Inelastic X-ray Scattering in the Hard X-ray Regime

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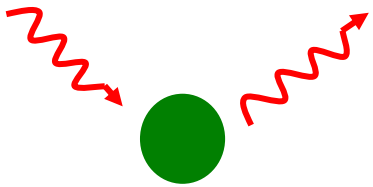
Inelastic X-ray Scattering

Photon-in – photon out



$q=k_i-k_f$

$\omega=\omega_i-\omega_f$

Probes charge-neutral excitations:

- Phonons, diffusive modes, orbitons, superconducting gaps……
- Excitons, plasmons, particle-hole creation, interband transitions…

Complements existing techniques:

- ARPES measures $A(q,\omega)$, surface sensitive.
- INS does not couple to charge, and requires large sample.
- EELS cannot measure to large q, or in fields.
- $\sigma(\omega)$, Raman are restricted to q=0.
- SX-RIXS, different intermediate states, smaller q range.

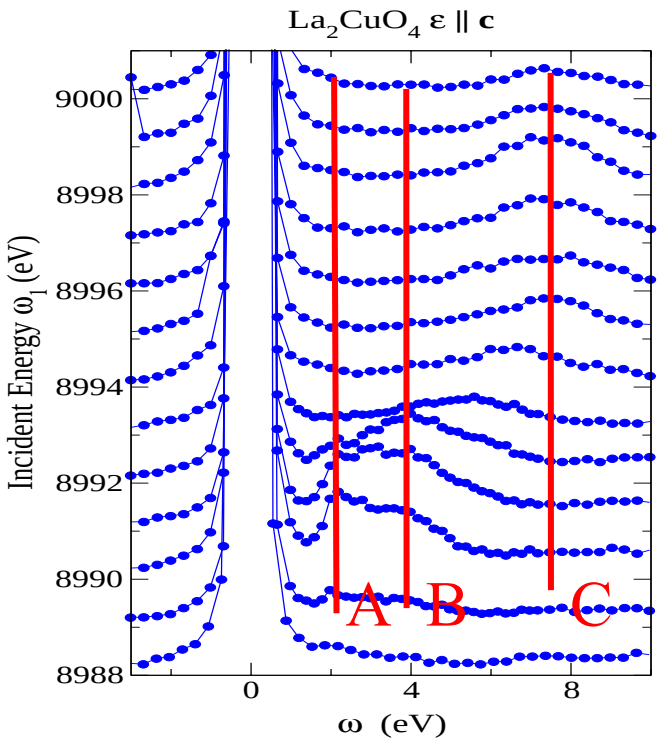
Example 1: Excitons in $La_{1-x}Sr_xCuO_4$

Resonant Inelastic X-ray Scattering

Tune to Cu K-edge
➡ x 300

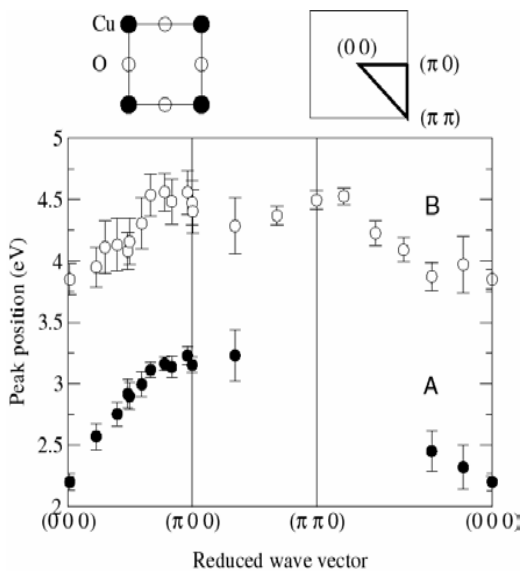
APS CMC-CAT

Si(3,3,3) inc. flux = 6×10^{11} ph. s^{-1}
ΔE=400 meV, ΔQ=0.3 A^{-1}



~ 2 cps

Measure dispersion and lifetime



(Highly dispersive and highly damped)

Y.-J. Kim *et al.*, PRL **89** 177003 (2002).

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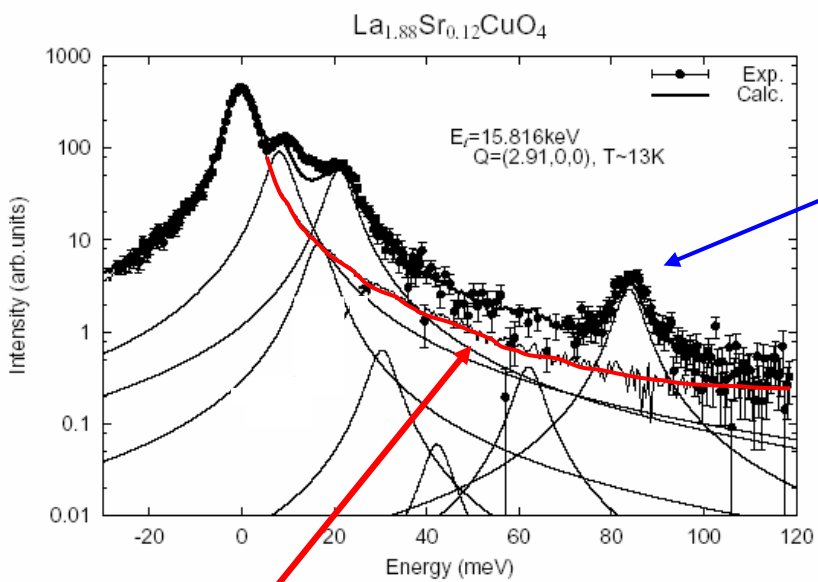
Example 2: Phonons in $La_{1-x}Sr_xCuO_4$

Fukada *et al.* , cond-mat/0306190

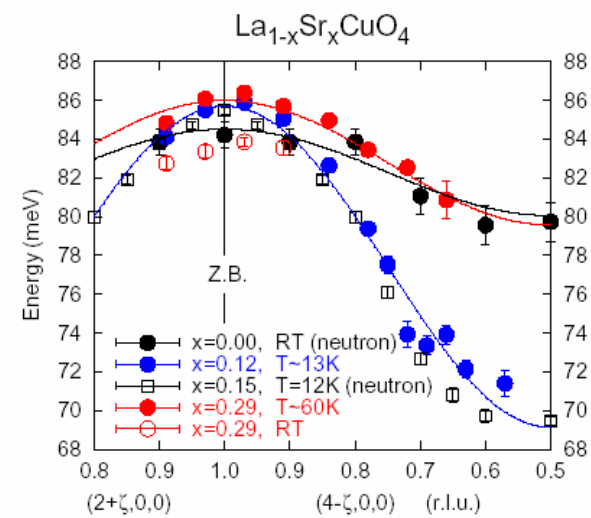
SPring-8 BL35XU

Si(8,8,8) inc. flux = 3×10^{10} ph. s^{-1}
ΔE=6 meV, ΔQ=0.076 A^{-1}



0.4 cps

Elastic tails



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Existing 3rd Generation Beamlines

ESRF (6 GeV, 200 mA)

1.5 x meV lines

0.5 x eV lines

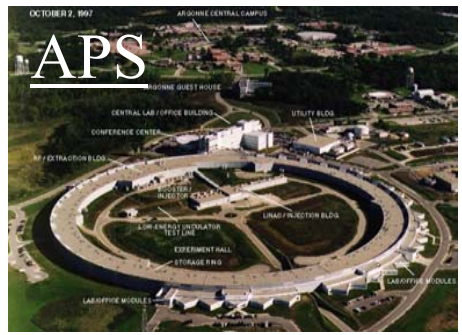
4.5m λ=3.5cm

APS (7 GeV, 100 mA)

0.5 x meV line — 2.4 m λ=3.3 cm

0.5 x meV line — 2.4 m SCU λ=1.4 cm

0.5 x eV line — 7.2 m λ=3.0 cm



SPring-8 (8 GeV, 100 mA)

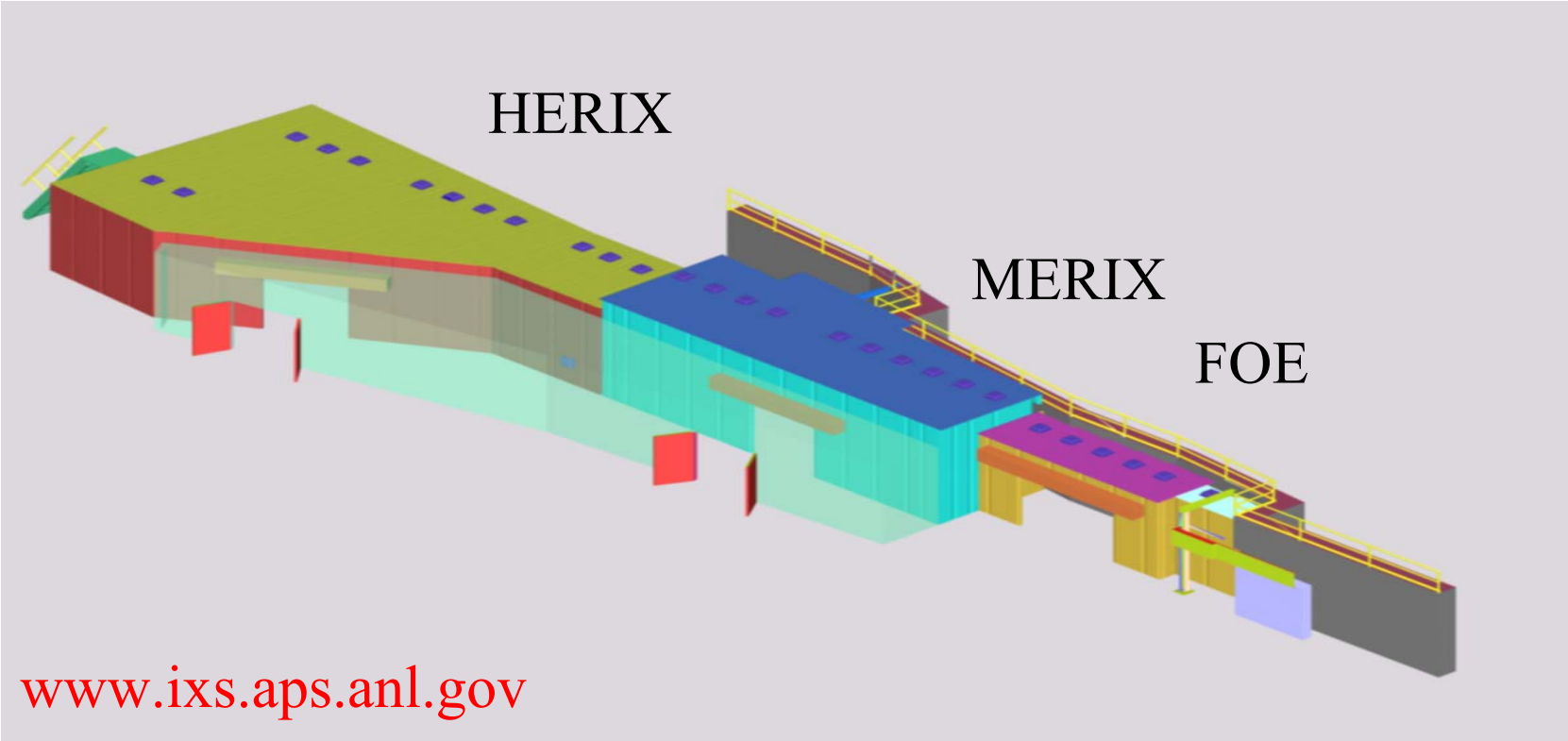
1 x meV line

1.5 x eV lines

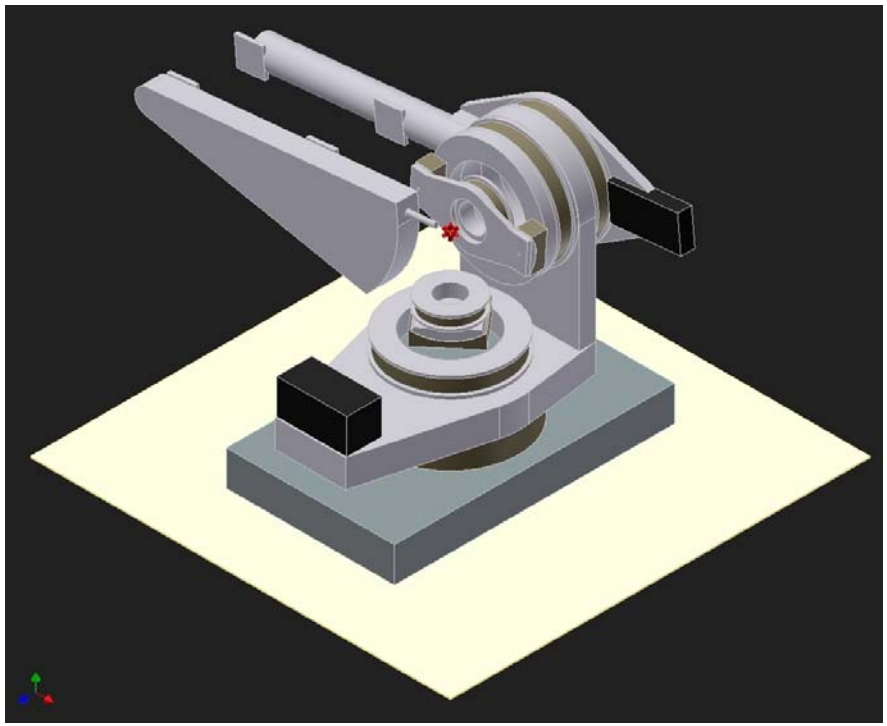
4.5m IVUN λ=3.2cm

Current State-of-the-Art: IXS-CDT

- Dedicated inelastic scattering beamline
- 100 meV endstation
- <1 meV endstation



MERIX



- 100 meV resolution.
- E_i= 4.9 – 11 keV.
- Vertical and horizontal scattering.
- Polarization selective.

- 2-3 x 2.4m undulators ($7x10^{19}$ ph/s/mrad^2/mm^2/0.1%bw @ 7 keV)

Operational in 3 years.

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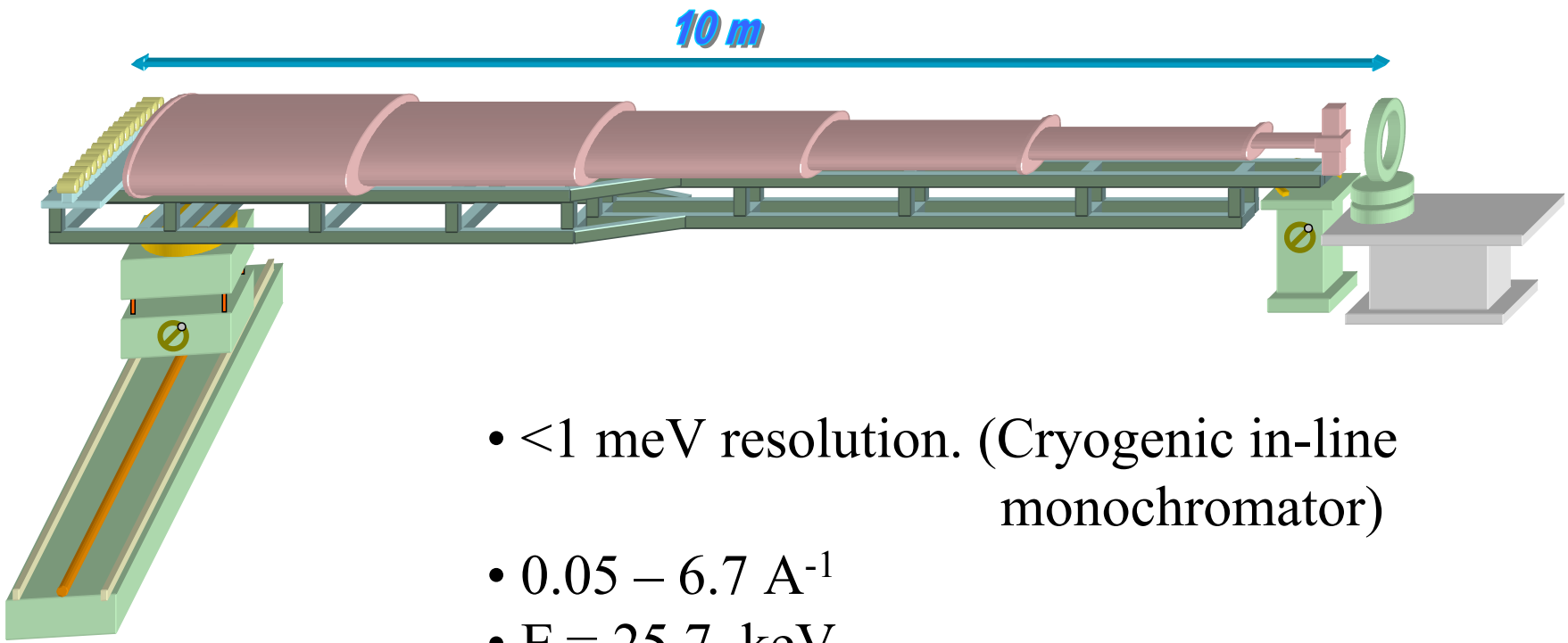
HERIX



- <1 meV resolution. (Cryogenic in-line monochromator)
- 0.05 – 6.7 A^{-1}
- E_i= 25.7 keV.
- Horizontal scattering (10m arm)
- 2.4m s.c. undulator

(10^{20} ph/s/mrad^2/mm^2/0.1%bw @ 21 keV)

Operational in 3 years.

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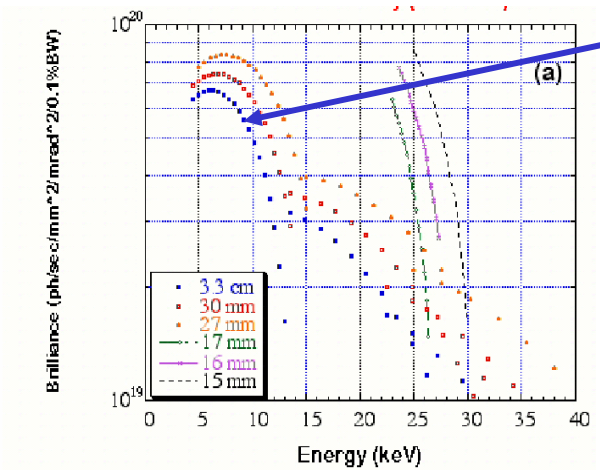
APS Undulator Curves

Medium Energy (5-10 keV)

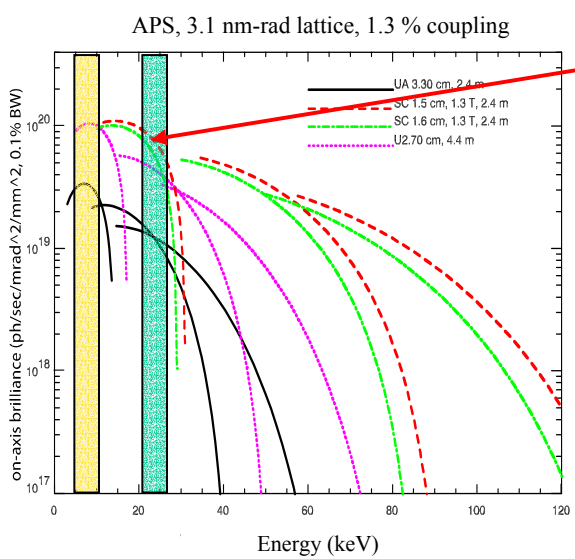


2 x Undulator A

I=100 mA
L=4.8 m

B~ 7 x 10^{19} ph/s/mrad^2/mm^2/0.1% BW

High Energy (21-25 keV)



SCU

L=2.4 m

B~ 10^{20} ph/s/mrad^2/mm^2/0.1% BW

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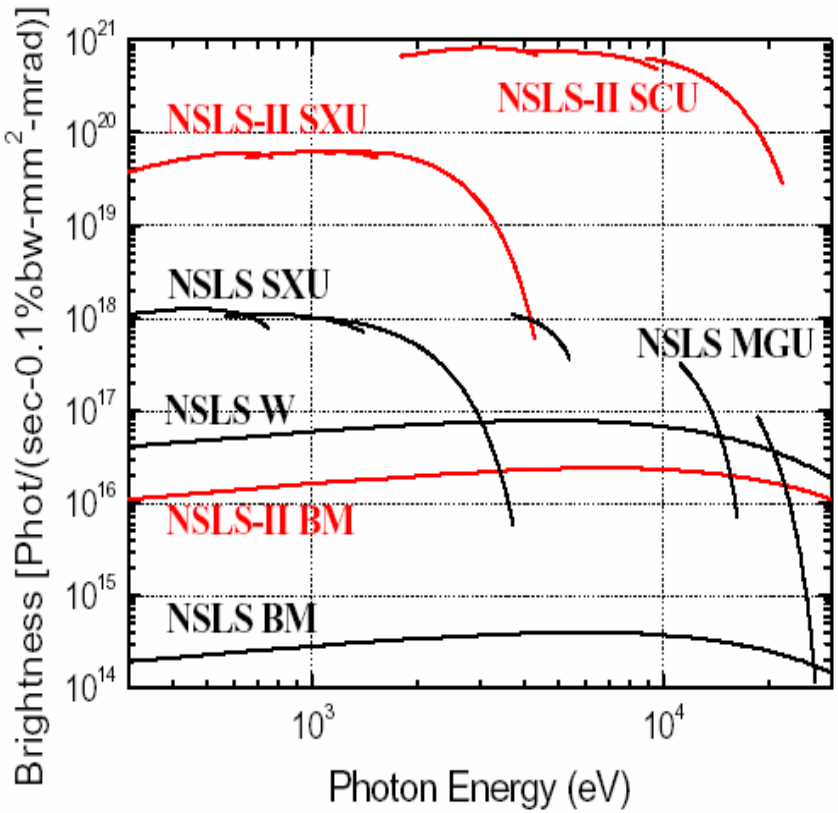
NSLS II Curves



NSLS II is 10 x brighter than APS extended straight section in medium energy

Will want as long a straight section as possible

I=500 mA

L=2.5 m

Flux proportional to L

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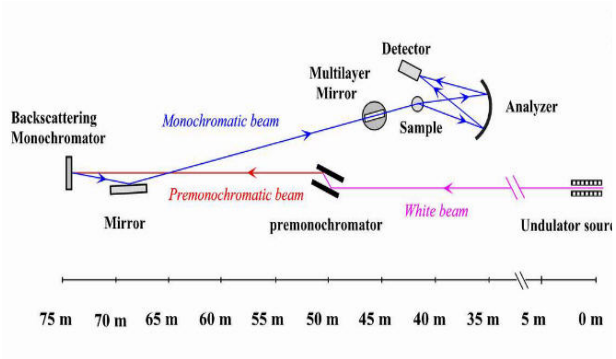
Monochromators

I. Back-Scattering



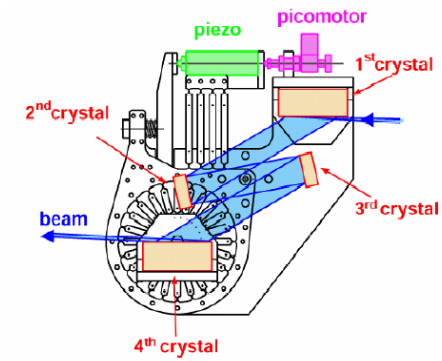
Advantages

- Proven design.
- Multiple resolutions.
- Efficient (?)

Disadvantages

- Must scan over elastic.
- Sample space constrained.
- Large footprint on floor.

II. In-line



Advantages

- Scan energy with a motor.
- Improved resolution tails.
- No encroachment on sample space.

Disadvantages

- Multiple bounces can cost efficiency
- Need new mono for each resolution
- Cryo-cooling ongoing R+D.

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Analyzers

Current technology is to dice and spherically bend Si or Ge

Works well for v. high resolution in back-scattering.

BUT….

- For RIXS, not always possible to find an appropriate reflection.
- Requires high-energy for high-resolution.

➡ Need for low symmetry analyzers

Possibly Al_2O_3?
Many reflections, e.g:
(5,2,6) ΔE=2.0 meV @ 9.8 keV

(also, quartz, semiconductor alloys…..)

➡ Polarization analyzers?

Si(333) θ_B=41° at 9 keV

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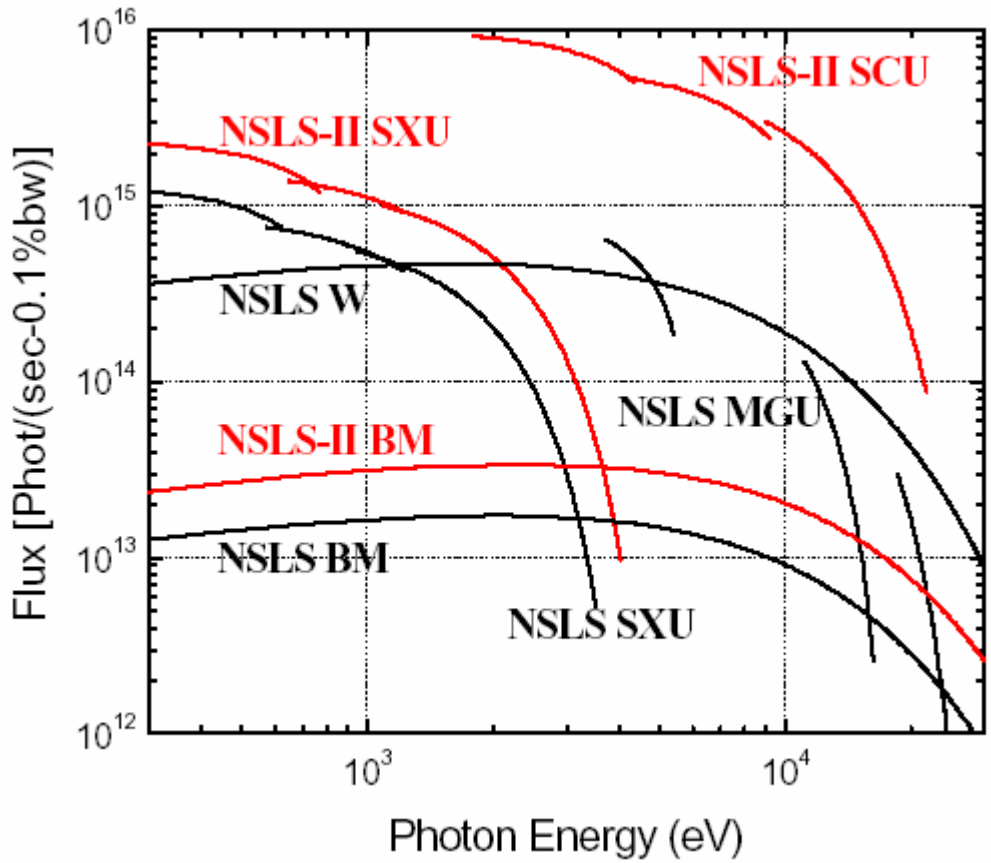
Summary

- IXS is an increasingly important tool in strongly correlated systems. There is a real need for more such facilities world-wide.

- NSLS II will give factor of 2 in flux over current state-of-the-art IXS facilities in medium energy regime and is competitive in the hard x-ray regime.

- A longer straight section to increase flux at the expense of brightness would directly benefit IXS experiments.

- Advances in analyzer design could cause significant gains, especially in the medium energy – high resolution regime.



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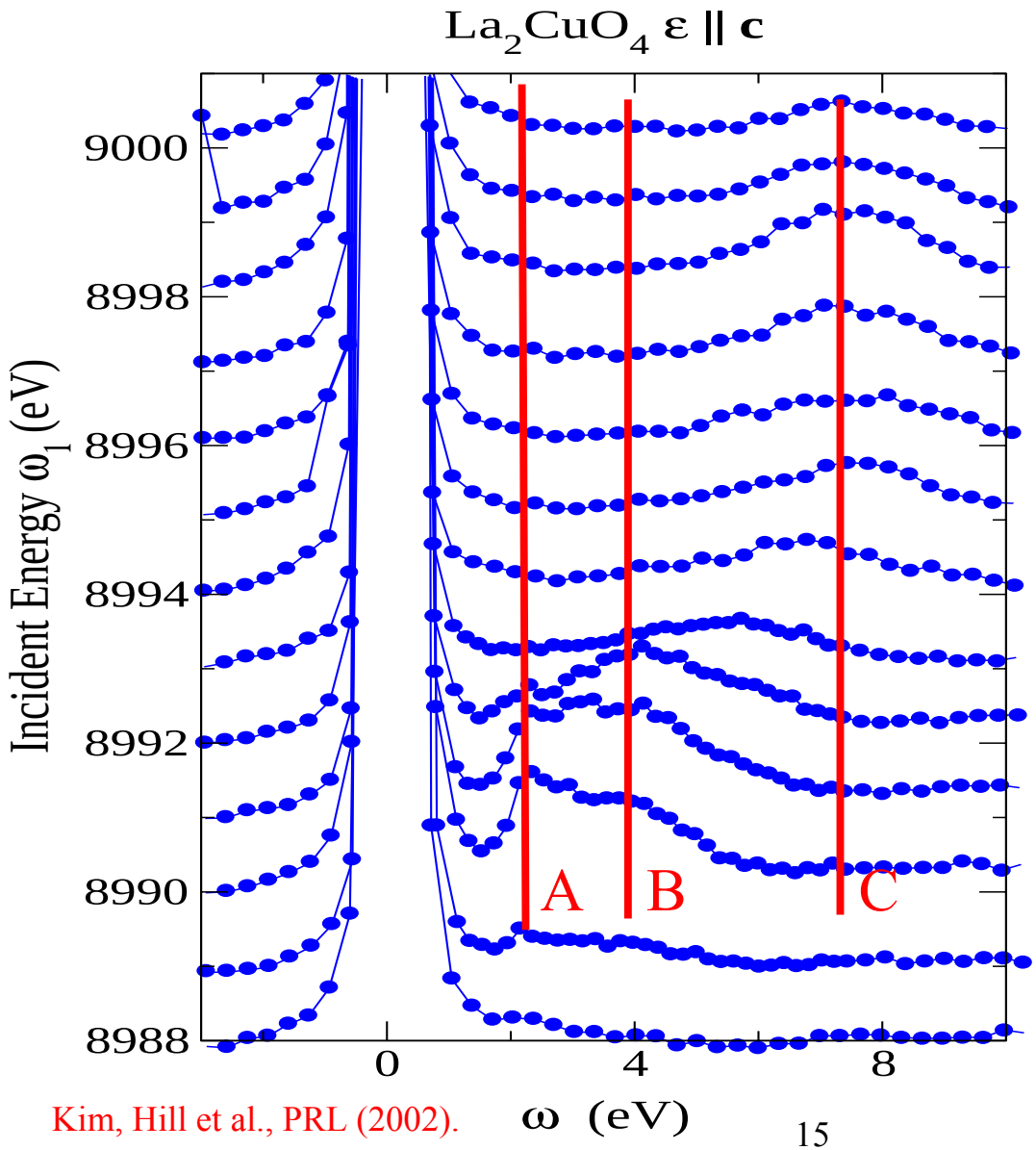
La_2CuO_4



Kim, Hill et al., PRL (2002).

Resonant Inelastic X-ray Scattering

Q=zone center

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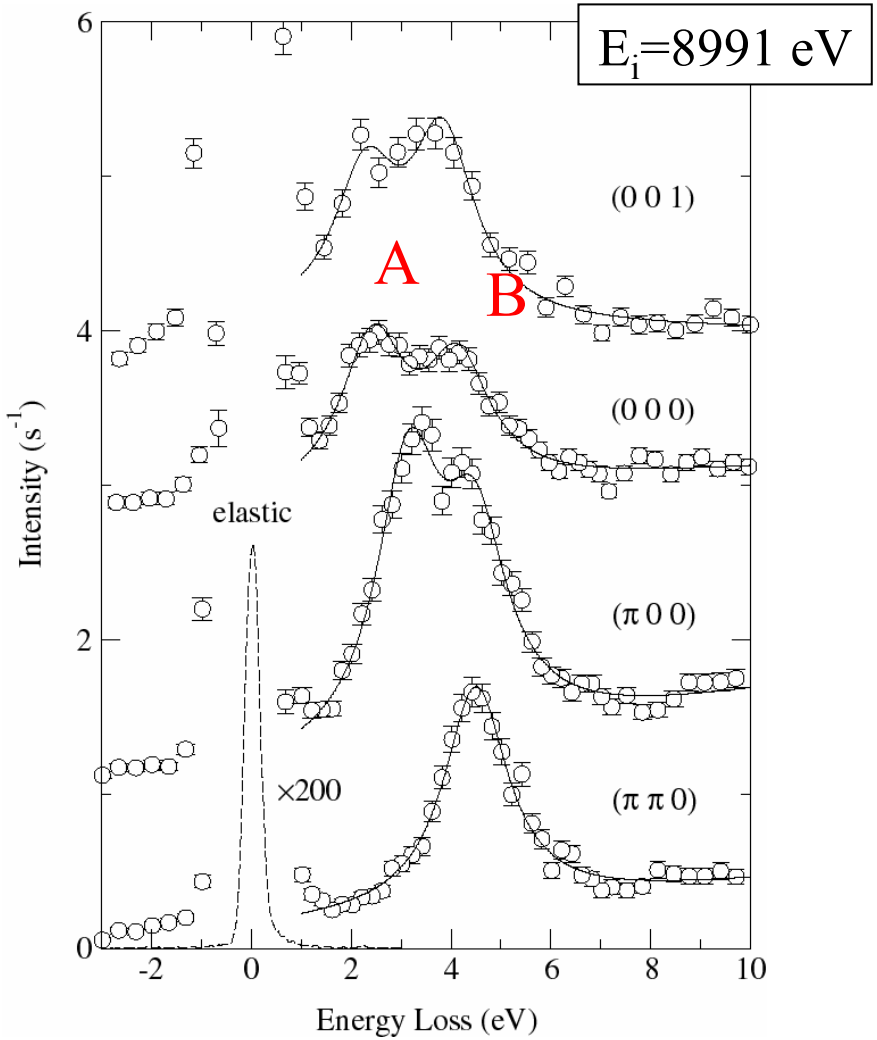
Gap Momentum Dependence



La_2CuO_4

Gap excitation shows structure and momentum dependence

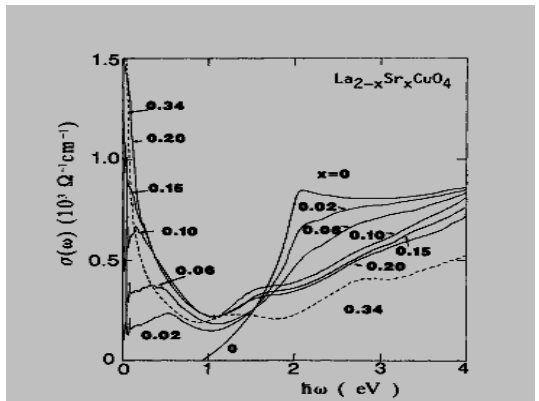


FIG. 7. Optical conductivity $\sigma(\omega)$ obtained from the Kramers-Kronig transformation of the *E*⊥*c* reflectivity spectra for various compositions *x*.

Uchida *et al*., PRB **43**, 7942 (1991)

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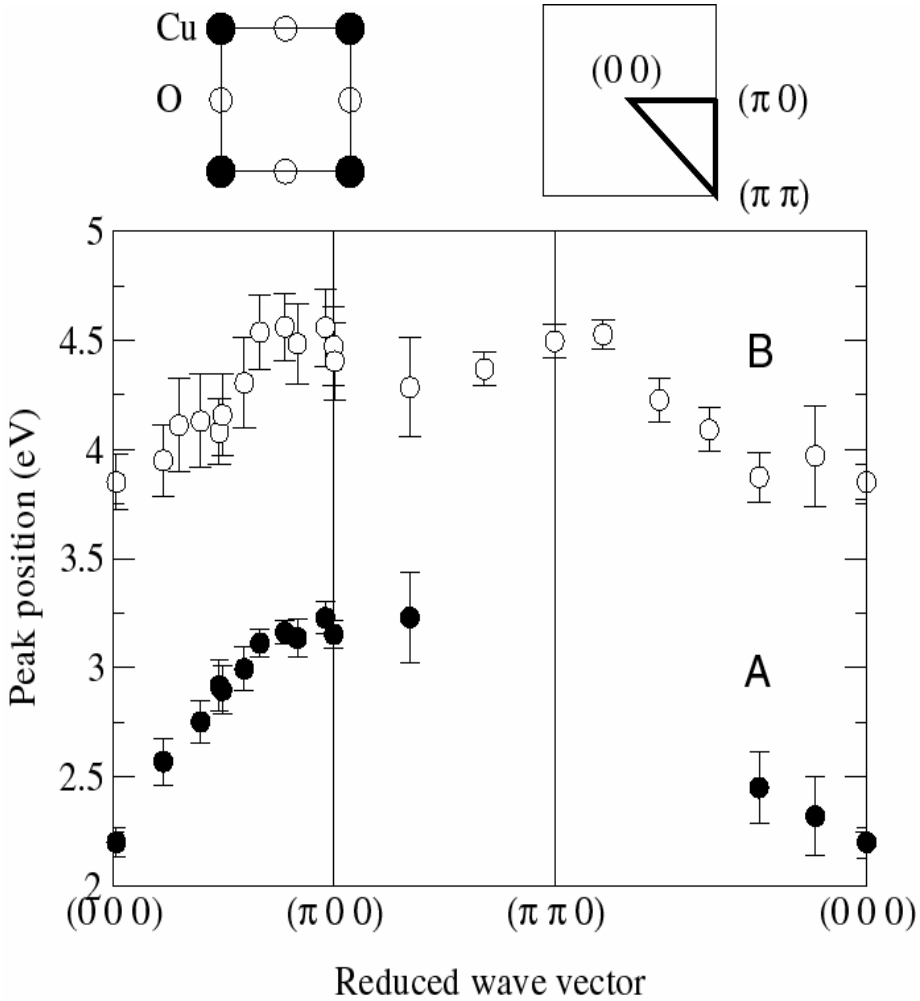
Dispersion Relations



Excitations A and B have;

- Large dispersion.
- Direct gaps.
- Momentum-dependent intensity (A).
- Large overlap with intermediate state.

→ Suggests they are *exciton-like*

Kim, Hill et al., PRL (2002).

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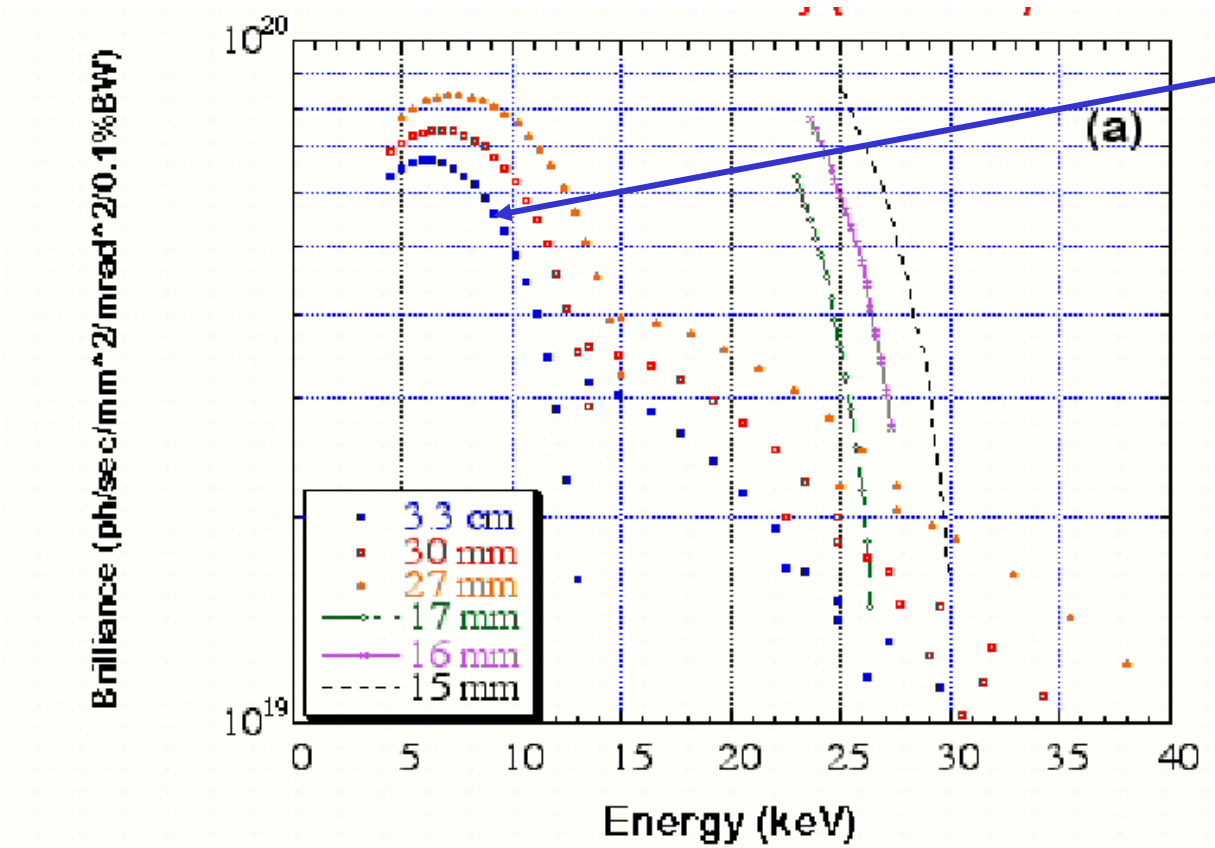
Undulators I



Undulator A

I=100 mA
L=9.6 m

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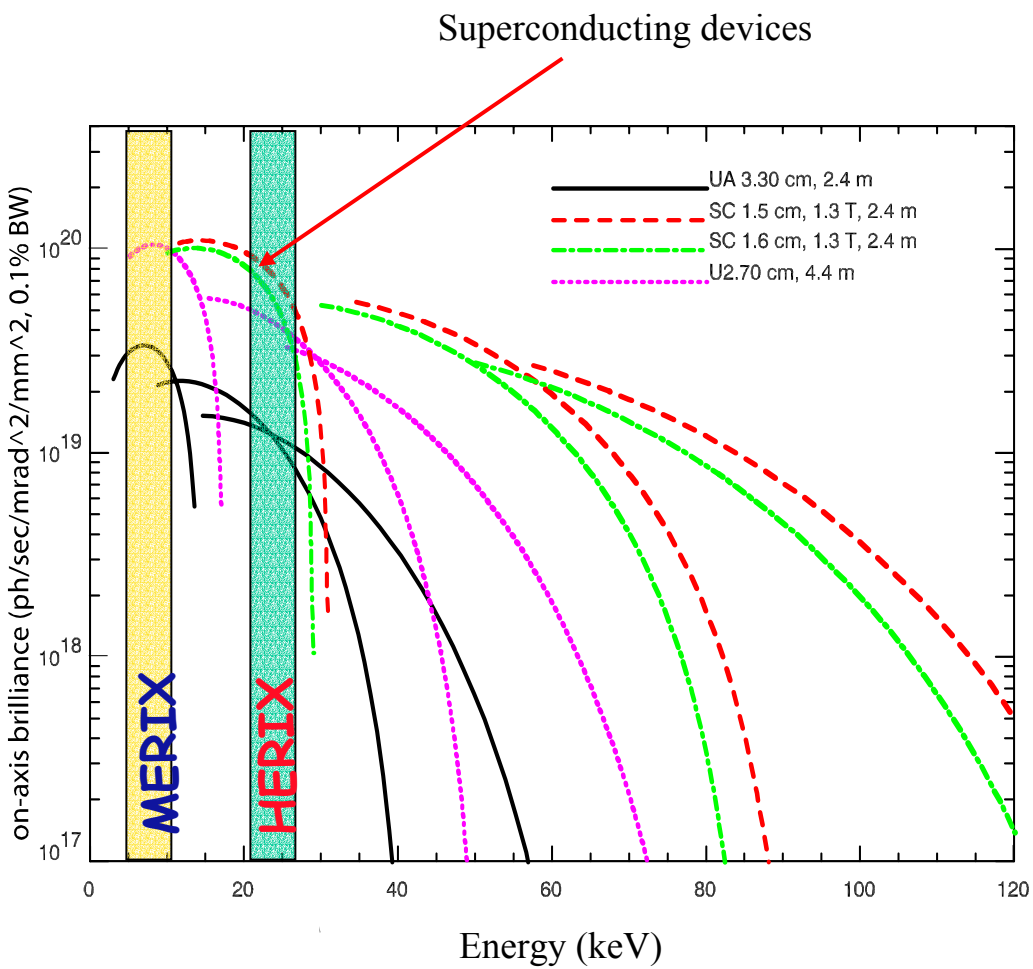
Undulators II



<u>HERIX Superconducting Device</u>

3.5 m long (2.5m magnet length)

14mm period

$1M delivery ~June 05

(Accel have built 8 period prototype and are currently building a 50 period device.)

<u>MERIX Device</u>

2-3 x Undulator A (33mm)

(Or 2-3 x 27mm device.)

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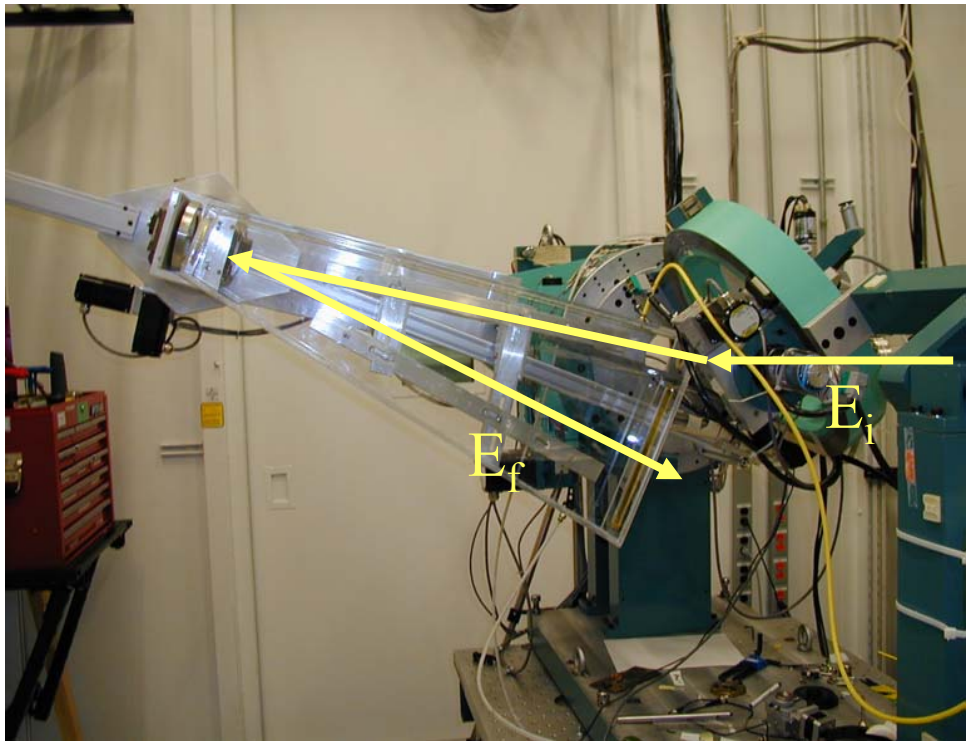
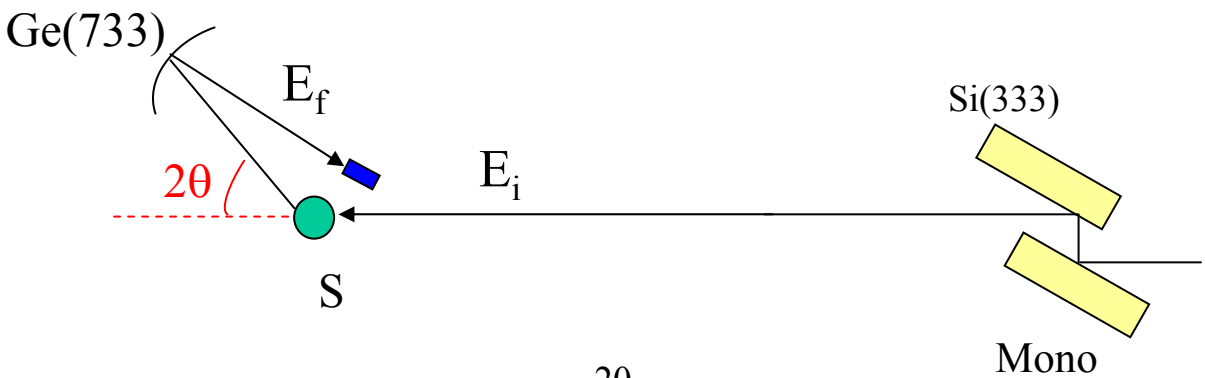
IXS Spectrometer at CMC-CAT



CMC-CAT 9IDB

Inelastic X-ray Scattering

- Measures two particle excitations.
- Is bulk sensitive.
- Can study insulators - and work in fields.



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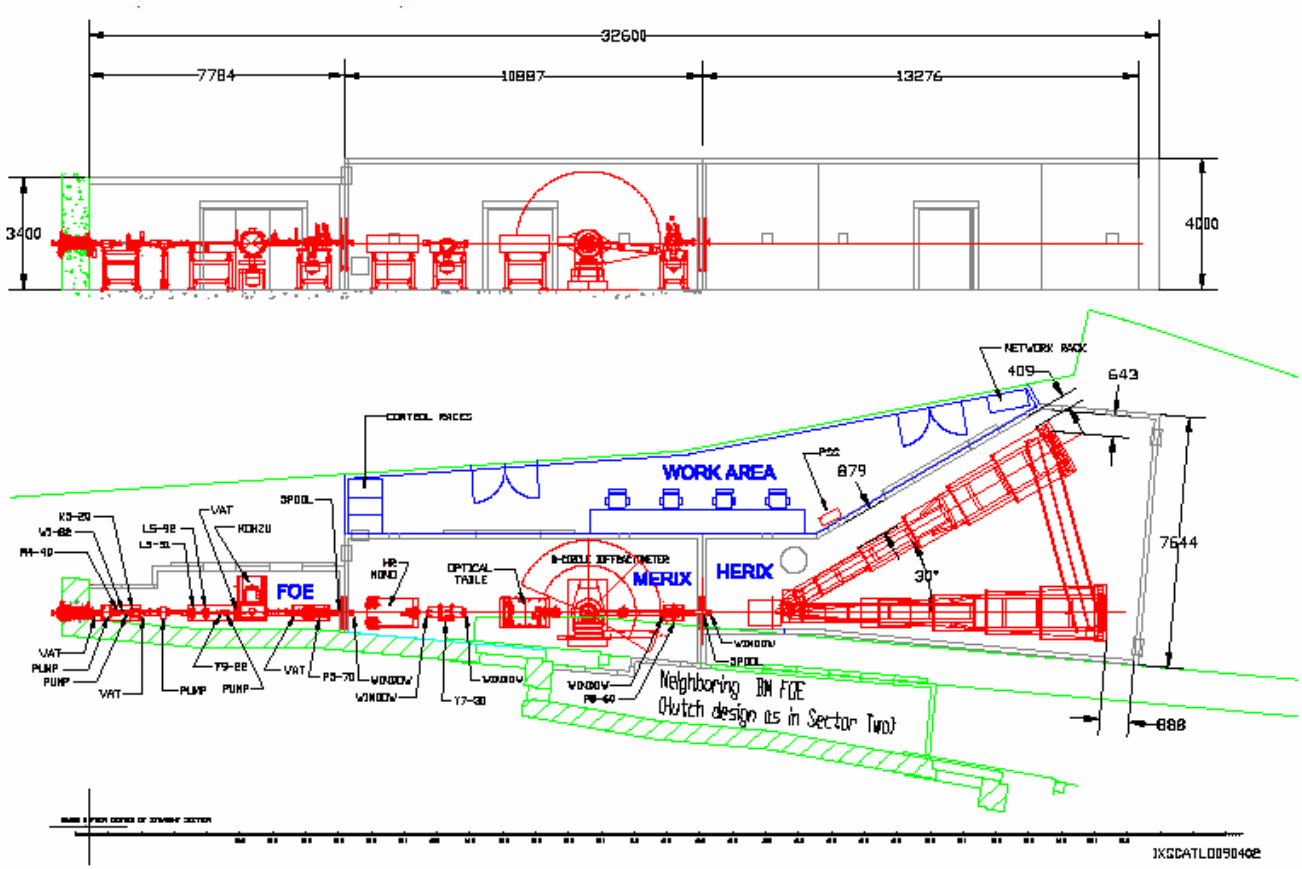
Inelastic X-ray Scattering Beamline

- Dedicated inelastic scattering beamline
- 100 meV endstation
- <1 meV endstation

www.ixs.aps.anl.gov



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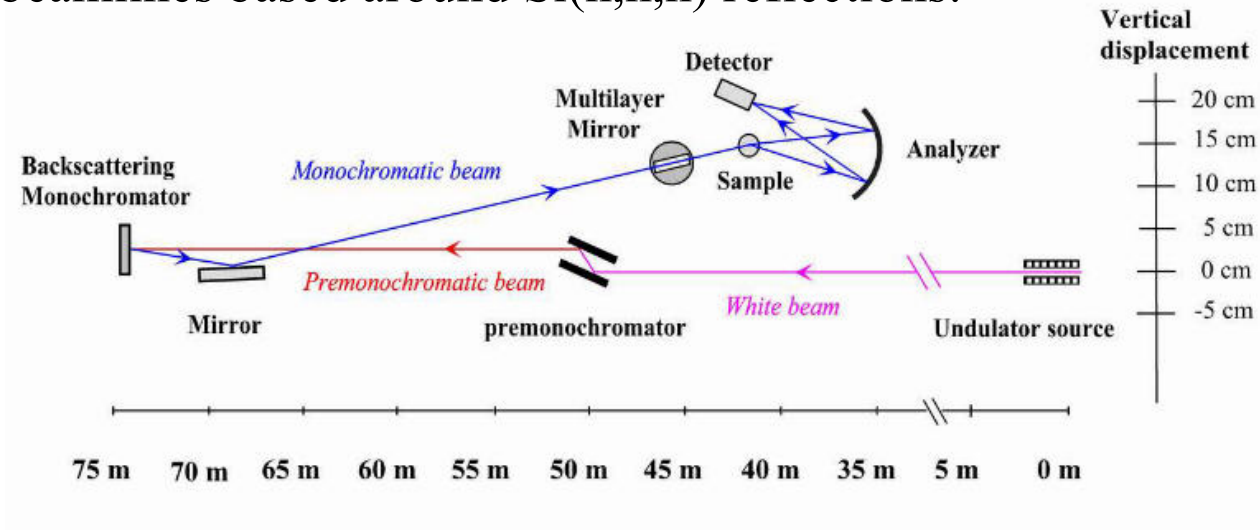
Monochromators I: Back-Scattering

ESRF and SPring-8 IXS beamlines based around Si(n,n,n) reflections:



Advantages

- Proven design.
- Multiple resolutions.
- Efficient (?)

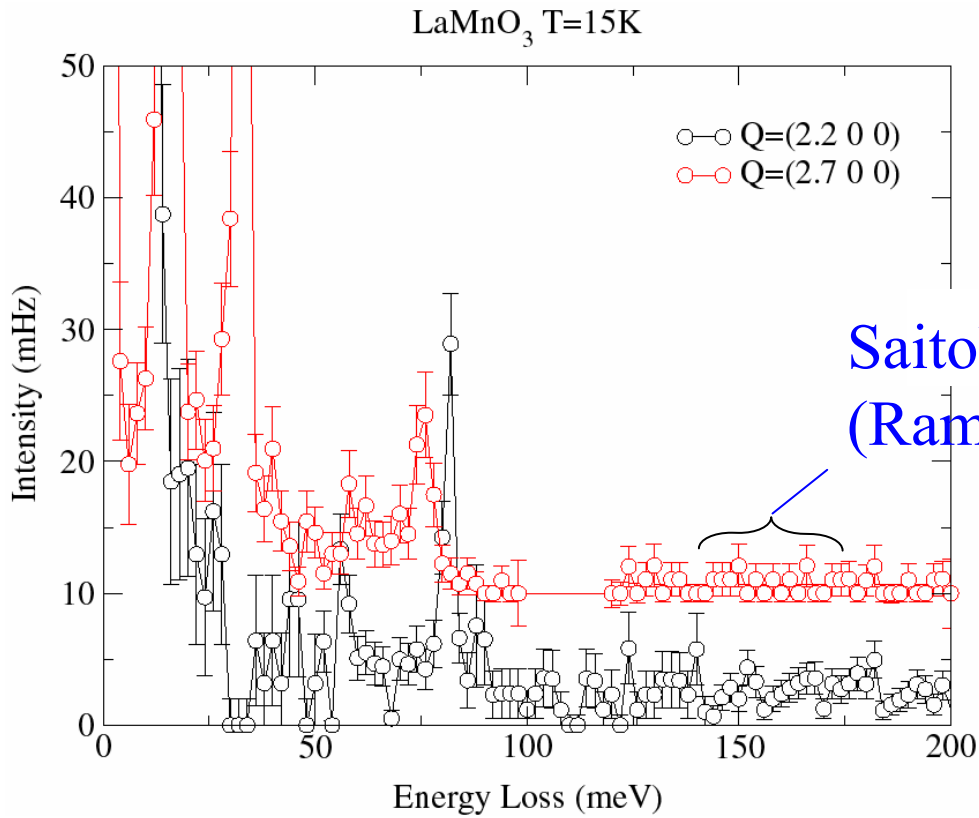
Disadvantages

- Must scan over elastic.
- Sample space constrained.
- Large footprint on floor.

Reflection	Energy	Inc. Resolution	Flux
(7,7,7)	13.8 keV	5.3 meV	1×10^{11}
(9,9,9)	17.8 keV	2.2 meV	2×10^{10}
(11,11,11)	21.7 keV	0.8 meV	7×10^{9}
(13,13,13)	25.7 keV	0.5 meV	2×10^{9}

Orbitons?

Non-resonant Inelastic X-ray Scattering



Saitoh *et al.* (Raman)

2 meV resolution

3ID, APS

Kim, Hill, Nelson, Alp, Sinn (2001).

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