Inelastic X-Ray Scattering

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Inelastic x-ray scattering – technical

$$H_0(t) = \int d\mathbf{x}^3\, \hat{\psi}^\dagger(\mathbf{x},t) \left[\frac{\mathbf{p}^2}{2m} + V(\mathbf{x})\right] \hat{\psi}(\mathbf{x},t)$$

$$\mathbf{p} \rightarrow \mathbf{p}_c - \frac{e}{c}\mathbf{A}$$

(Lorentz force law)

$$\hat{\mathbf{A}}(\mathbf{x},t) = \sum_{k,\lambda} c\sqrt{\frac{\hbar}{2\omega_k}} \left[a_{k,\lambda}\, \epsilon_\lambda\, e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)} + a^\dagger_{k,\lambda}\, \epsilon^*_\lambda\, e^{-i(\mathbf{k}\cdot\mathbf{r}-\omega t)}\right]$$

$$H = H_0 + H_1 + H_2$$

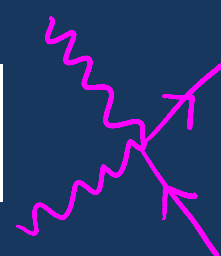
$$H_2^I(t) = -\frac{e^2}{2mc^2}\int d\mathbf{x}^3\, \hat{\psi}^\dagger(\mathbf{x},t)\ \hat{\mathbf{A}}^2(\mathbf{x},t)\ \hat{\psi}(\mathbf{x},t) = -\frac{e^2}{mc^2}\int d\mathbf{x}^3\ \hat{\mathbf{A}}^2(\mathbf{x},t)\ \hat{n}(\mathbf{x},t)$$

$$H_1^I(t) = -\frac{e}{mc}\int d\mathbf{x}^3\, \hat{\psi}^\dagger(\mathbf{x},t) \left[\mathbf{p}\cdot\hat{\mathbf{A}}(\mathbf{x},t)\right] \hat{\psi}(\mathbf{x},t)$$

$$U_I(\infty,-\infty) = \exp\left[-i\int_{-\infty}^{\infty} dt\, H^I(t)\ e^{-\eta|t|}\right]$$

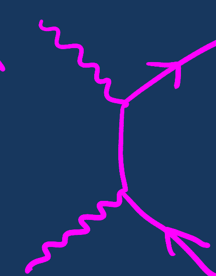
Nonresonant inelastic x-ray scattering

$$w_{f\leftarrow i} = r_0^2(\epsilon_f^*\cdot\epsilon_i)^2 \sum_{n,m} |< n|\hat{n}(\mathbf{k})|m >|^2\, P_m\, \delta(\omega - \omega_n + \omega_m)$$



Resonant inelastic x-ray scattering (RIXS)

$$w_{f\leftarrow i} = \left|\frac{e^2}{mc^2\hbar^2}\sum_m \frac{< f|\mathbf{p}\cdot\mathbf{A}|m >< m|\mathbf{p}\cdot\mathbf{A}|0 >}{\omega - \omega_m + i\gamma}\right|^2 \delta(\omega - \omega_f + \omega_0)$$



Nonresonant IXS

"Nonresonant" inelastic x-ray scattering

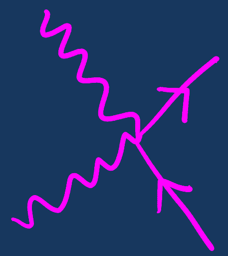
$$w_{f\leftarrow i} = r_0^2(\epsilon_f^* \cdot \epsilon_i)^2 \sum_{n,m} |< n|\hat{n}(\mathbf{k})|m >|^2 \, P_m \, \delta(\omega - \omega_n + \omega_m)$$

$$P_m = \frac{e^{-\hbar\omega_m/kT}}{Z}$$

Dynamic structure factor:

$$S(k,\omega) = \sum_{n,m} |< n|\hat{n}(\mathbf{k})|m >|^2 \, P_m \, \delta(\omega - \omega_n + \omega_m)$$

$$\mathbf{k} = \mathbf{k}_f - \mathbf{k}_i \qquad \omega = \omega_f - \omega_i$$



S(k,ω) is the Fourier transform of the Van Hove density correlation function:

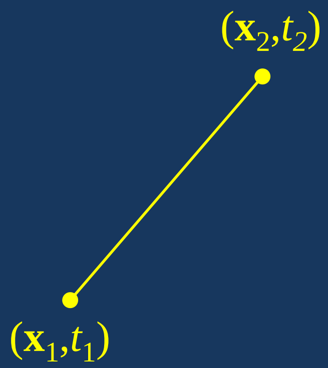
$$S(\mathbf{k},\omega) = \int d\mathbf{x}dt \; G(\mathbf{x},t)\, e^{-i(\mathbf{k}\cdot\mathbf{x}-\omega\mathbf{t})}$$

$$G(\mathbf{x},t) = \int d\mathbf{x}'dt' < \hat{n}(\mathbf{x},t)\, \hat{n}(\mathbf{x}+\mathbf{x}', t+t') >$$

X-ray "diffraction" actually measures an equal-time correlation function

$$\int S(\mathbf{k},\omega)\, d\omega = G(\mathbf{k},t)\Big|_{t=0} = \int d\mathbf{x}\, G(\mathbf{x},0)\, e^{-i\mathbf{k}\cdot\mathbf{x}}$$

Nonresonant IXS – dynamics!

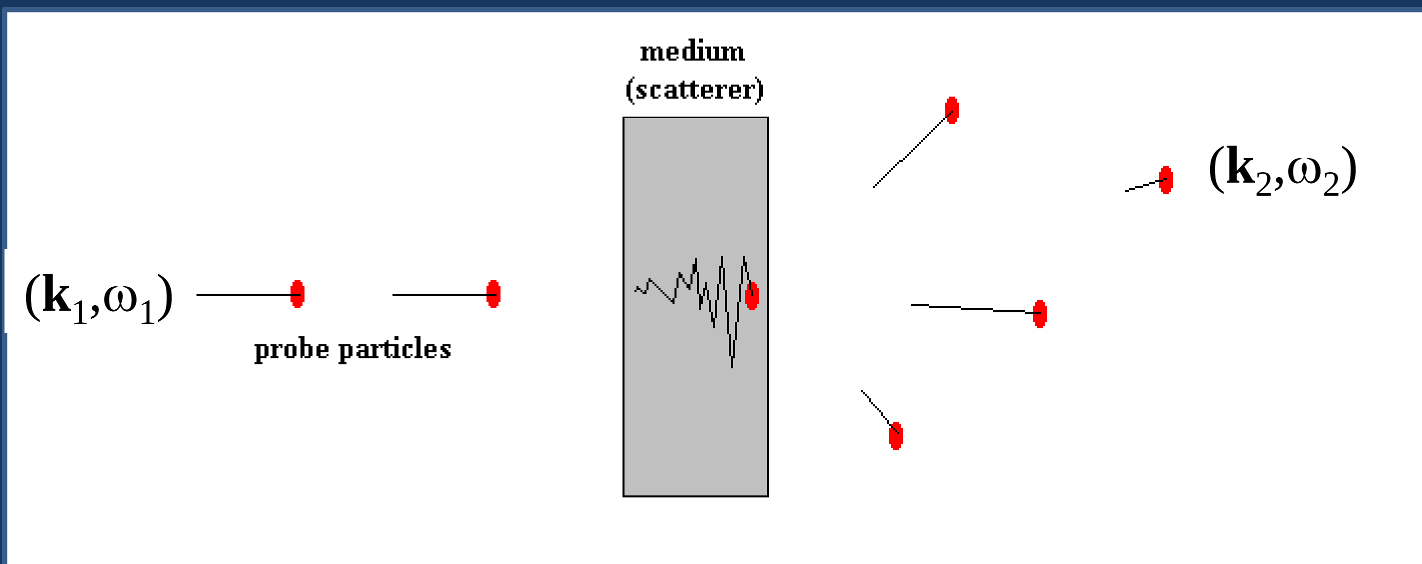


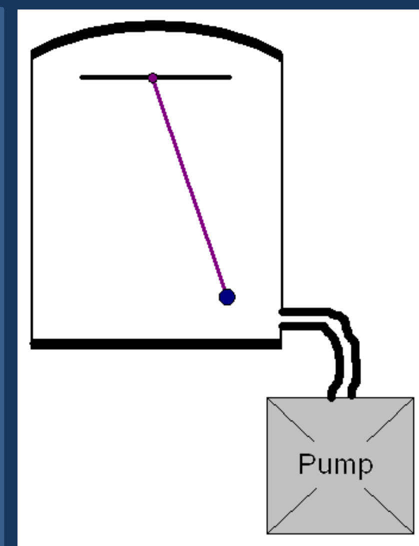
$$S(\mathbf{k},\omega)=\frac{1}{\pi}\frac{1}{1-e^{-\hbar\omega/kT}}\operatorname{Im}\left[\chi(\mathbf{k},\omega)\right]$$

$$\chi(\mathbf{x},\omega)=-\frac{i}{\hbar}\left\langle\left[\hat{n}(\mathbf{x},t),\hat{n}(0,0)\right]\right\rangle\theta(t)$$

Fluctuation-dissipation theorem

Retarded density Green's function



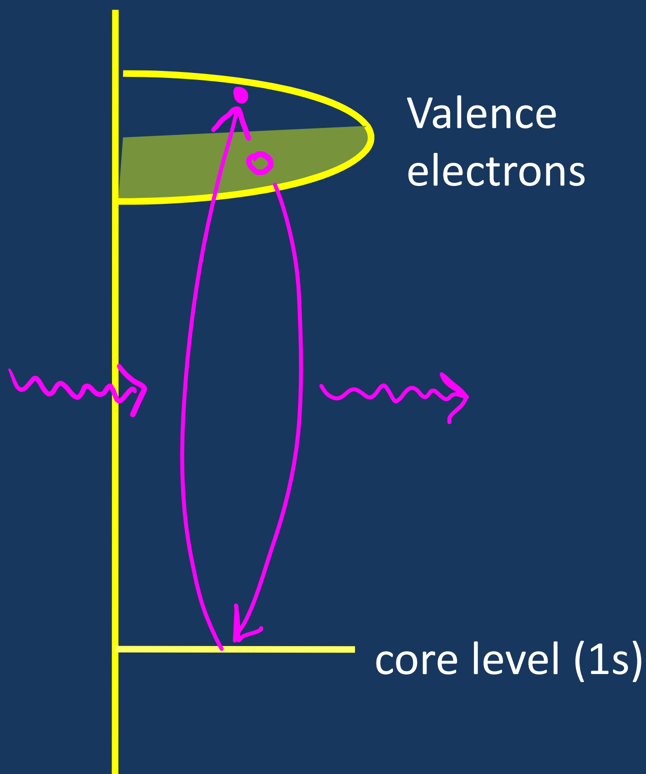


Describes how charge propagates around a system:

- Phonons
- Plasmons
- Excitons
- Band structure
- Etc.

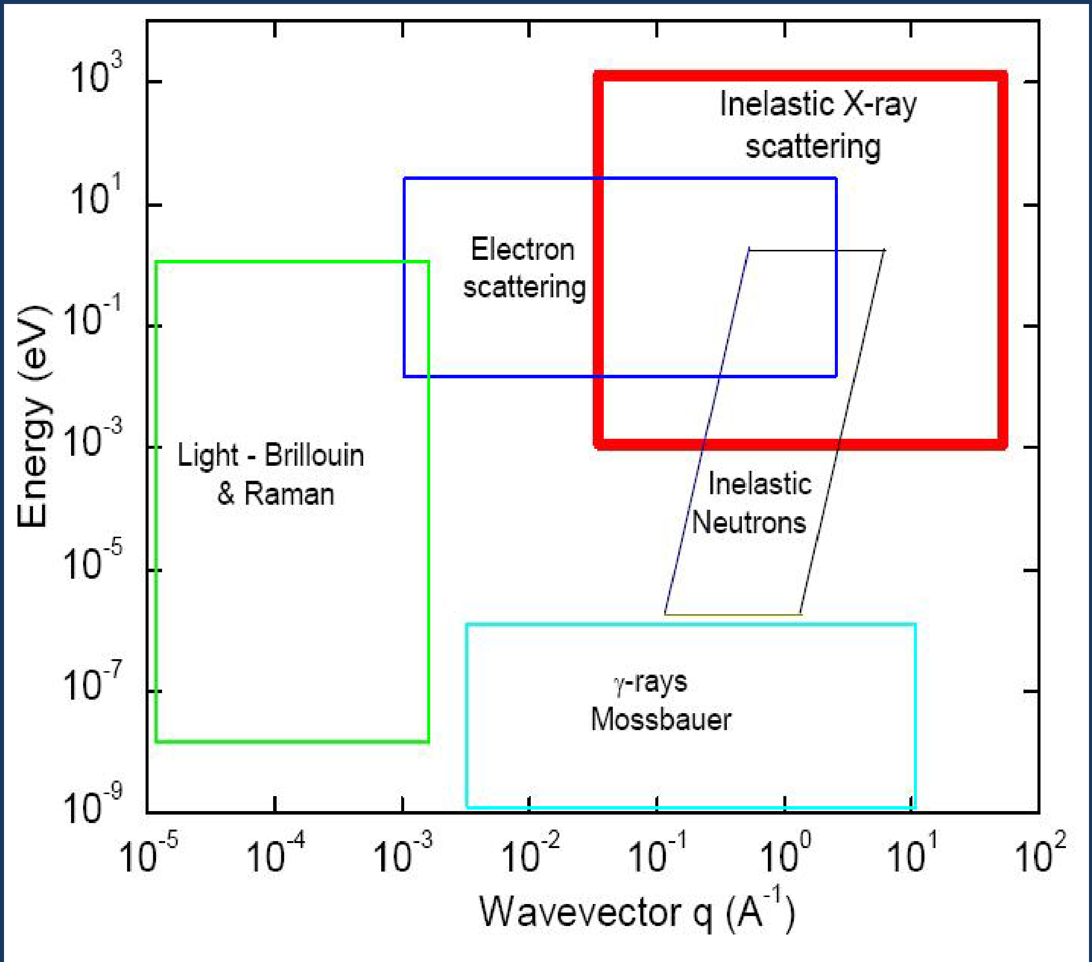
Resonant IXS (RIXS) – physical picture?

$$w_{f\leftarrow i} = \left| \frac{e^2}{mc^2\hbar^2} \sum_m \frac{< f|\mathbf{p}\cdot\mathbf{A}|m >< m|\mathbf{p}\cdot\mathbf{A}|0 >}{\omega_i - \omega_m + i\gamma} \right|^2 \delta(\omega - \omega_f + \omega_0)$$



- Coherent two-step process (absorption / emission)
- Denominator can diverge. More *intense.*
- Polarization-dependent; sensitive to symmetry of excitations
- Couples to many types of excitations (magnons?)
- No causality - *cross section not related to a correlation function. Often hard to decipher.*

Comparison to other techniques



Inelastic Neutron Scattering

- Low energy probe (ω_i = 25 meV)
- Better energy resolution
- Couples only to nuclei and spins
- Not sensitive to charge excitations (e.g. plasmons)
- Large samples required

Inelastic Electron Scattering

- Can focus beam ~ 1Å
- Decent energy resolution (0.5 eV)
- Sample damage
- Multiple scattering
- Does not work in $H \neq 0$
- Requires UHV

Light (Raman) Scattering

- q=0 probe
- Super high energy resolution (0.1 meV)
- Super high flux (10^{22} photons/sec on sample)
- Polarization selection rules
- cheap

Advantages / disadvantages of IXS

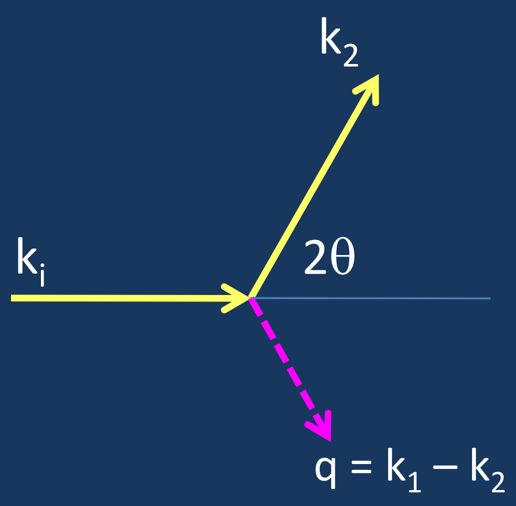
☺

- Direct coupling to charge excitations
- High resolution (<1 meV possible)
- Broad kinematic range
- Small samples OK
- Works in environments (high H, P, etc.)
- No need for high vacuum

☹

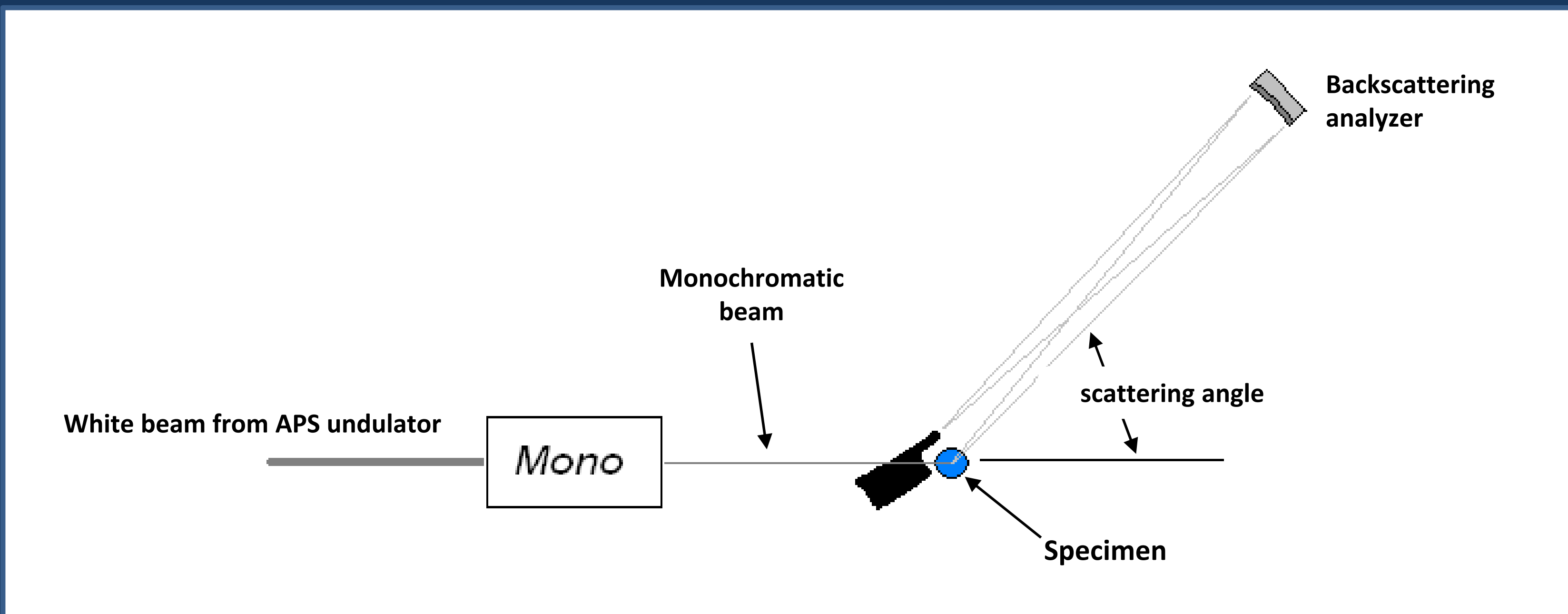
- Not sensitive to spin (possible exception from RIXS)
- Only measures excitations that modulate the electron density
- Longitudinal excitations only (cannot detect TO phonons, transverse plasmons, …)
- Prohibitive flux limitations at high resolution (only phonons practical)

Instrumentation: Overview



Requirements / Challenges

- Cross section small: only 1 in 10^8 photons are inelastically scattered – high flux needed
- Very high resolving power needed. 2 meV / 20 keV = 10^{-7}
- Must be able to tune energy; both ω_1 and ω_2 for RIXS
- Broad angular acceptance required for enough signal



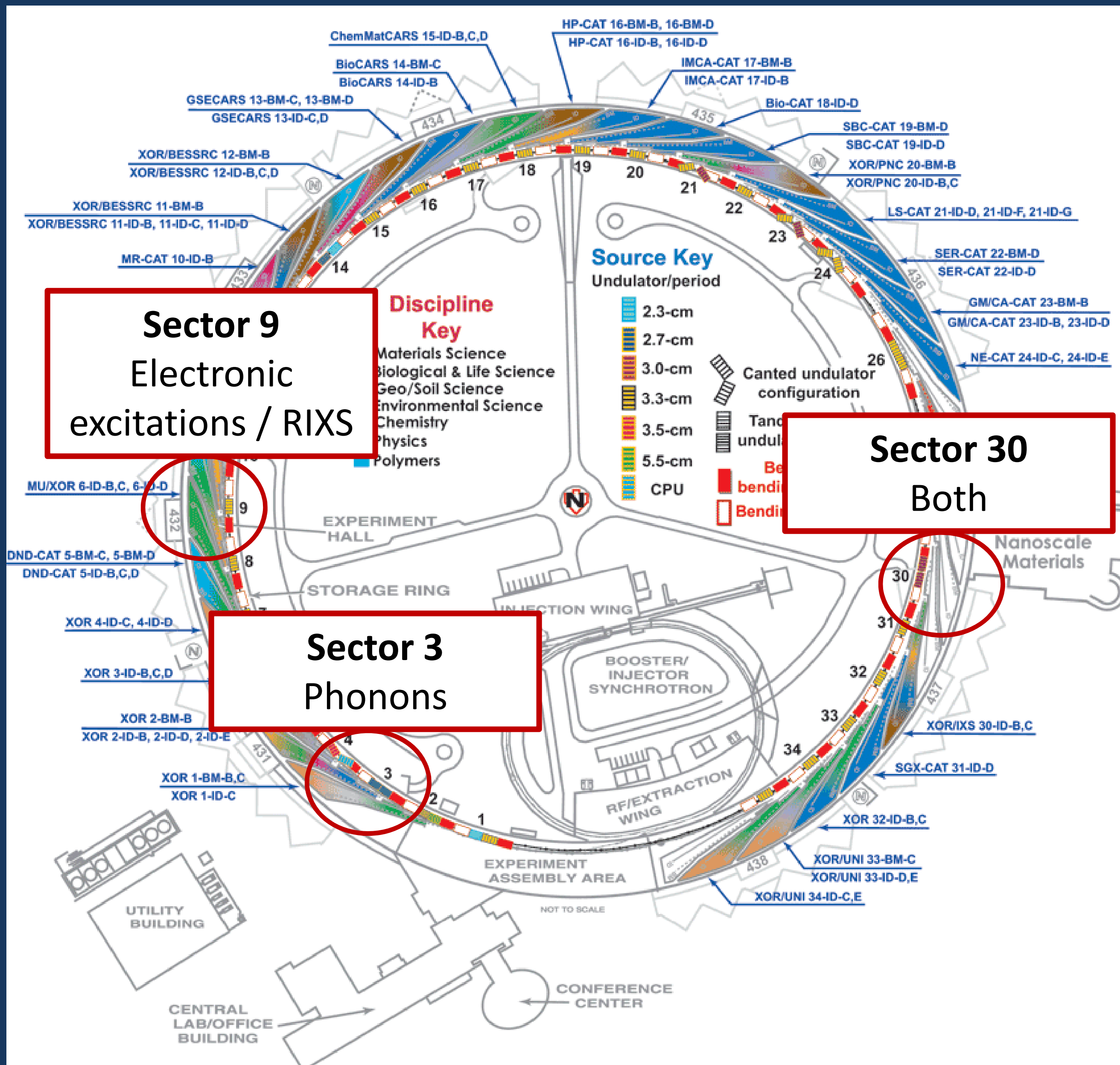
Instrumentation: Source

Spring-8 (Hyogo, Japan)

ESRF (Grenoble, France)

Advanced Photon Source
(Chicago, IL, USA)

Instrumentation: Source



Instrumentation: Sector 30 XOR-IXS Spectrometers

MERIX

ΔE = 10-100 meV

Electronic excitations:

- plasmons
- excitons
- Mott-gap excitations
- two-magnons

HERIX

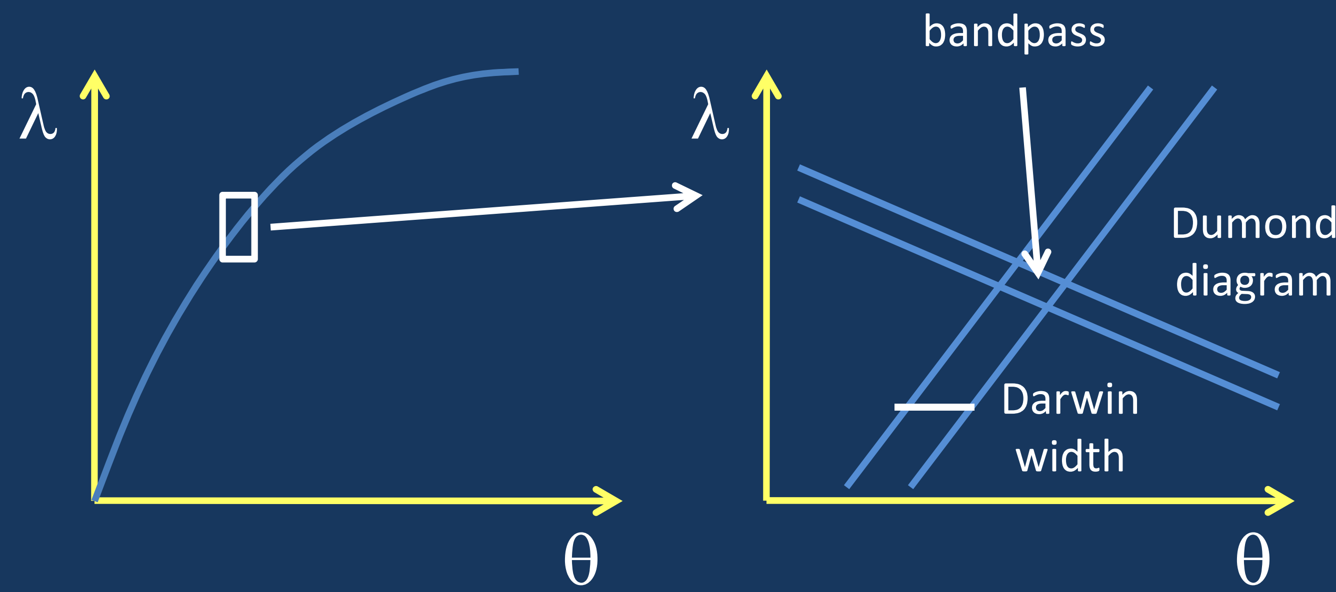
ΔE = < 1 meV

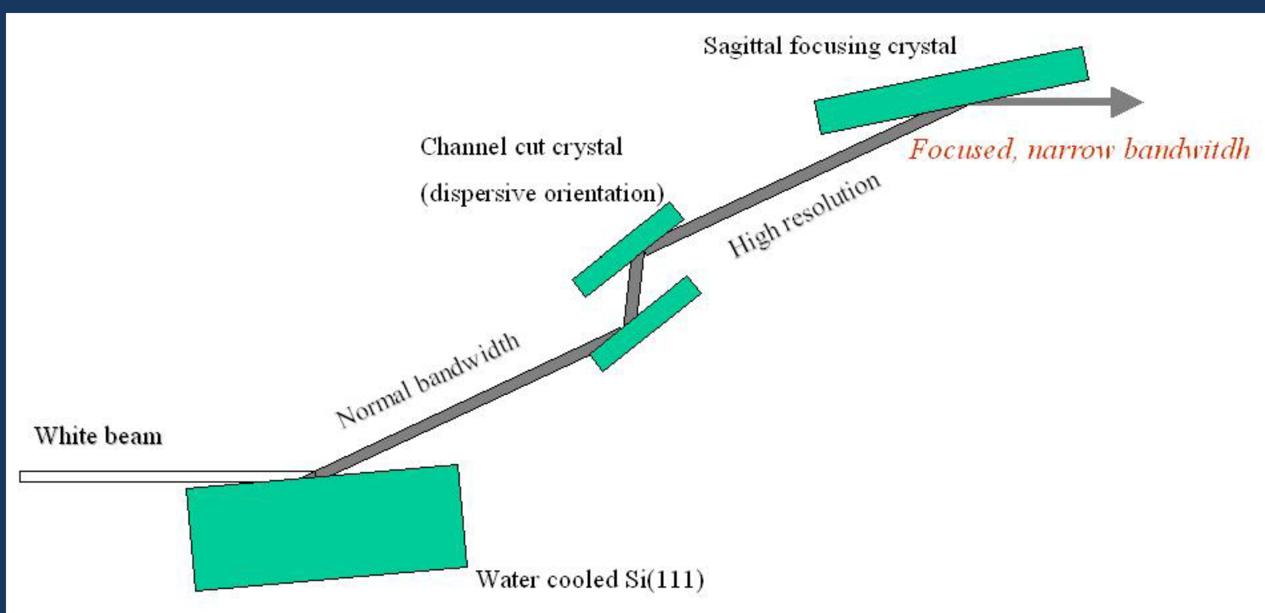
Vibration modes:

- acoustic phonons (sound)
- optical phonons

Instrumentation: Monochromator

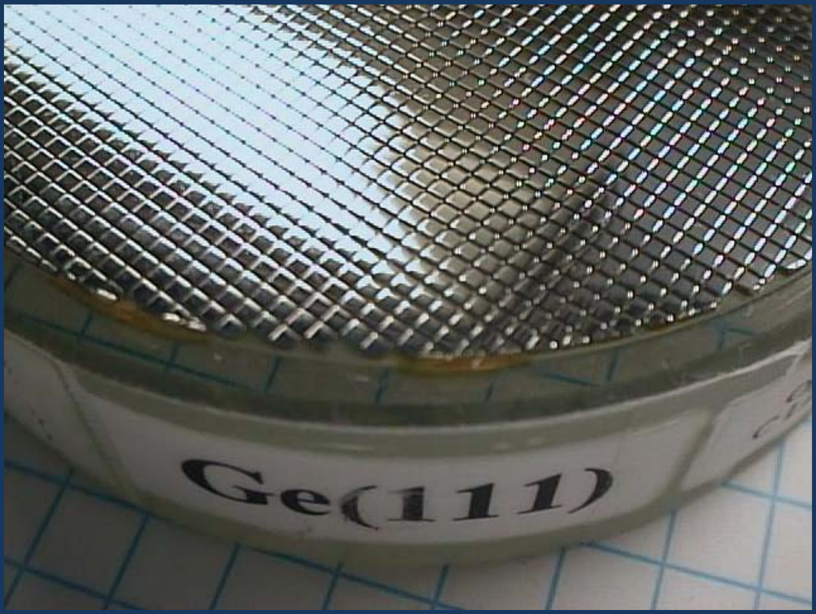
Bragg's law:
$2d \sin\theta = \lambda$

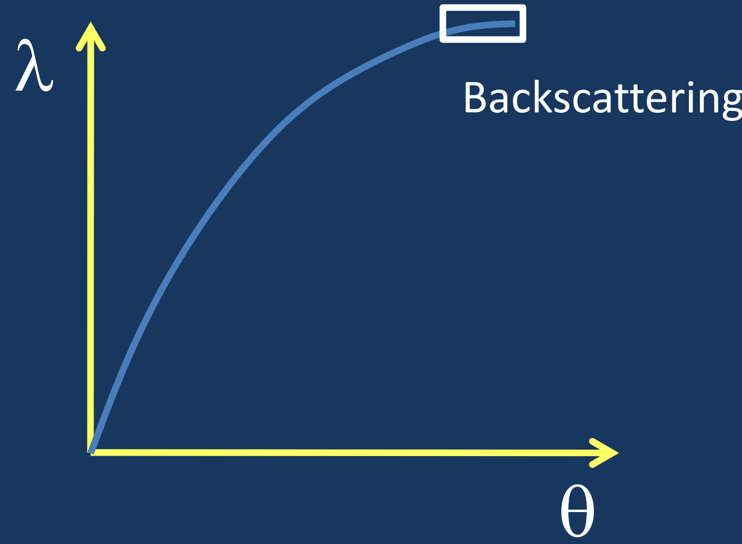




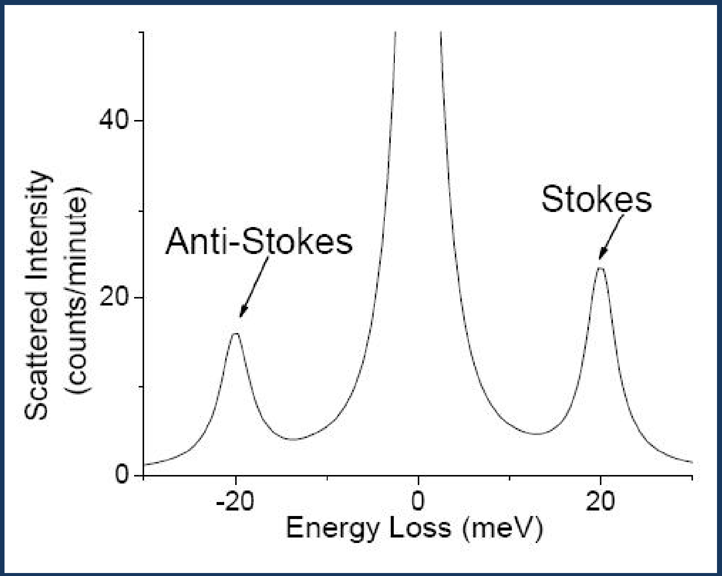
Tunable by rotating crystals in a coordinated fashion.

Instrumentation: Analyzer

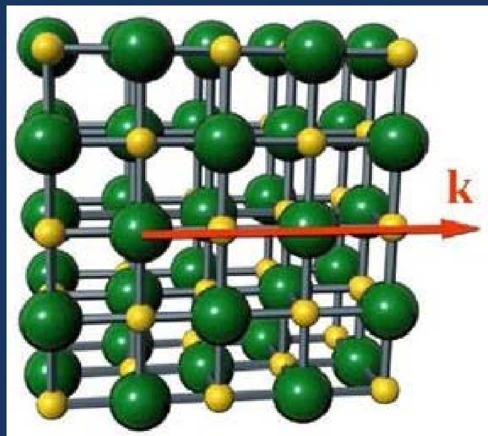


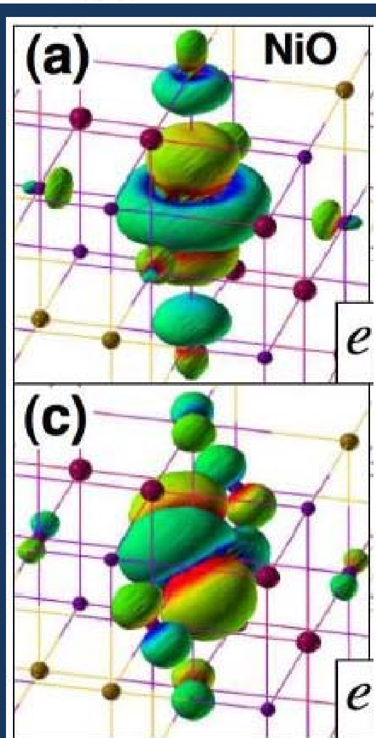


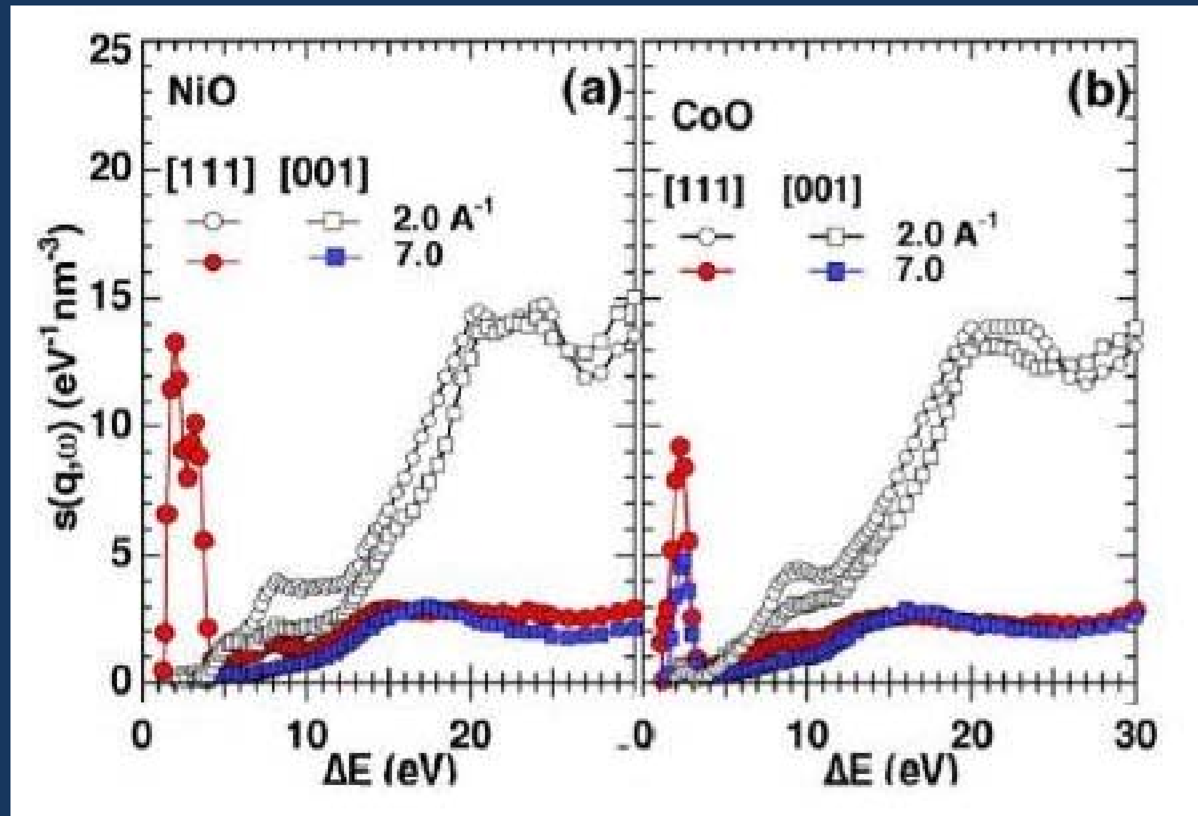
- Need "curved" crystal to accept large $\Delta\Omega$
- Mosaic of ~10^4 aligned blocks
- Glued on spherical surface
- Used near backscattering
- *Slightly* tunable by rotating angle



IXS Example 1: Excitons

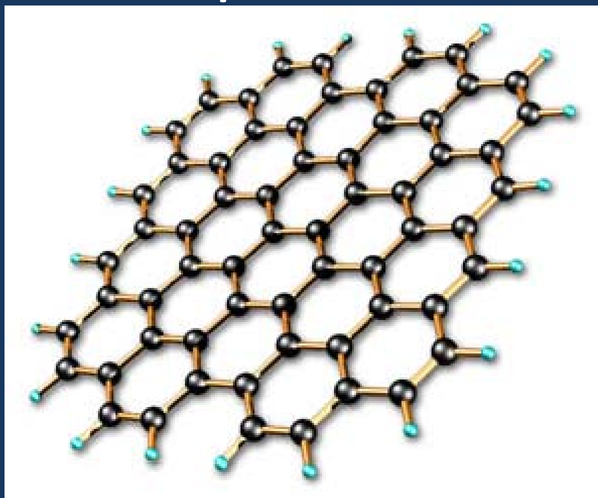


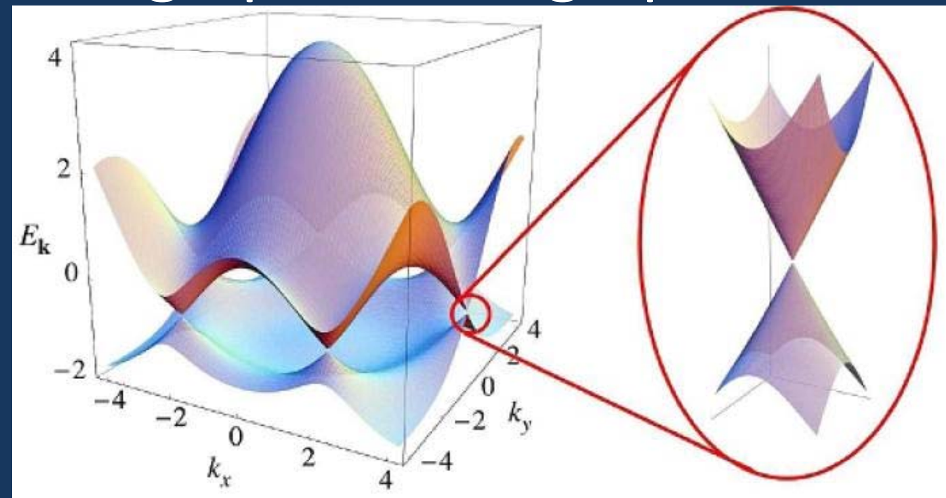




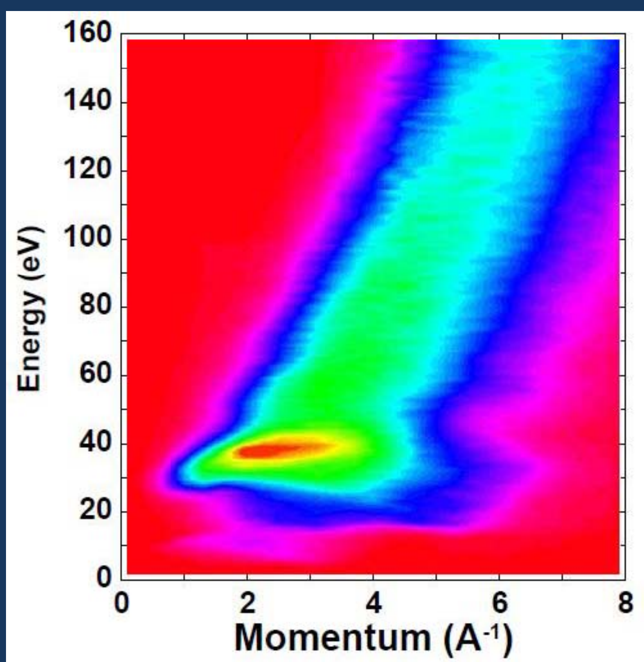
B. C. Larson, et. al., Phys. Rev. Lett. **99**, 026401 (2007)

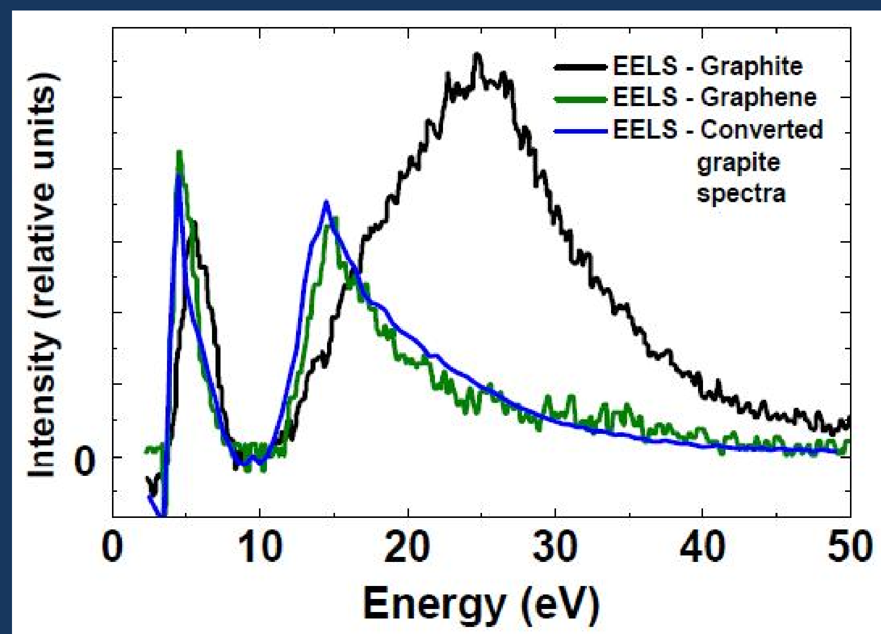
IXS Example 2: Plasmons in graphite and graphene





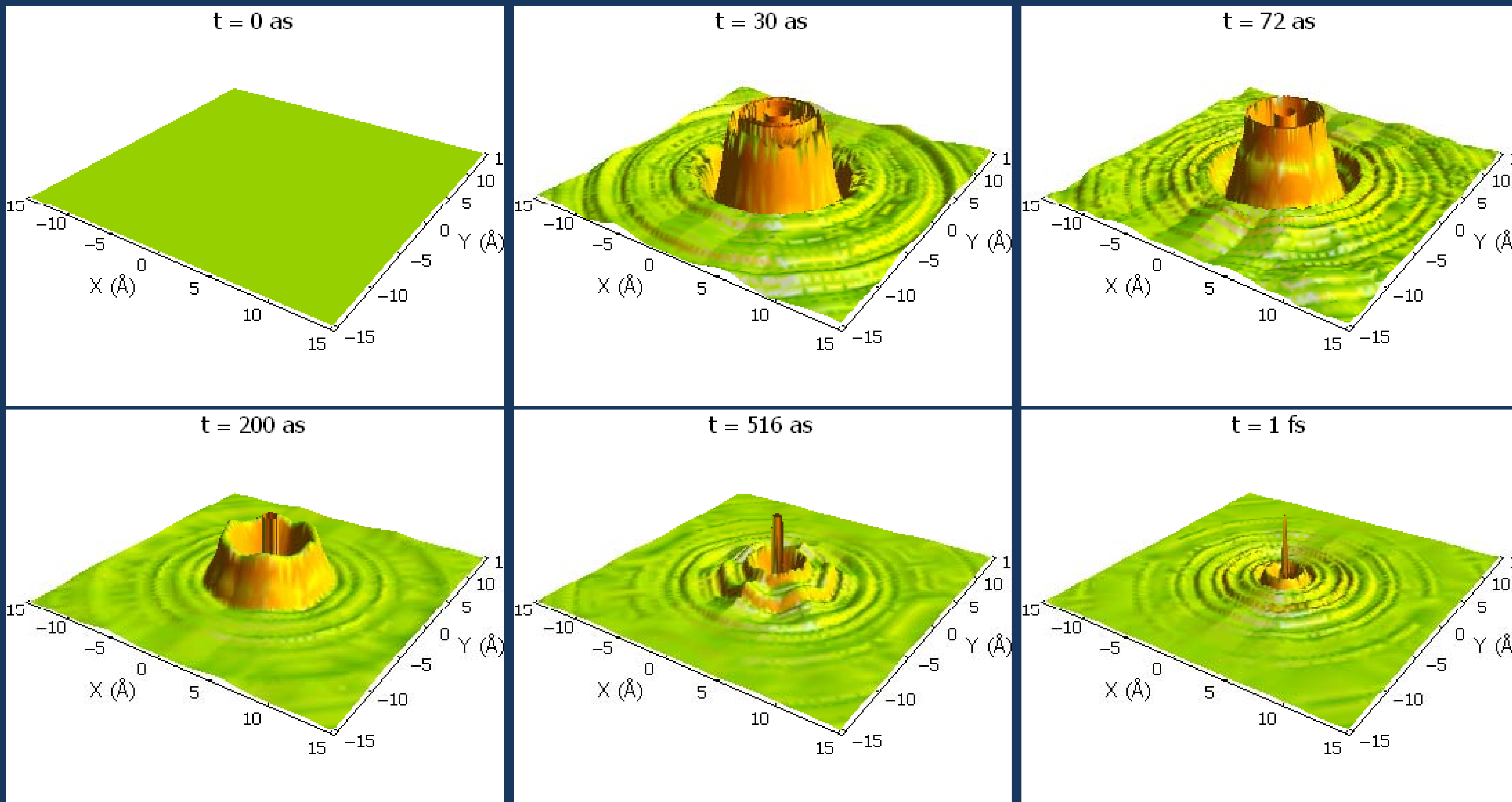
A. H. Castro- Neto, et al., Rev. Mod. Phys. **81**, 109 (2008)





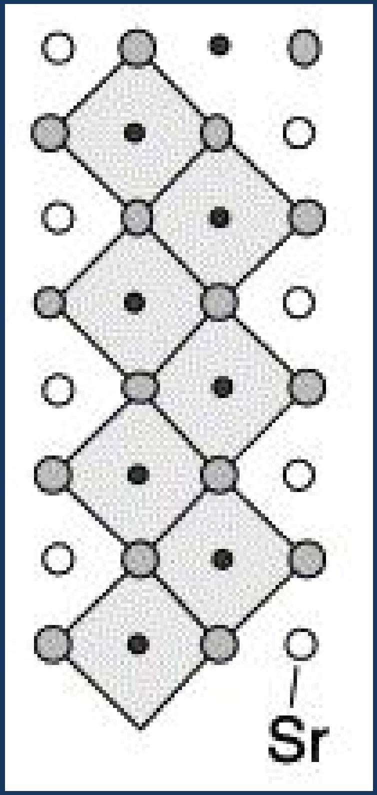
IXS Example 2: Plasmons in graphite and graphene

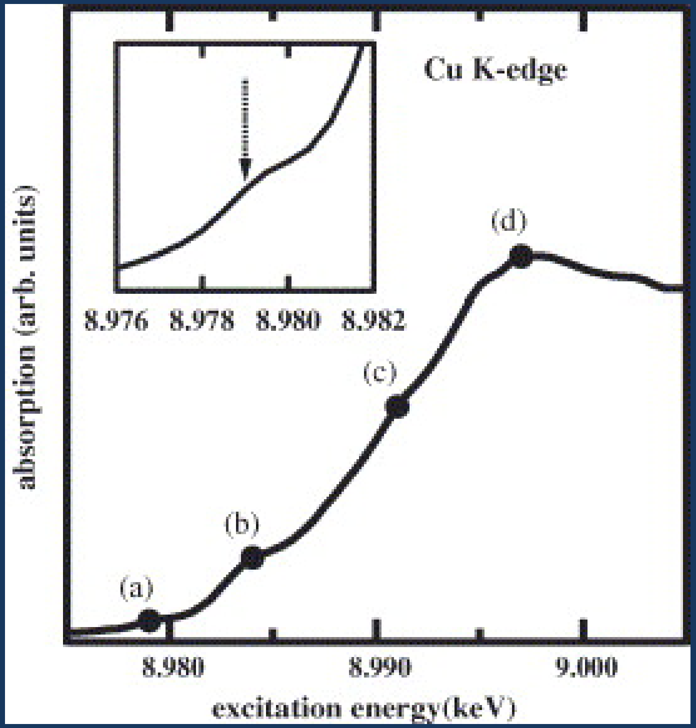
$$\chi(\mathbf{x},\omega) = -\frac{i}{\hbar}\left\langle \left[\hat{n}(\mathbf{x},t), \hat{n}(0,0) \right] \right\rangle \theta(t)$$

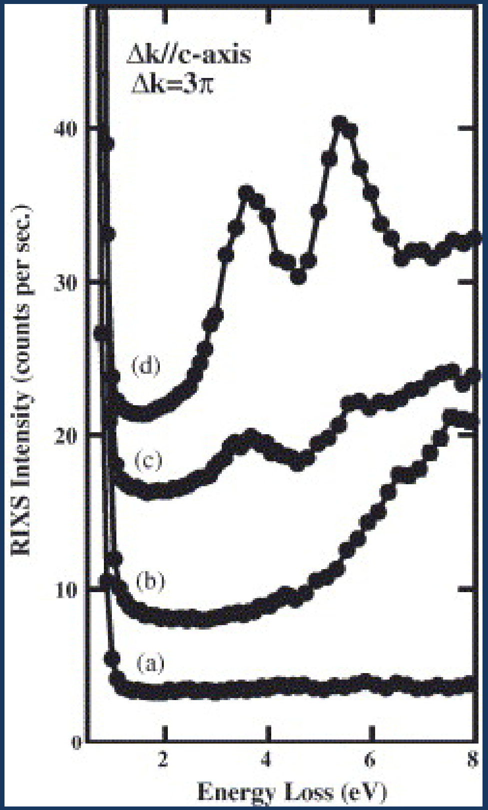


J. P. Reed, et al., submitted

RIXS Example 1: Crystal field excitations in $SrCuO_2$

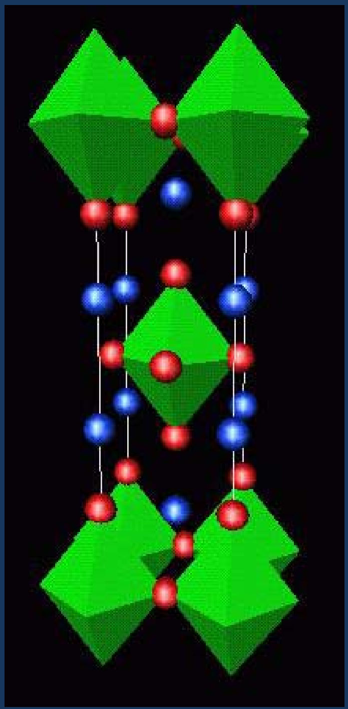


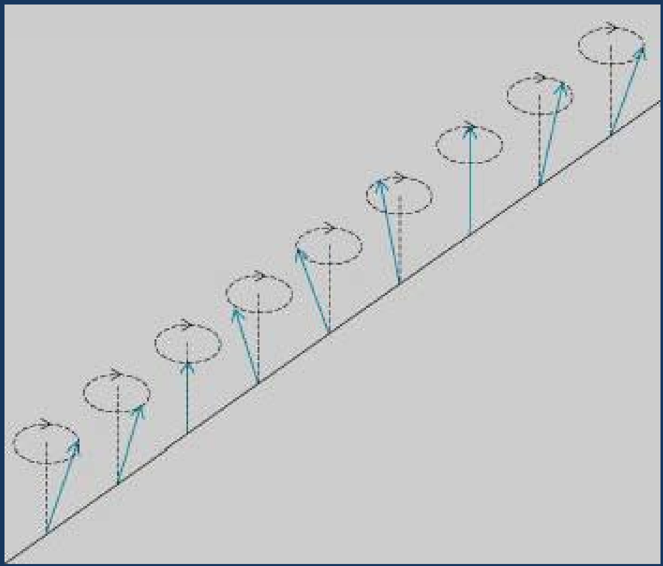


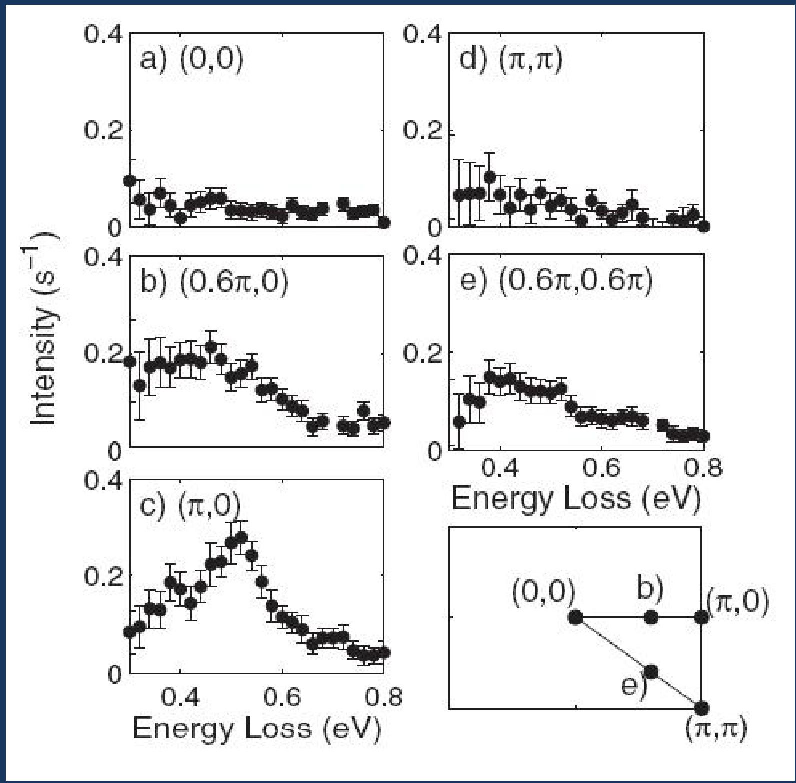


A. Higashiya et. al., J. Elect. Spect. Rel. Phen. **144-147**, 685 (2005)

RIXS Example 2: Magnons in La_2CuO_4







J. P. Hill, et. al., Phys. Rev. Lett. **100**, 097001 (2008)

The Future

- Join mainstream of experimental probes (with STEM, ARPES, etc.)
- Imaging
- Strip detectors (no more analyzers?)
- Surface dynamics [B. Murphy, et. al., Phys. Rev. Lett. **95**, 256104 (2005)]
- High pressure (fluctuations near quantum phase transitions)
- ??? New ideas from new people.