Inelastic X-ray Scattering at LCLS-II-HE

Some points to form a basis for discussion

Not a concrete scientific plan nor a proposal

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Flux at synchrotrons

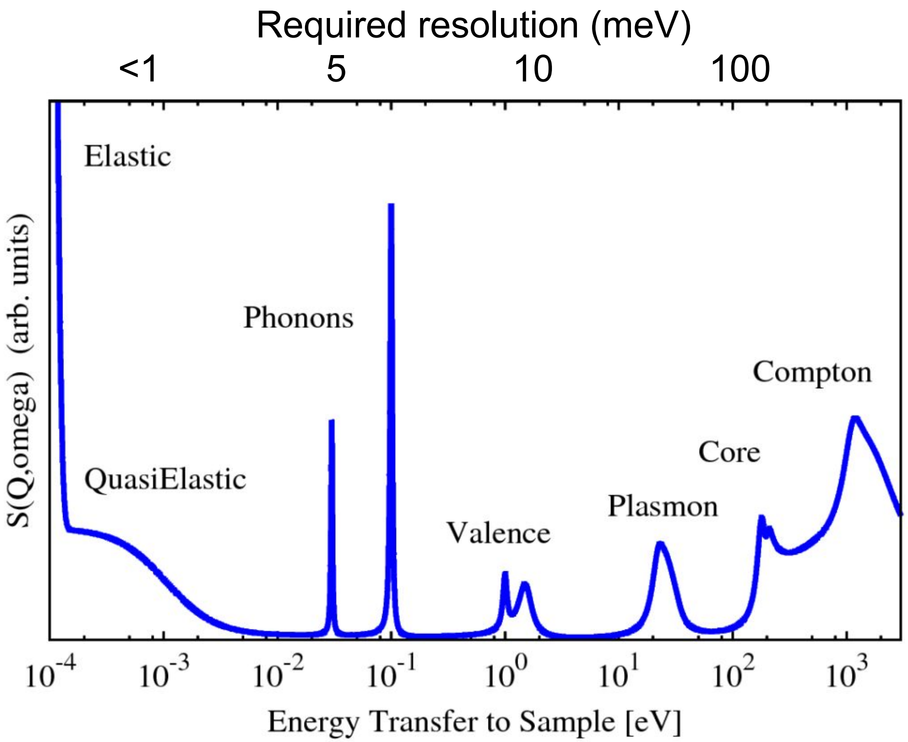
	Hard x-ray flux on sample per meV	Power on the sample (Watt)
Spring-8	~10^{11} ph/s (BL43)	0.0032 (~20 keV)
ESRF	~10^{10} ph/s (ID16, ID28)	0.00032 (~20 keV)
APS	~10^{10} ph/s (27-ID, 30ID) ~10^{9} ph/s (UHRIXS)	0.00032 (~20 keV) 0.00001 (~9 keV)
NSLS-II	~10^{10} ph/s (10-ID)	0.00014 (~9 keV)

A generic diffraction beamline with Si(111) at most 3rd gen. synch. has ~10^{13} ph/s/eV (0.14W @ ~9 keV)

10^{13} ph/s on sample with 100 kHz rep-rate = 10^{8} ph/pulse
= 0.0023 eV/atom per pulse (9 keV photons, 30 micron beam on YBCO)
Less than 0.01 eV per atom (a safe threshold)

Let's start from the assumption that we will have 10^{13} photons on the sample per second
We can/will go beyond that if sample/machine/optics cooperates

Types of IXS and typical resolution

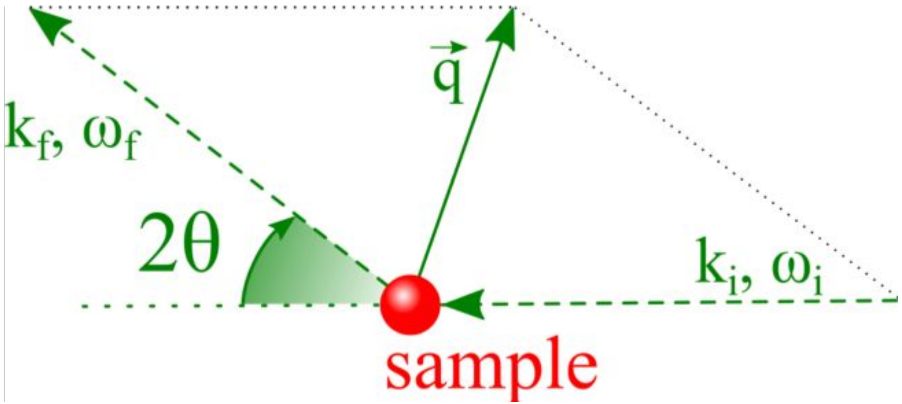


Goal (for the sake of argument):
5 meV overall resolution (we can discuss trade off later)
4 meV incident beam (5 meV overall)
< 0.5 ps time resolution
Phonon measurements
(electron-phonon, spin-phonon coupling, and a lot more where low-energy excitations are relevant)

Bonus option-1: broader resolution for finer time res.
Bonus option-2: magnetic inelastic signal (non-resonant)

Quick summary before we go further

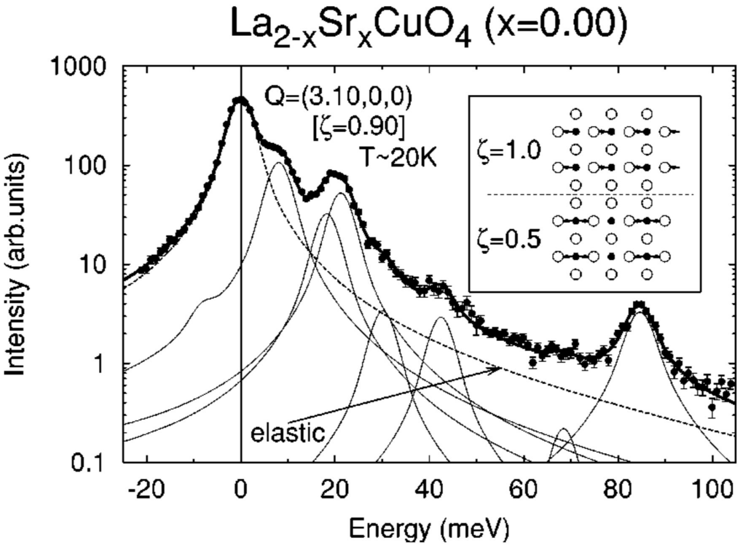
- 10^{13} photons per second on the sample with 4 meV bandwidth
  - For reference:
    BL43@SPring8: **~5x10^{10} ph/s** @ 17.8 keV (2.8 meV), 5 x 5 μm^2
    MERIX@APS and ID-20@ESRF: ~**2.5x10^{11} ph/s** @11.2 keV (15 meV), 20 x 10 μm^2
- 5 meV overall energy resolution: 4meV incident ⊕ 3meV analyzer (crystal, geometry, etc.)
  - 4 meV ↔ 450 fs
  - 0.014 Watt on the sample (9 keV), compatible with low-temperature measurements
  - Less than 0.01 eV/atom dose assuming 9 keV and a typical high-Z sample like YBCO



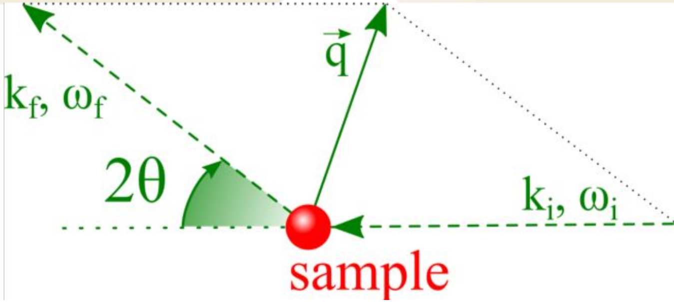
Takeaway (so far):

1. Moderate resolution of 5 meV enables phonon measurements in crystalline samples
2. Relevant samples endure 10^{13} photons/second
3. 10^{13} Hz is comparable to -if not better than- synchrotron beamlines for med-resol. IXS

Case study for phonon measurements



T. Fukuda, et al., *Phys. Rev. B*, **71**, 060501, 2005.



Incident beam:
3×10^{10} ph/s, 15.816 keV, 4 meV, ~100 μm

Analyzer:
Si(888): 4.5 meV (intrinsic + geometry)
100mm@9.8m: ~10x10 mrad^2, 0.076 Å^{-1}

~6 meV overall resolution
30 second per data point (120 pts = 1 hour per spectrum)

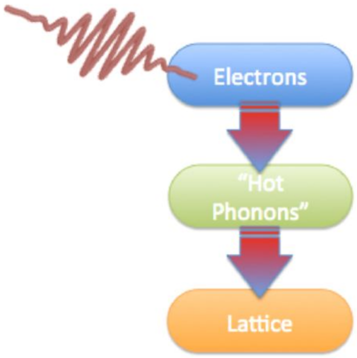
IF we recreate the "same" setup at 9 keV

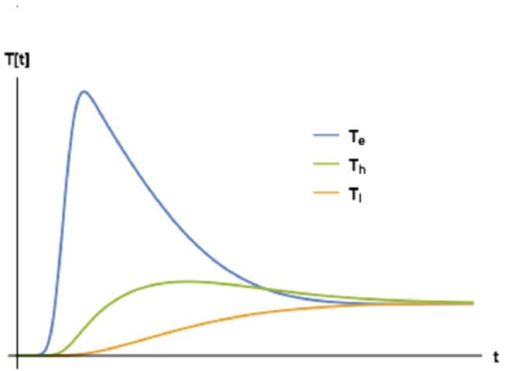
- Factor of 4 to 6 would be lost due to Thomson/photoelectric absorption assuming thick samples
- $10^{13}/3\times10^{10}$ = ~300 gain from the incident flux

~ x50 overall gain, i.e. less than 2 minutes per spectrum
This would allow another parameter to investigate: **time!**
Cherry on top:
If we could serialize data collection with a dispersive analyzer

Possible science experiment (one of many)

Just to demonstrate the sensitivity of an efficient IXS spectrometer (not the ONLY application)

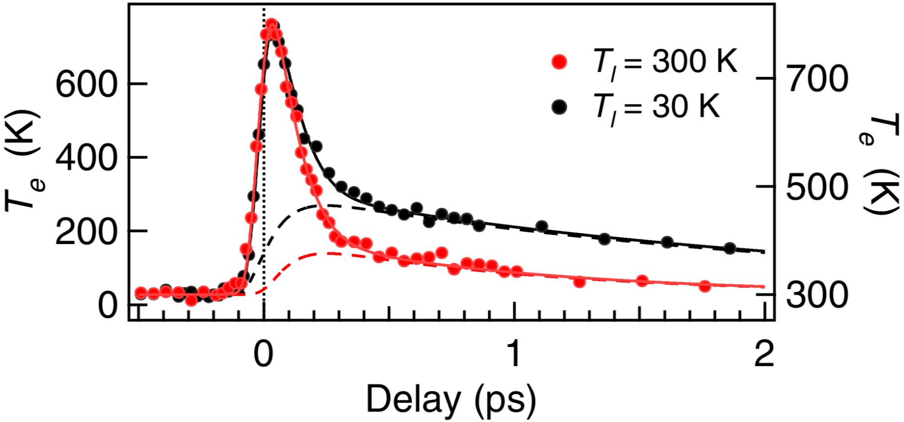




3-temperature model for energy flow in a system

S. L. Johnson, et al., Struct. Dyn. **4**, (2017).

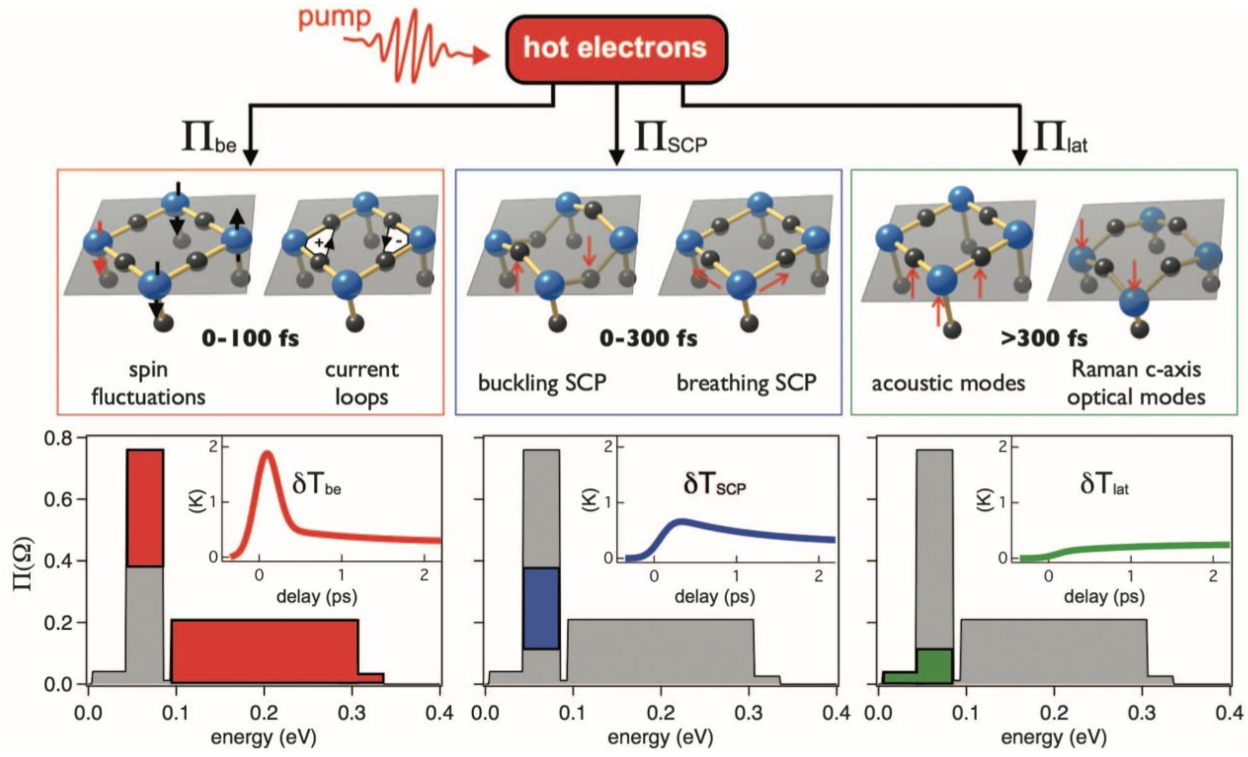
The Fermi edge spread in ARPES measurements allows to estimate the temperature of the electron system



L. Perfetti, et al., Phys. Rev. Lett. **99**, 197001 (2007).

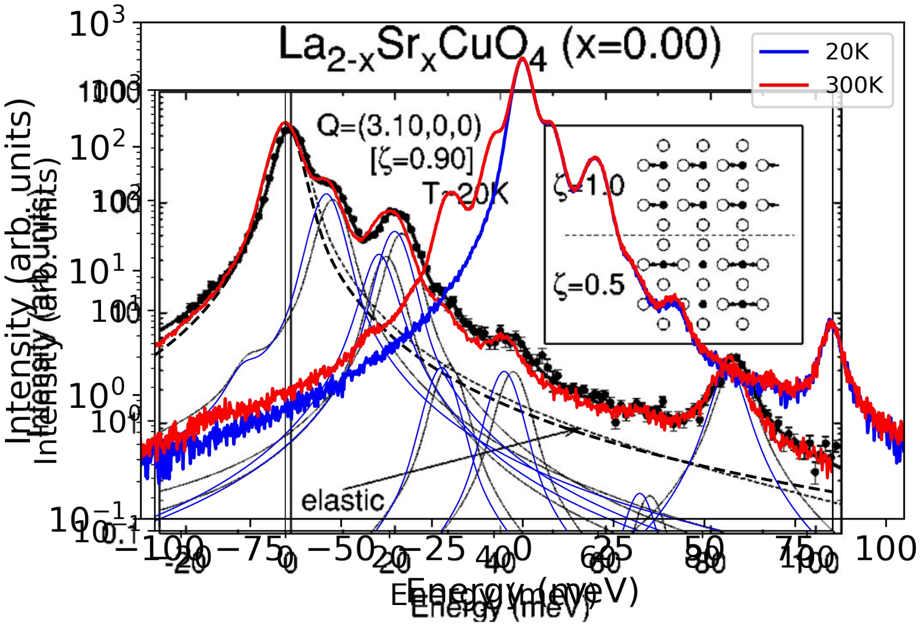
Two kinks in the temperature curve indicates two sets of phonons thermalizing after the pump.
Model calculations estimate ~20% of the phonons thermalize first (hot phonons) before the rest of the lattice warms up

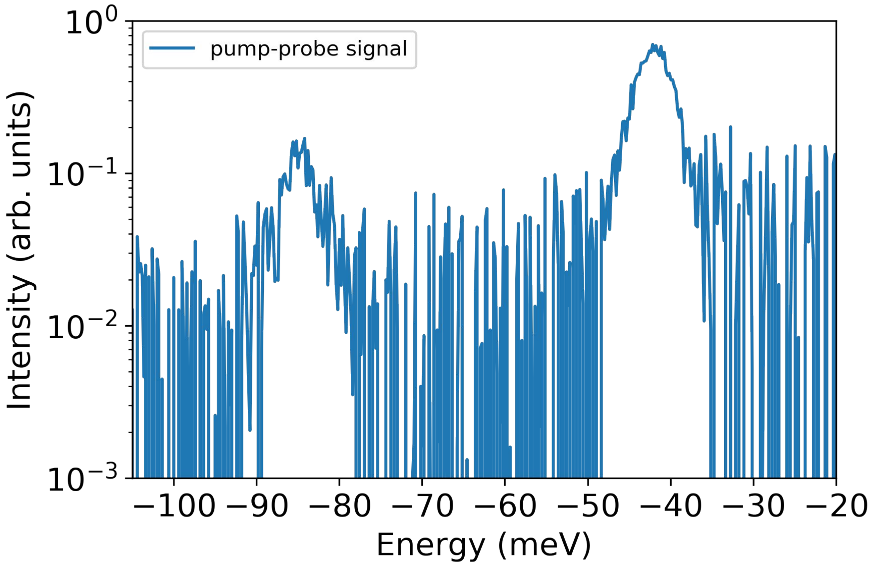
Complicated dynamics, where mode-specific probe would be helpful



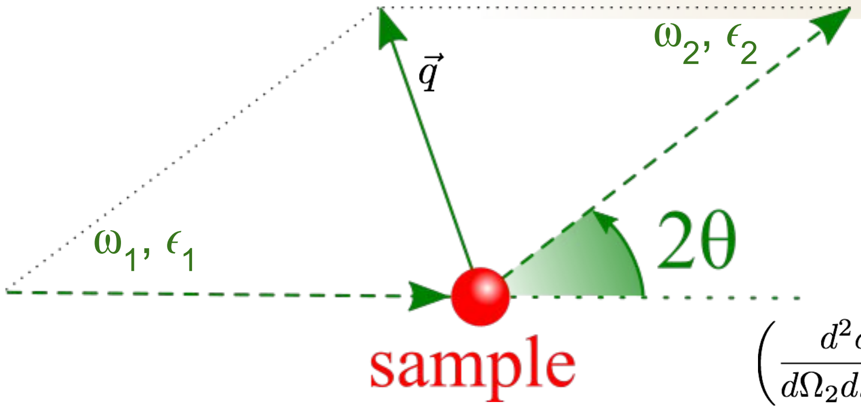
Possible science experiment (one of many)

This is an exercise to demonstrate the sensitivity of a high-efficiency phonon spectrometer. There is no other claim. The data is reproduced with realistic statistics.

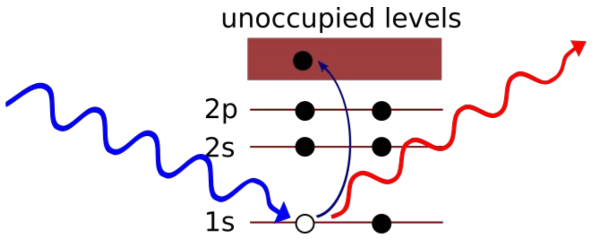




Non-resonant IXS (phonons and more)



$E_i \rightarrow E_f$



$$\left(\frac{d^2\sigma}{d\Omega_2 d\hbar\omega_2}\right)_{i,1\to f,2} = \left(\frac{e^2}{mc^2}\right)^2 \left|\left\langle f \left| \sum_j e^{i\vec{q}\cdot\vec{r_j}} \right| i \right\rangle \vec{\epsilon_1}\cdot\vec{\epsilon_2}\right|^2 \delta(E_i - E_f + \hbar\omega_1 - \hbar\omega_2)$$

$$\left(\frac{d^2\sigma}{d\Omega_2 d\hbar\omega_2}\right)_{i,1\to f,2} = \left(\frac{e^2}{mc^2}\right)^2 \left|\underbrace{\left\langle f \left| \sum_j e^{i\vec{q}\cdot\vec{r_j}} \right| i \right\rangle \vec{\epsilon_1}\cdot\vec{\epsilon_2}}_{\text{charge}} - i\underbrace{\frac{\hbar\omega'}{mc^2}\left\langle f \left| \sum_j \vec{s_j} e^{i\vec{q}\cdot\vec{r_j}} \right| i \right\rangle}_{\text{spin}} \cdot \vec{\epsilon_1}\times\vec{\epsilon_2}\right|^2 \delta(E_i - E_f + \hbar\omega_1 - \hbar\omega_2)$$

$\omega' \approx \omega_1 \approx \omega_2$

$\hbar\omega' = 10^4$ eV , $mc^2 = 0.5\text{x}10^6$ eV → spin term is 2 orders of magnitude smaller.
This is not the whole story. Signal on the detector is much weaker.

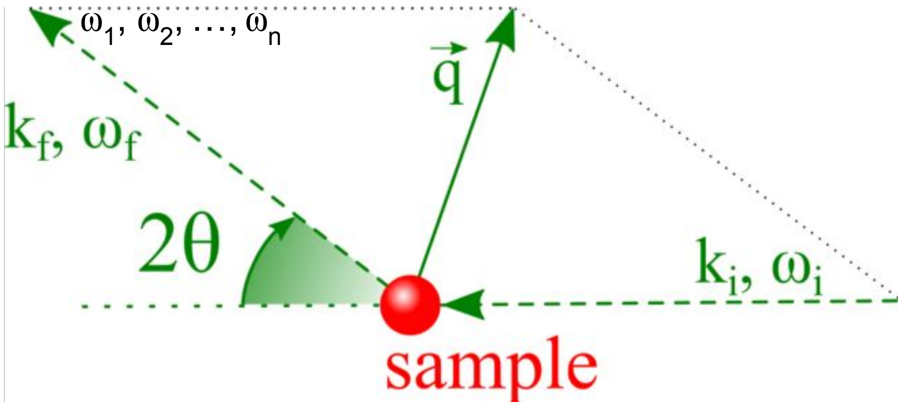
Can we make it up?

	Hard x-ray flux on sample per meV
LCLS-II-HE (seeded)	**~10^{14} ph/s**
LCLS-II-HE (SASE)	**~10^{13} ph/s**
Spring-8	~10^{11} ph/s (BL43)
ESRF	~10^{10} ph/s (ID16, ID28)
APS	~10^{10} ph/s (27-ID, 30ID) ~10^{9} ph/s (UHRIXS)
NSLS-II	~10^{10} ph/s (10-ID)

- The resolution can be relaxed in favor of higher flux if the sample can tolerate

$$\left(\frac{d^2\sigma}{d\Omega_2 d\hbar\omega_2}\right)_{i,1\to f,2} = \left(\frac{e^2}{mc^2}\right)^2 \left| \left\langle f \left| \sum_j e^{i\vec{q}\cdot\vec{r_j}} \right| i \right\rangle \vec{\epsilon_1}\cdot\vec{\epsilon_2} - i\frac{\hbar\omega'}{mc^2} \left\langle f \left| \sum_j \vec{s_j} e^{i\vec{q}\cdot\vec{r_j}} \right| i \right\rangle \cdot \vec{\epsilon_1}\times\vec{\epsilon_2} \right|^2 \delta(E_i - E_f + \hbar\omega_1 - \hbar\omega_2)$$

What kind of spectrometer we should have, why?



Let's assume we have the monochromator that delivers the FT-limited beam at the photon energy we want with the desired bandwidth

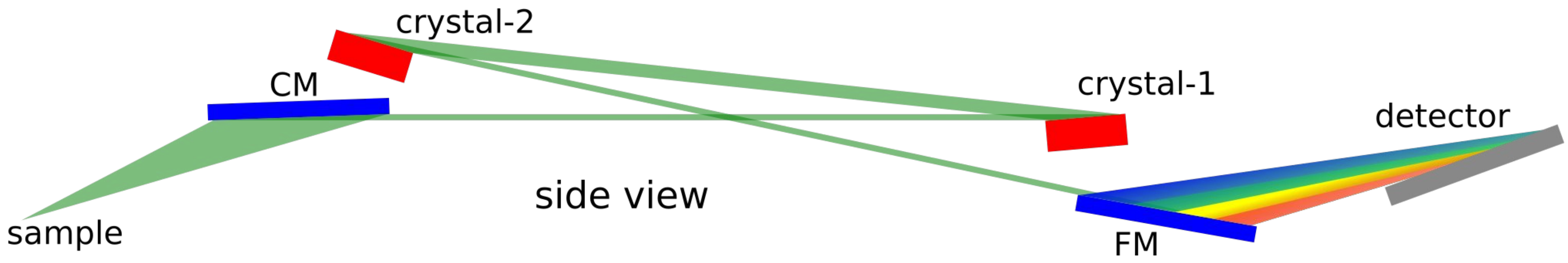
We should have an energy dispersive analyzer that could capture a spectral window as broad as possible.

- Limits/reduces the radiation damage on the sample
- Eliminates/reduces the complications due to source fluctuations

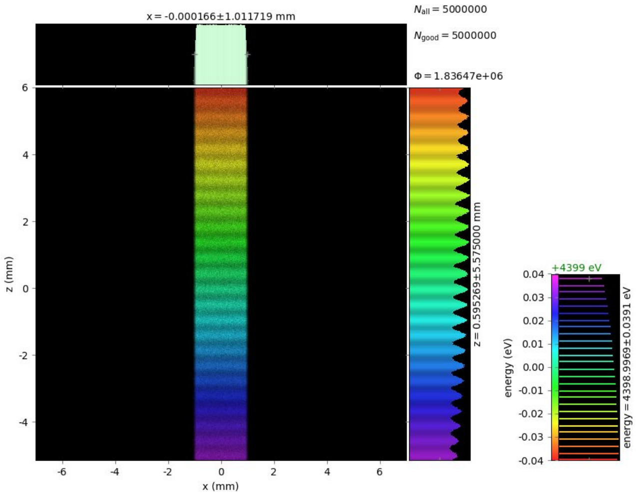
- Allows bining the spectral window to improve statistics when energy resolution is not the priority
- It is more efficient so gives us a better chance to detect "faint" signals like magnetic excitations.

For example: The total number of electrons in YBa2Cu3O7 is 294. Looking for a single unpaired spin is akin to finding a needle in the haystack.
Ratio of non-resonant magnetic signal to Thomson can be on the order of $10^{-8} = (10^{-2} \times 10^{-2})^2$
10x10 mrad^2 solid angle at 90° eliminates ~6×10^{-7}, which can be further improved (more discussion/work needed)

A "prototype" #1



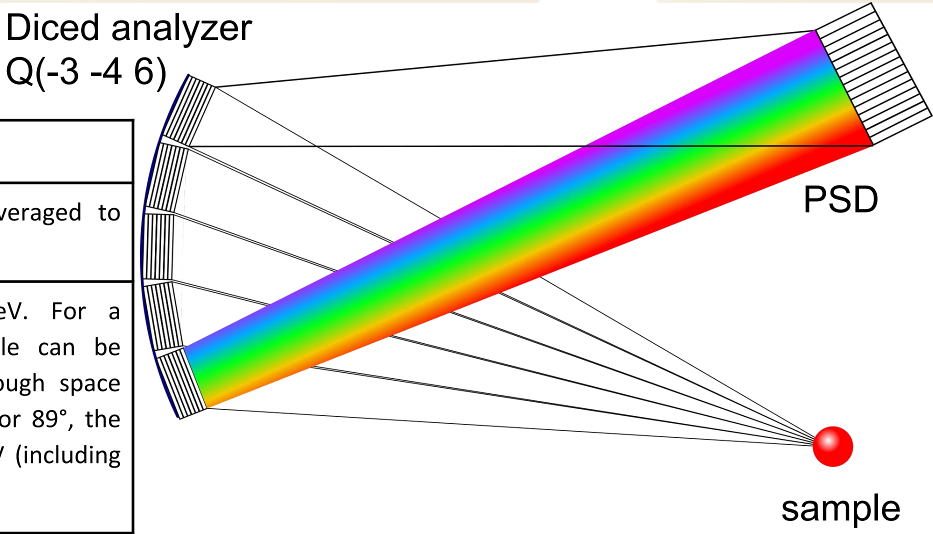
Parameter		Value	Notes
Beam size on the sample		10µm(V)	Larger horizontal beam size can be leveraged to reduce the heat-load on the sample
CM	Focal distance	200 mm	These parameters may not be optimal. A more careful analysis with an experienced vendor should be done. The beam size at the center of the mirror is taken as 2x2 mm^2. Better than 2 µrad slope error can be achieved for elliptical mirrors. So this value can be better.
	Coating pair	Ni-C	
	Optical length	150 mm	
	Slope error	2µrad(rms)	
Analyzer crystals		Ge(400)	Bragg and asymmetry angles are 85°and 80°, respectively. The optical length along the beam path is less than 30 mm.



A “prototype” #2

Aconventional approach can be suggested as a baseline.

Parameter		Value	Notes
Beam size on the sample		10µm(V)	Larger horizontal beam size can be leveraged to reduce the heat-load on the sample
Analyzer	Radius	2000 mm	Photon energy is around 11.216 keV. For a non-resonant approach, the Bragg angle can be arbitrarily small as long as there is enough space between the sample and the detector. For 89°, the analyzer resolution will be around 3 meV (including the geometrical contributions)
	Material	Quartz	
	Miller indices	-3 -4 6	
	Intrinsic BW	2.12 meV	
Incident BW		4 meV	With 4 meV incident and 3 meV analyzer bandwidth, the overall resolution will be around 5 meV
Detector pixel size		50xL μm^2	A stripe detector would work. It a 2D detector, the nondispersive direction should be integrated



Dispersion:
With analyzer cube size of 1 mm, ~100 meV spectral range can be recorded without any scanning.

Works at higher photon energies, polarization analysis of the scattered photons is not as easy.

A possible layout

Pump Laser

Monochromator → Sample chamber → Analyzer

Stage-I

~4 meV @ ~10 keV

Optimal photon energy(s) to be determined

Resolution can also be tweaked

Versatile sample chamber

~3 meV resolution, dispersive (~5 meV overall)

Resolution can be tweaked

Stage-II

Versatile monochromator

Tunable bandwidth? Swapping crystals?

~10 meV & ~100 meV on-demand

→

Lattice and spin (~5 meV and 10s meV)

Core-level excitations (100s meV)

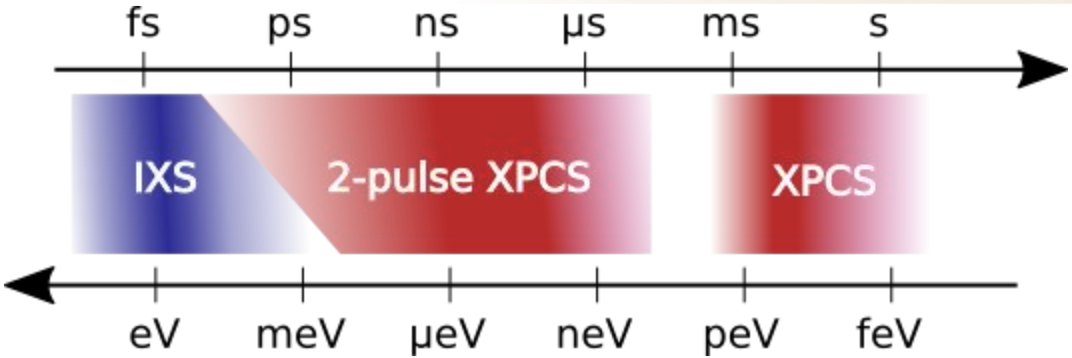
RIXS (10s meV)

Resonant IXS option

Conventional analyzers (spherical, diced)

PAX analyzer (photoemission - G.D.)

Do we want/need to push the energy-resolution?



Also, FT-IXS?