10 DLS

Soft x-ray RIXS

SIX NSLS-II



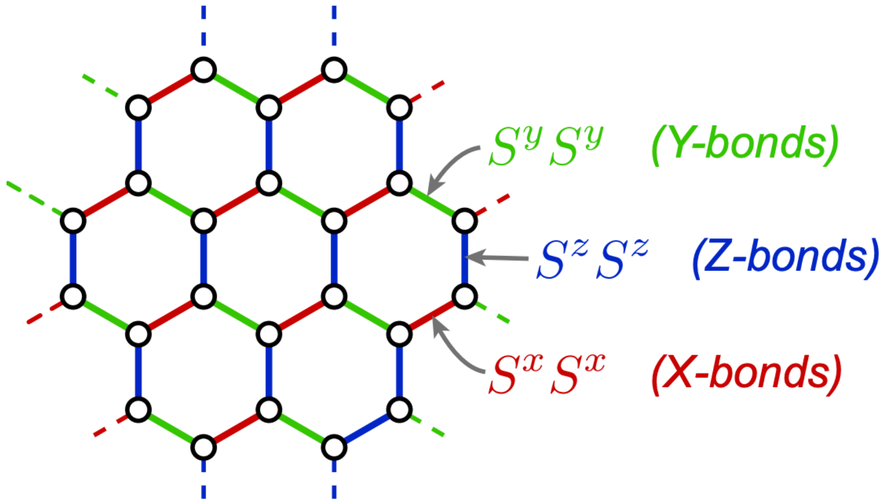
Some other beamlines: I12 DLS, ID32 ESRF

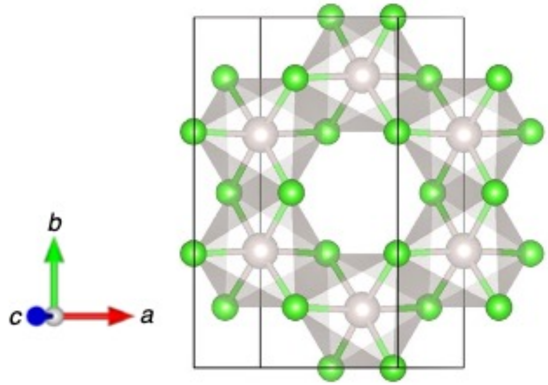
Example 1: REXS
Spin direction in proximate Kitaev magnet

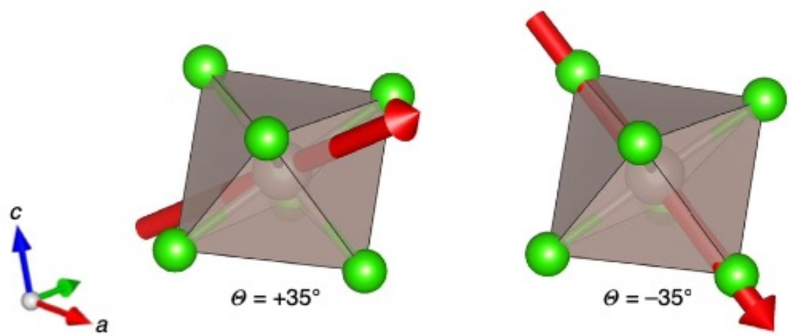
Kitaev magnetism in $RuCl_3$

Quantum spin liquid with Majorana Fermions

$$\mathcal{H} = \sum_{\langle ij \rangle} S_i^\gamma S_j^\gamma + \cdots$$







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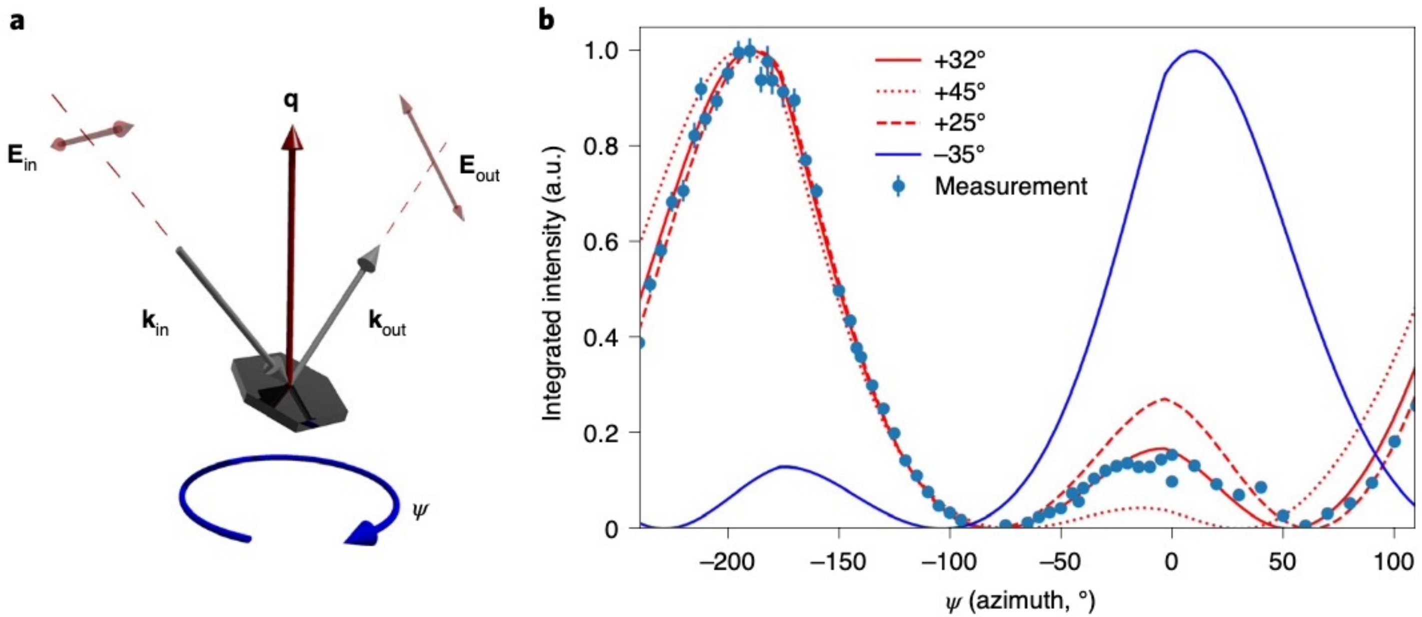
J. Sears et al. Nature Physics 16, 837–840 (2020)

Azimuthal scan

Ru L-edge 2.8 keV. $\frac{d^2\sigma}{d\Omega \mathrm{d}\omega} \propto |\epsilon_i \times \epsilon_f \cdot M|^2$



J. Sears et al. Nature Physics 16, 837–840 (2020)

Example 2: RIXS
Oxygen states in superconducting nickelates

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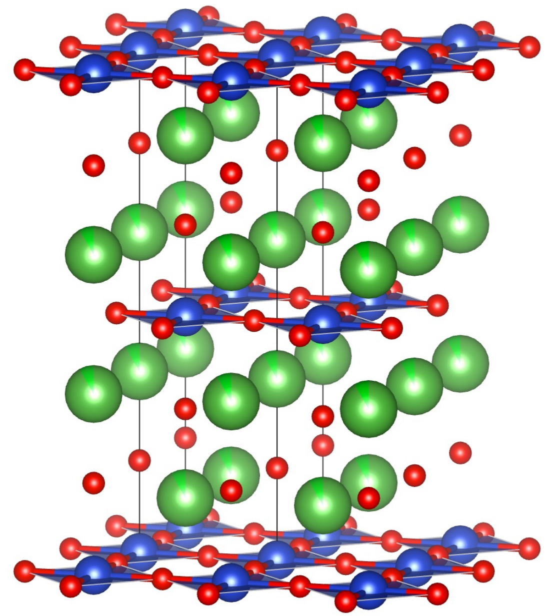
Superconducting nickelates

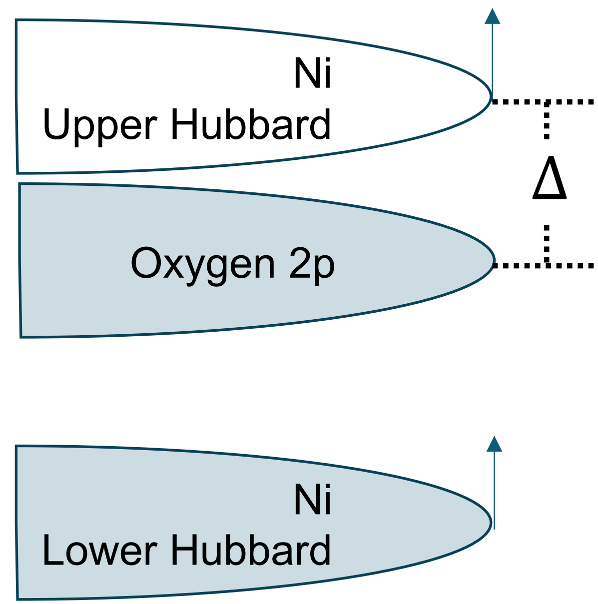
d^9

Possible analogs of cuprate superconductors

Role of oxygen states

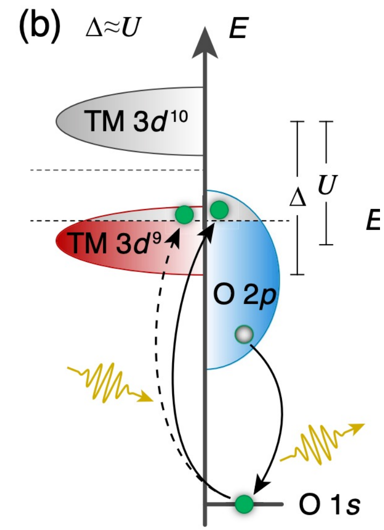
Strength of magnetism



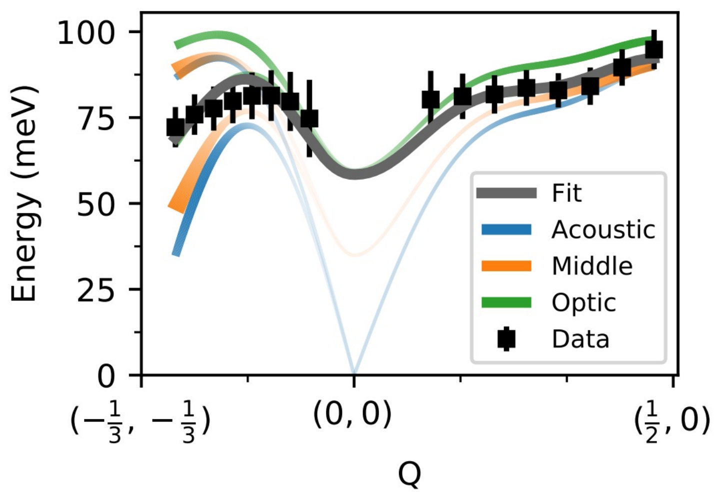


D. Li et al., Nature 572, 624–627 (2019)
G. Pan et al., Nature Materials 21, 160–164 (2022)

RIXS

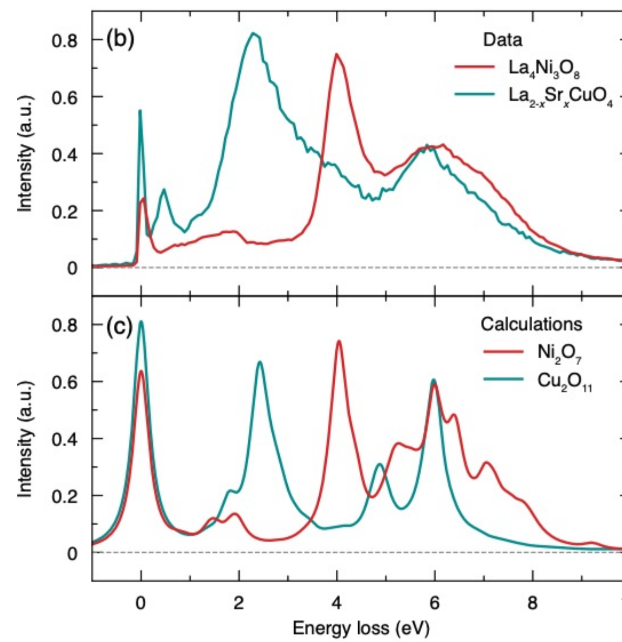


Ni L-edge RIXS



Relatively strong magnetic interactions

O K-edge RIXS



Substantial oxygen involvement

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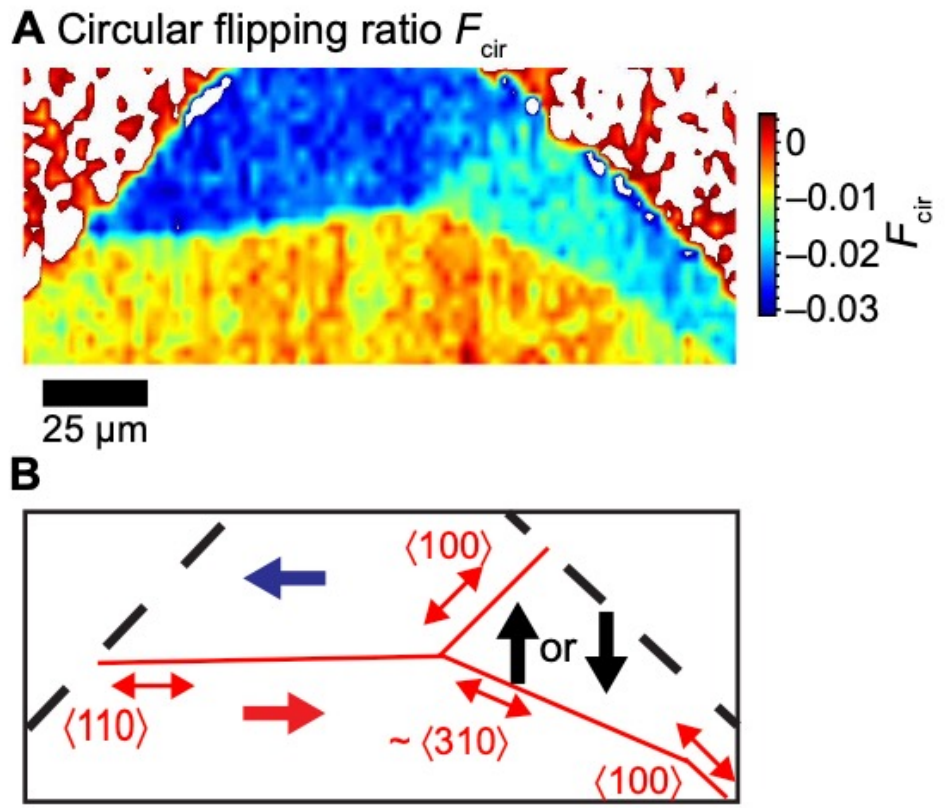
J. Li et al., Phys. Rev. Lett. 126, 087001 (2021)
Y. Shen et al., Phys. Rev. X 12, 011055 (2022)

Extended versions of resonant scattering

REXS imaging

Nanobeam magnetic diffraction imaging of $Gd_3Fe_5O_{12}$ at Gd L_2 edge



P. Evans et al., Sci. Advances 6 eaba9351 (2022)

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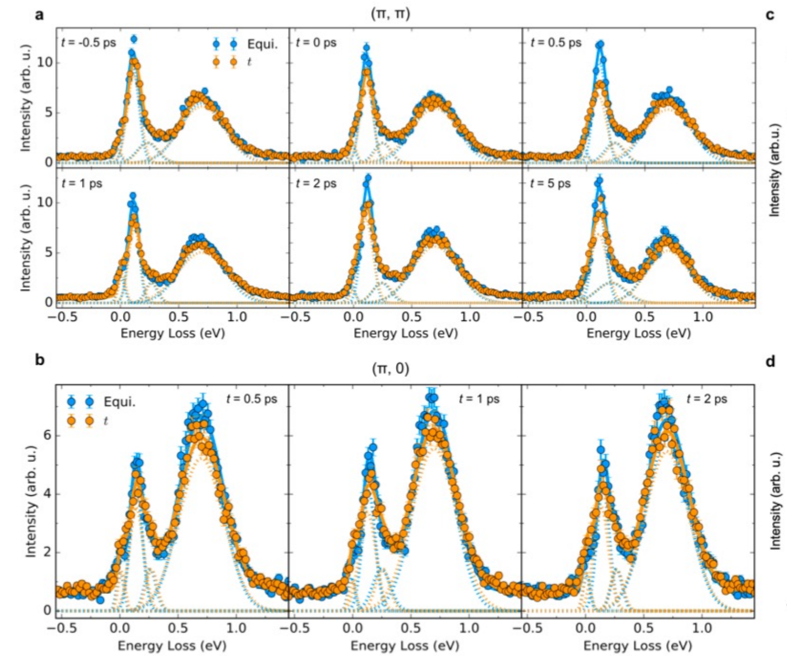
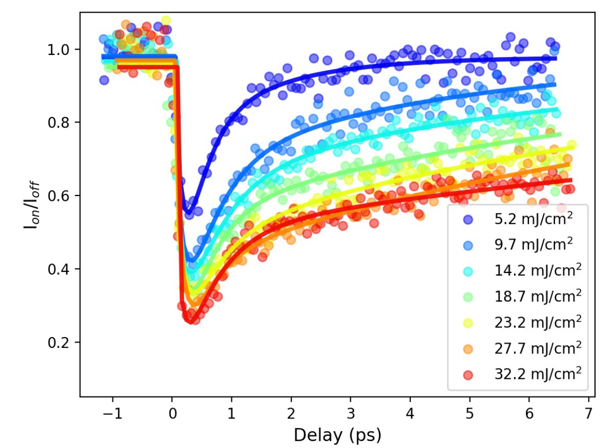
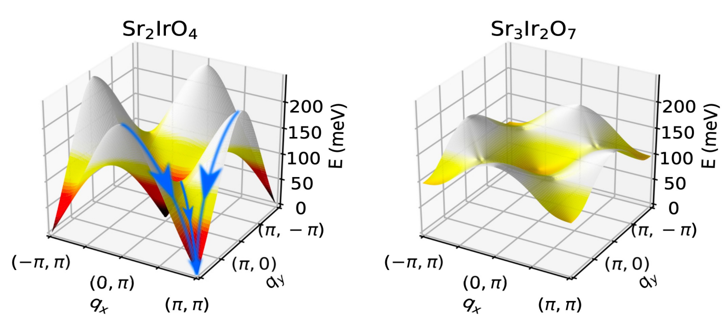
Ultrafast study of magnetism $Sr_3Ir_2O_7$

Magnetic state gets “stuck” in suppressed state

Spin-gap bottleneck?







D. Mazzone, D. Meyers, … MPMD et al., PNAS 15, 601–605 (2021)

Conclusions

Resonant scattering uses x-ray core hole states to access important information that is inaccessible to regular-ray scattering

The technique is readily extensible and can be performed in imaging, time-resolved, and ultrafast modes

Feedback

Lecture – 9:45 – 10:45
Resonant elastic and inelastic scattering - Mark Dean
https://forms.office.com/g/QR5WYreeUh