Inelastic X-ray Scattering Spectroscopy for Material Excitations

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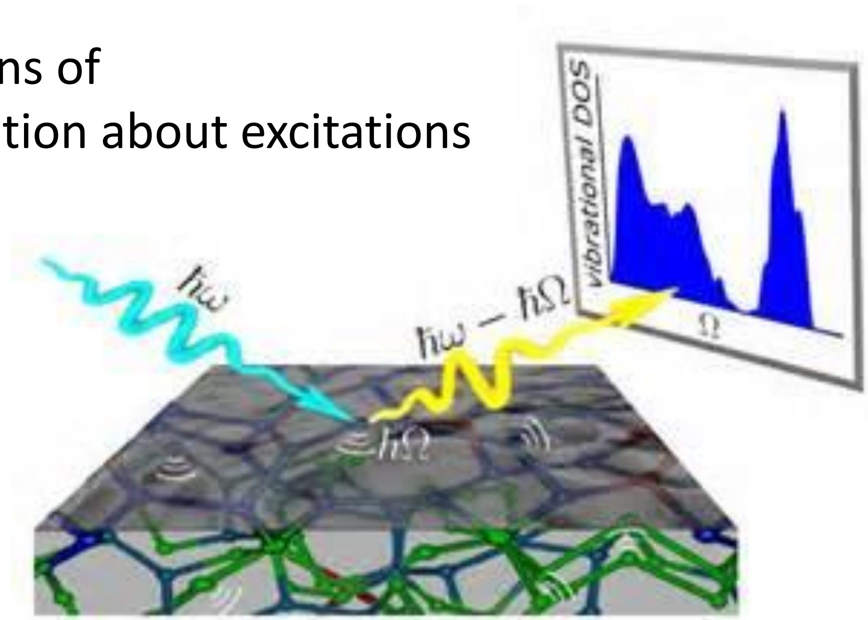
Outline

- IXS & Low Energy Excitations in Materials
- Why Synchrotron Radiation?
- Resonant IXS
- Typical Beamlines for RIXS
- IXS for Dynamics
- Typical Beamline of IXS
- Summary

IXS

Inelastic X-ray Scattering   Spectroscopy

Photon in-Photon out spectroscopy
Hard & Soft x-ray energy range
Energy, Momentum & polarizations of
outcoming photons gives information about excitations



Resonant IXS (RIXS)
The incoming photon resonates by absorption edges

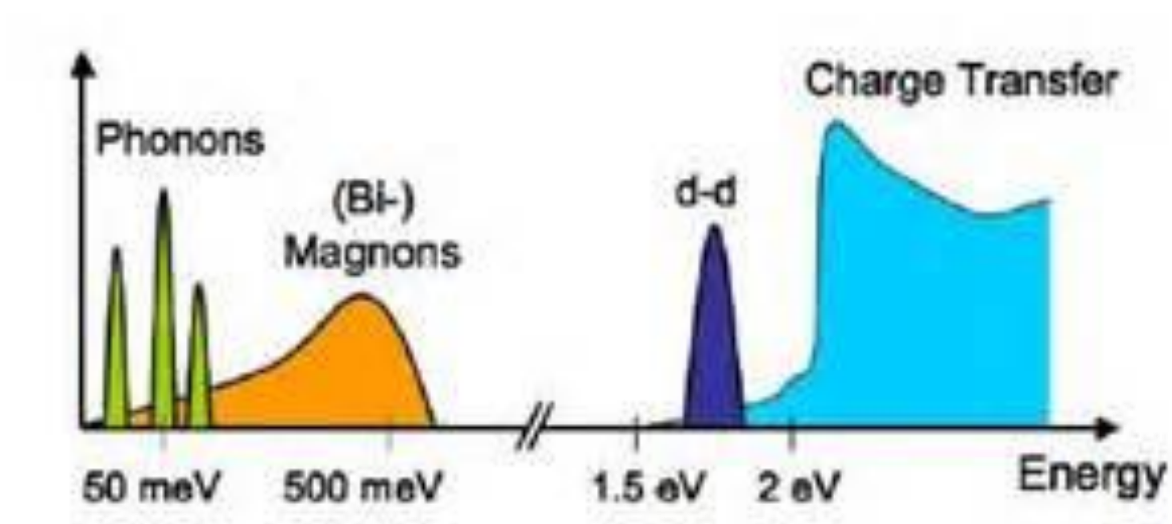
Non resonant IXS (IXS)
Scatters from Charge density distribution

Excitations

Low Energy Excitations: meV-eV

Electronic: d-d, charge transfer
Dynamic: phonons
Magnetic: magnons

Measuring the energy-momentum dispersion of these excitations gives very important microscopic information of the sample under studies

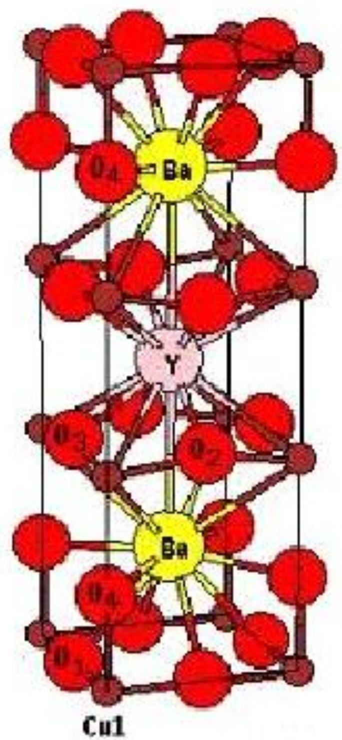


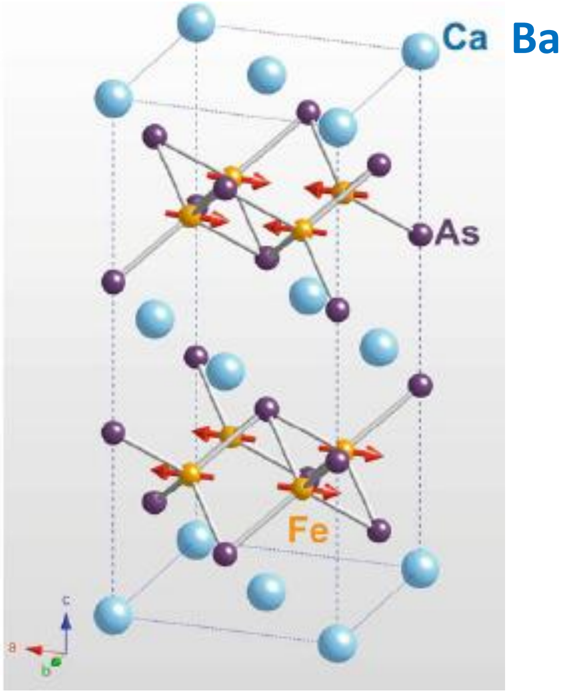
Fields of Study

Condensed Matter Physics, Material Science
Understand the physical properties like
Transport properties,
Quantum state,
Phase transitions,
Superconductivity mechanism,
Charge Ordering Phenomenon, etc

Interests
Unconventional Superconductivity,
High Tc superconductivity: cuprates & Fe based
Manganite, Niclates
Strongly Correlated Materials, Mott-Hubbard Insulators
Semiconductors, etc

Samples



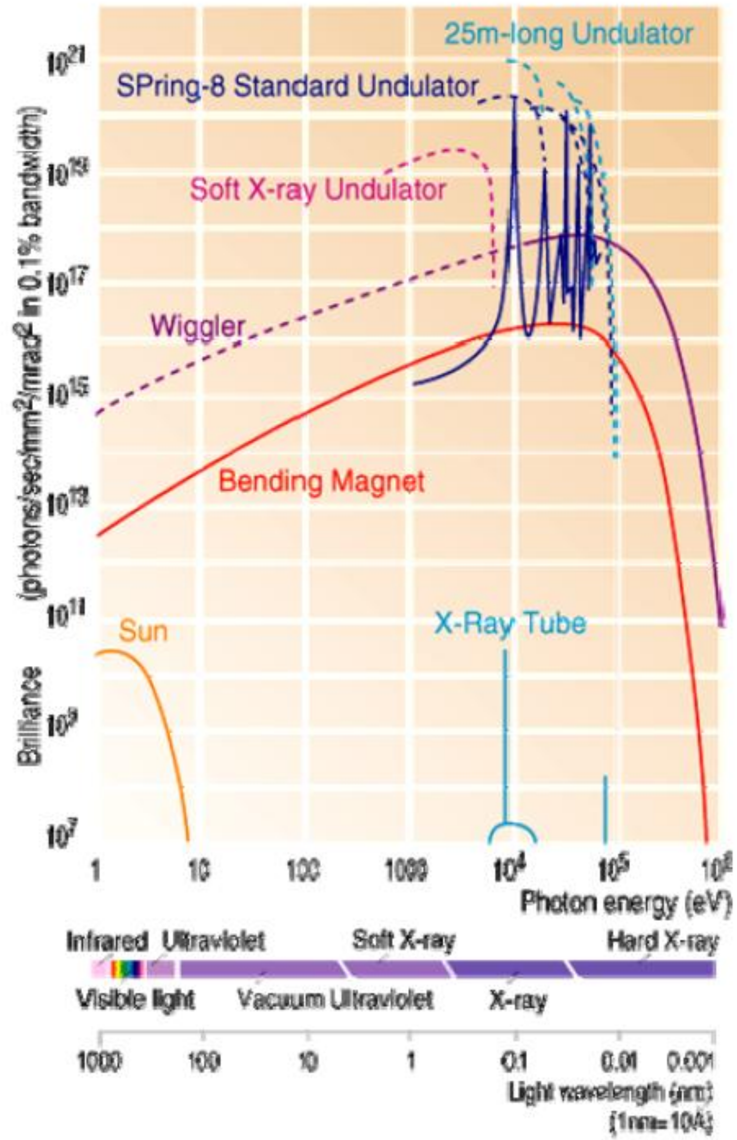


YBCO High Tc
Superconductor
T_c~90 K

Fe based Superconductor
T_c~40 K

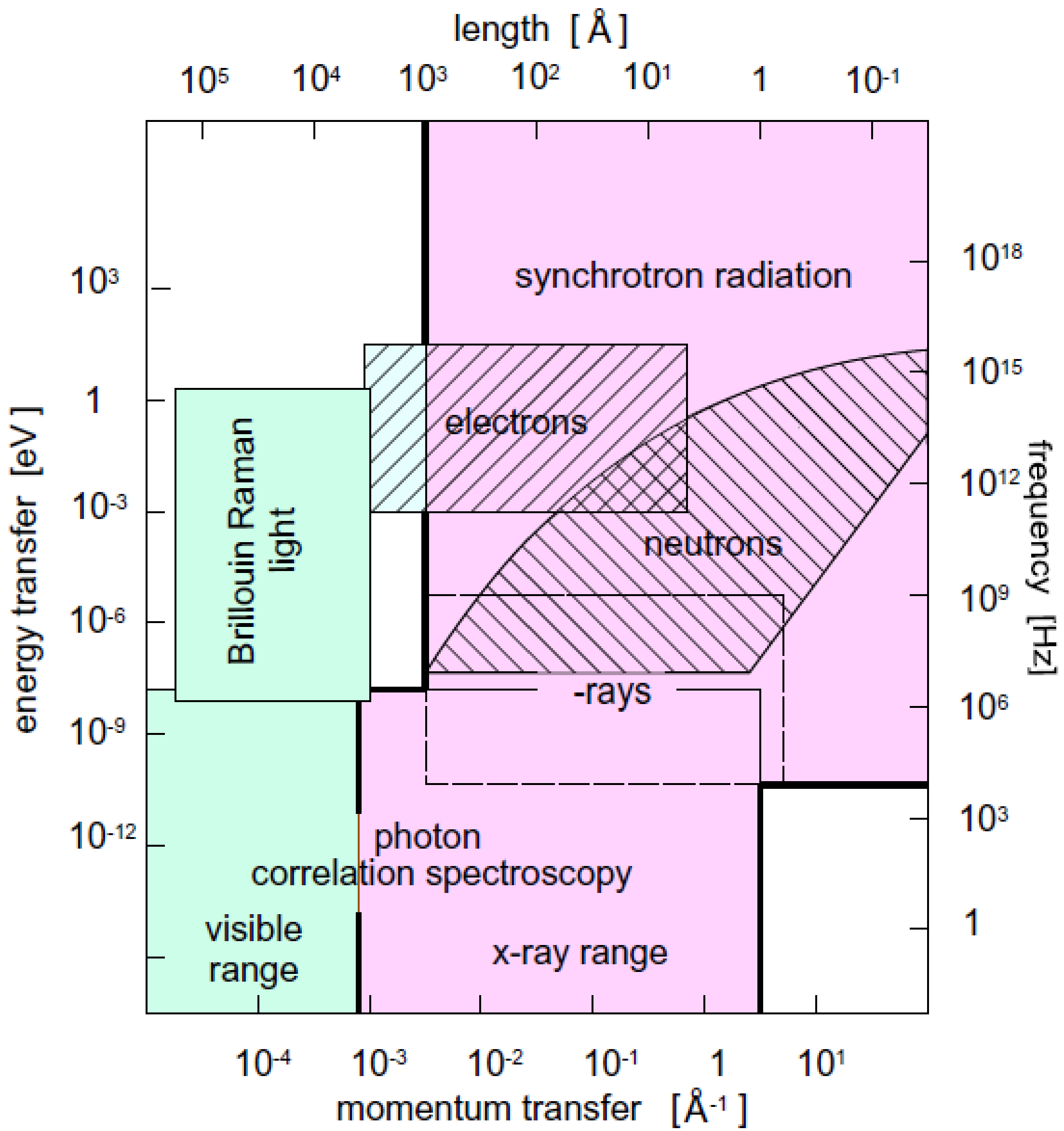
Synchrotron Radiation

SPring-8 Japan 1997
8 Gev, 1436 m



- Very High Brightness Source
- Very high photon flux
- Collimated beam
- Tuning the incoming energy
- Possible to reach very high energy resolution ($\Delta E/E \sim 10^{-8}$)

Energy-momentum of Synchrotron Radiation



Resonant Inelastic X-ray Scattering

REVIEWS OF MODERN PHYSICS, VOLUME 83, APRIL–JUNE 2011

Resonant inelastic x-ray scattering studies of elementary excitations

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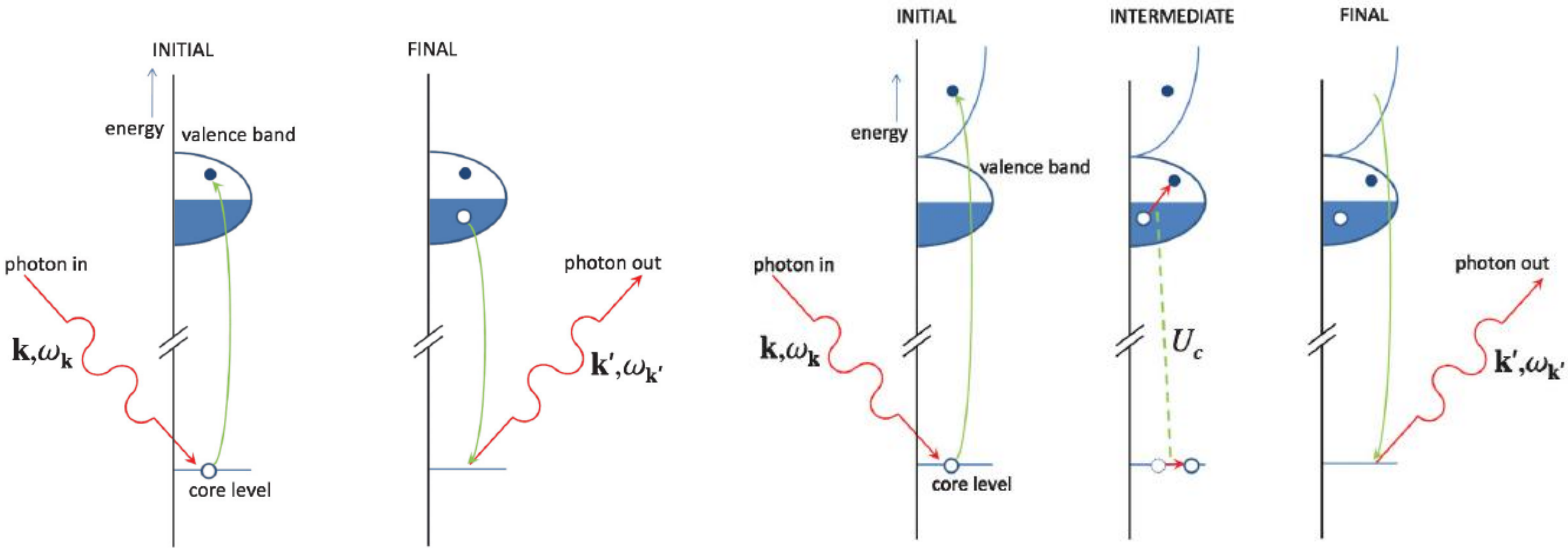
In the past decade, resonant inelastic x-ray scattering (RIXS) has made remarkable progress as a spectroscopic technique. This is a direct result of the availability of high-brilliance synchrotron x-ray radiation sources and of advanced photon detection instrumentation. The technique's unique

Physical Process

Resonant Inelastic X-ray Scattering (RIXS)

Direct RIXS

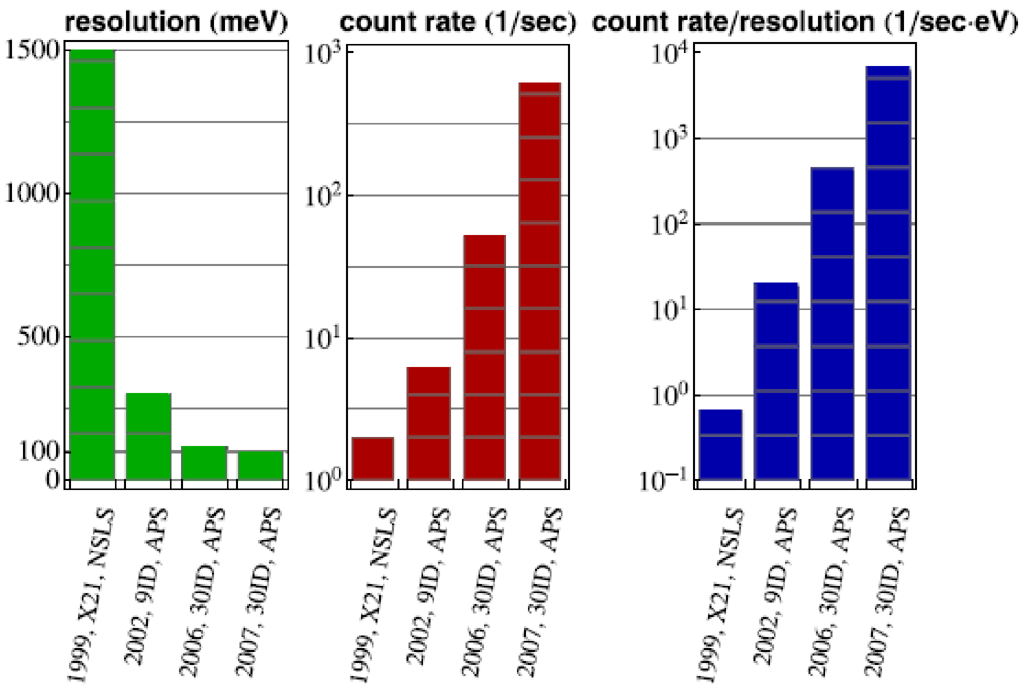
InDirect RIXS



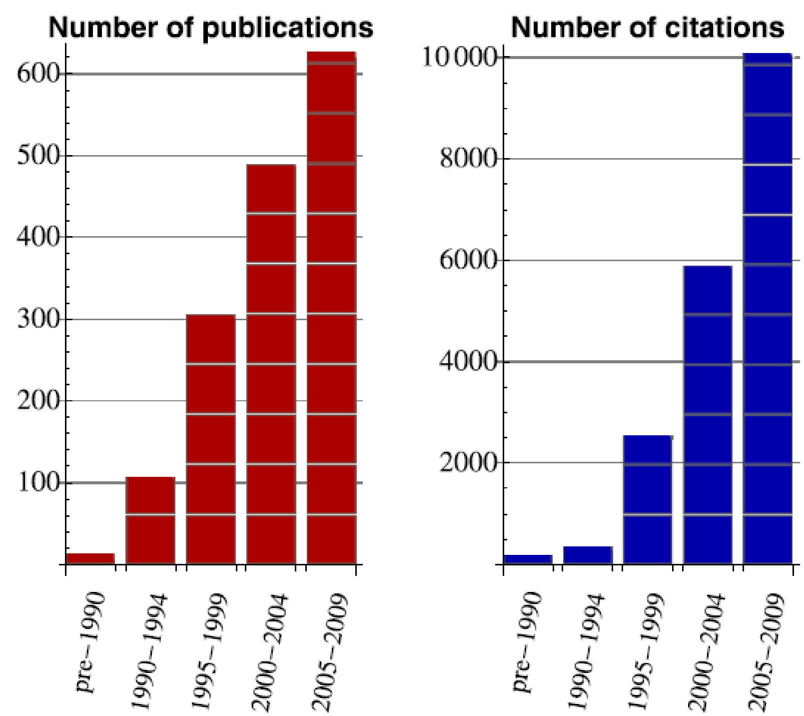
Schematic picture is understood simply
But measuring data and interpreting the data has many challenges

Many works has been done to reach the state of the art of this measurement using synchrotron radiation

Progress

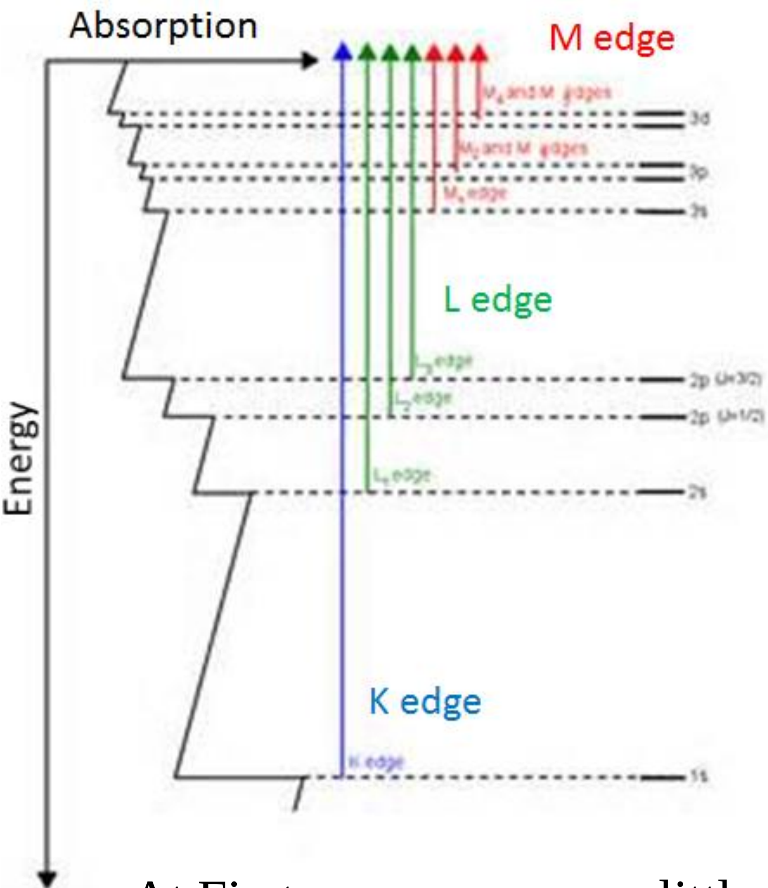


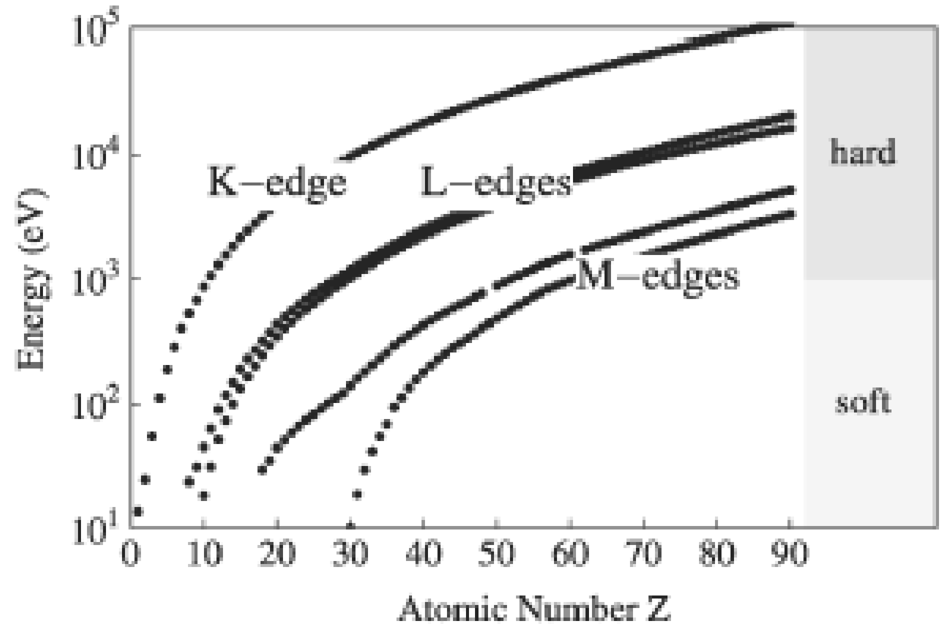
Increasing number of publications and citations



Energy Resolution improvements
Count Rate increase
Count rate/resolution increase

Decision for Absorption Edge





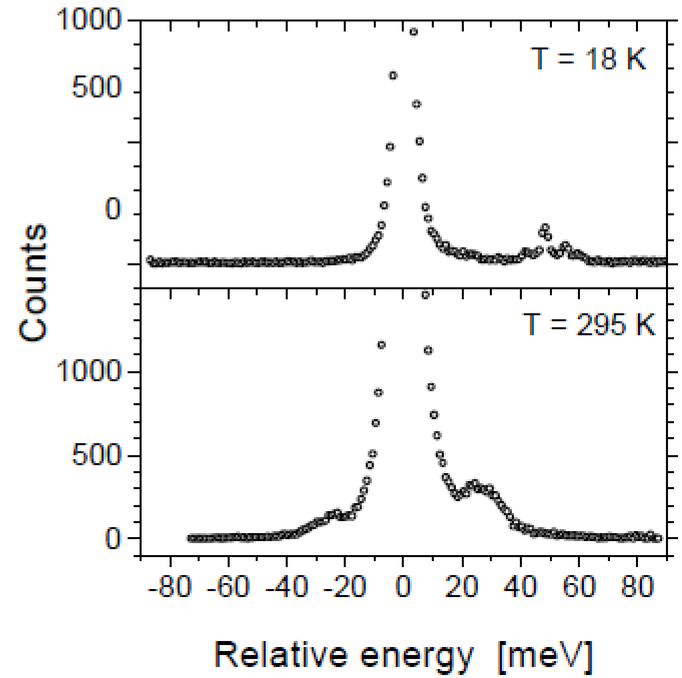
At First we may assume little difference between different Edge, but it is not!!

- Intensity changes with dioplar or quadropole transitions
- Using higher energy lets scans higher momentum transfer

1s to 4s transition is dipole, strong intensity and large momentum transfer
Incident photon energy is tuned by energy scan measurement of absorption edge

Reduction of Elastic Peak

Tail of the Elastic Scattering peak can hide or disturb the low energy inelastic peaks
So it has to be reduced, determined and subtracted from the measured data



Main Contributions of the elastic peak

- Phonon

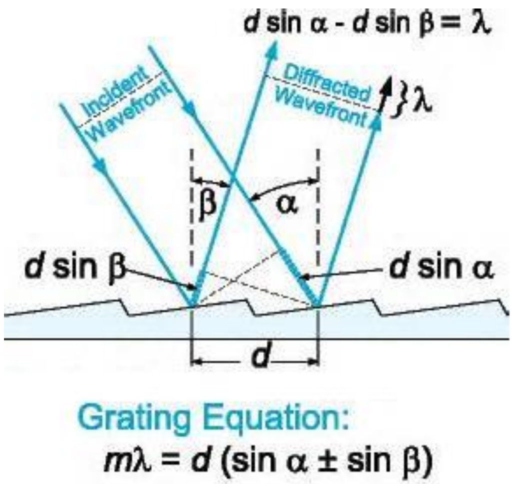
as much as 50 % of the peak
Reduce the temperature as much as possible
In long momentum transfer intensity α $(q.\delta)^2$

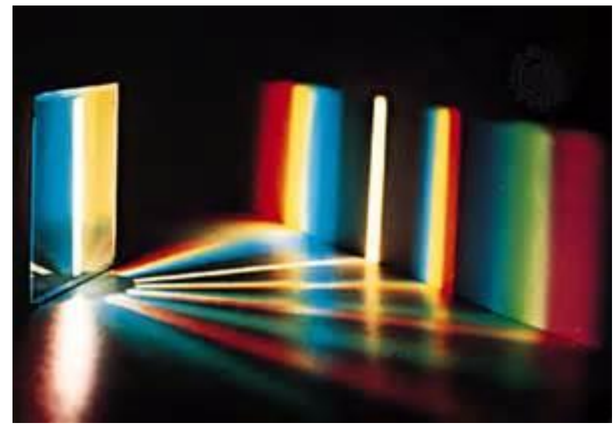
- Diffuse scattering due to the strain & defects

Energy & Resolutions

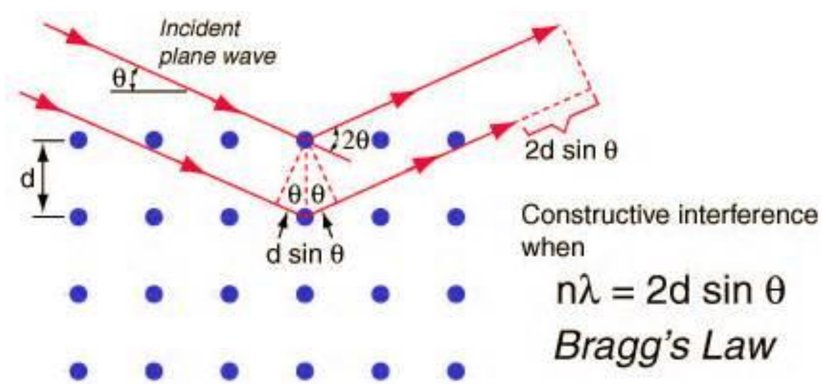
Energy transfer: Less than a few electron volts
Energy Resolution: less than 100 meV
For phonons ~meV

Soft X-ray: diffraction grating





Hard X-ray: highly ordered crystalline lattice of Si, or Ge



RIXS Instrument

Experimental Instrument:

1- Beamline: deliver a well collimated, highly monochromatic, and often focused beam at sample

2- Spectrometer: sample environment, manipulate scattering angle, collect scatter radiation, energy analyzer

Two typical examples

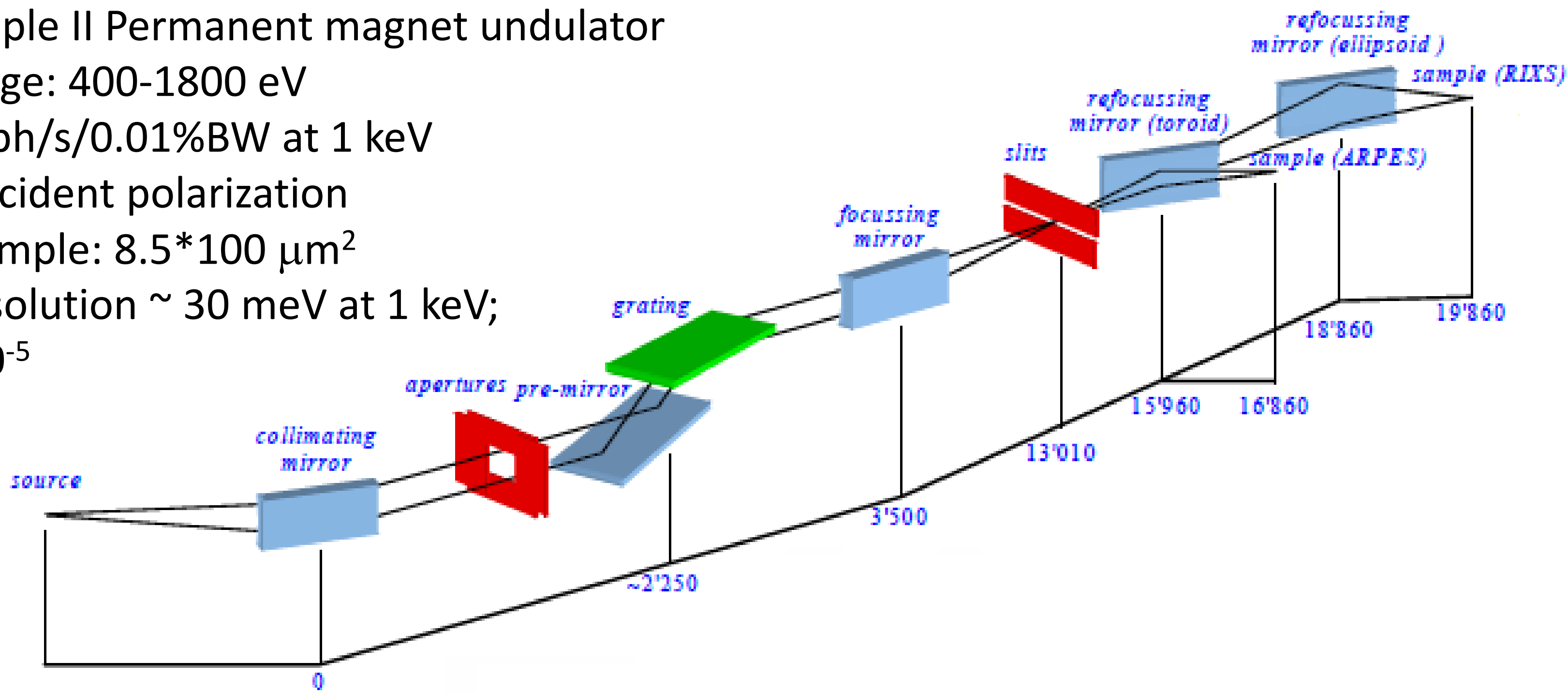
Soft x-ray: ADRESS beamline and SAXES spectrometer

Hard x-ray: MERIX-HERIXS bemline and MERIX spectrometer

ADRESS Beamline

ADvanced RESonant Spectroscopies (ADRESS) Beamline
Swiss Light Source (SLS)
For RIXS and ARPES studies on correlated & nano-strucured materials

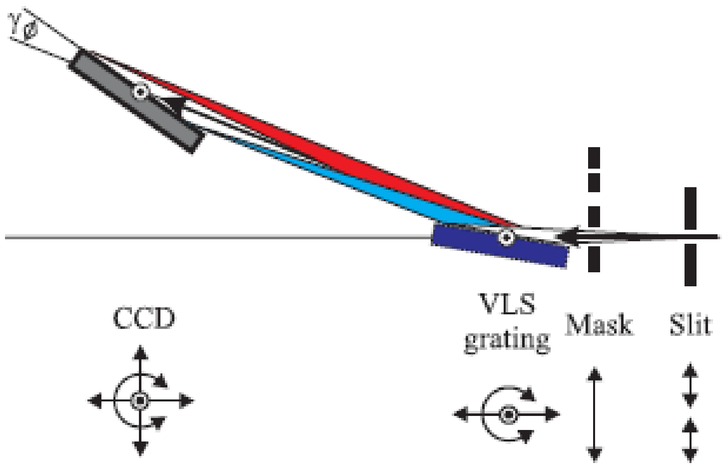
Source: Apple II Permanent magnet undulator
Energy range: 400-1800 eV
Flux: 10^{13} ph/s/0.01%BW at 1 keV
Variable incident polarization
Spot on sample: 8.5*100 μm^2
Energy Resolution ~ 30 meV at 1 keV;
$\Delta E/E=3*10^{-5}$



Collimating mirror
Plane grating monocromator (PGM): line spacing 4200 mm^{-1}
Focusing mirror & Refocusing mirror
Sample
SAXES Spectrometer

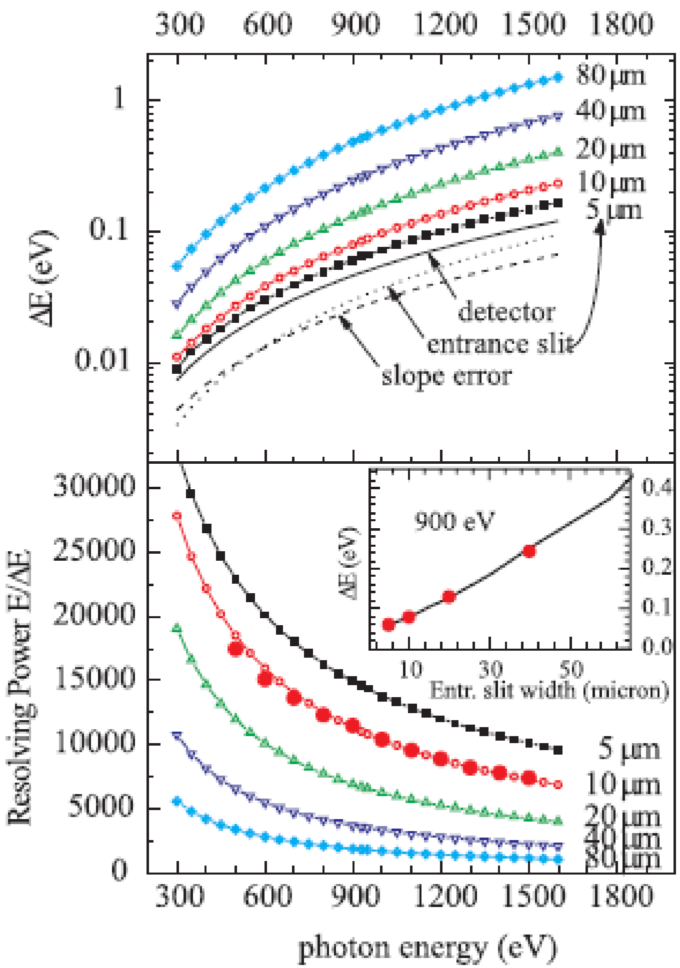
SAXES Spectrometer

SuperAdvanced X-ray Emission Spectrometer (SAXES)



Spherical Variable Line Space (VLS) grating rather than a plane grating and concave collection mirror (minimize higher order optical aberrations in cost of reduced collection angle)
3200 mm^{-1}, R=58.55 m,

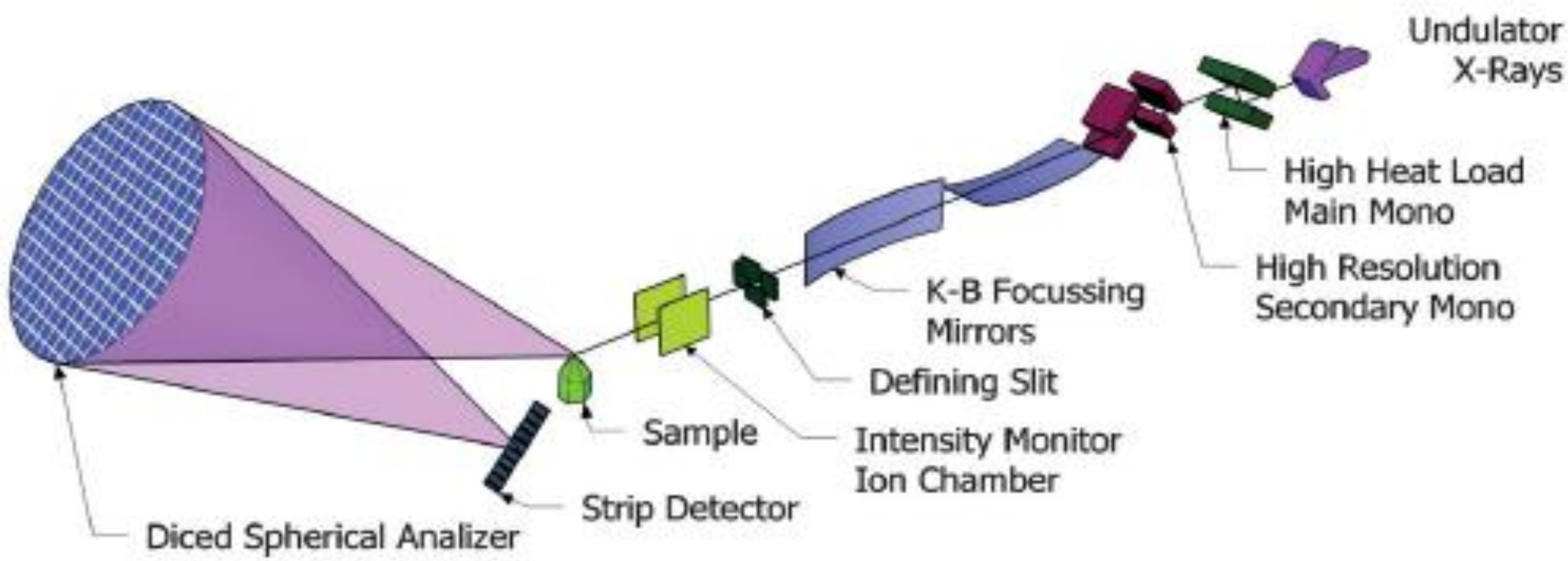
CCD detector
Pixel size: 13.5*13.5 μm^2
Total resolving power ~ 10,000 in the whole energy range



Calculated performance of SAXES

MERIX Beamline

Example: BL30 ID
APS, USA
MERIX (Medium Energy Res. Inelas. X-ray)
HERIX (High Energy Res Inelas. X-ray)



Source: two linear polarized Und, 2*2.5 m long, 30 mm period
Energy Range~ 4.9-15 keV
Energy Resolution ~ 70 meV; $\Delta E/E=1*10^{-5}$
Flux at sample (ph/s)=$2*10^{12}$ at 9 keV; Spot size=45*6 μm^2-2.2*0.4 mm^2

MERIXS Beamline

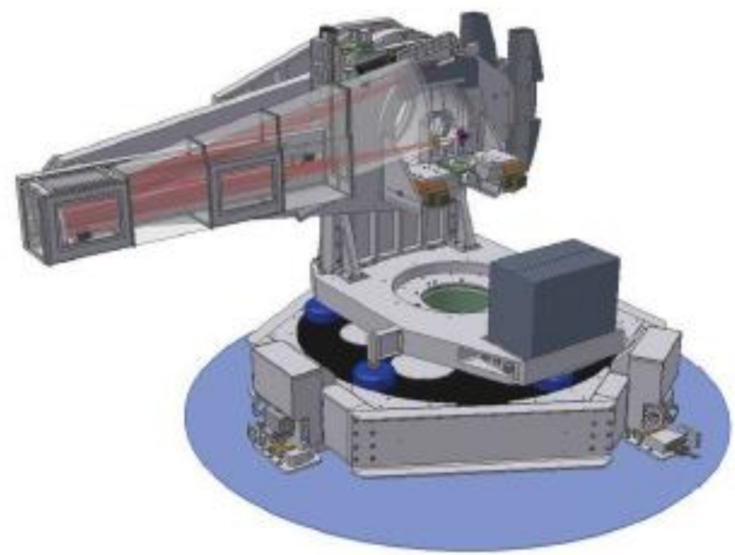
Monochromator: water cooled diamond(111) crystal

Secondary Monochromator: four asymmetrical cut Si(111) crystal, zero offset
Constant band pass in the whole energy range~ 70 meV

Quickly Switch between
high resolution narrow band with low flux to & low resolution and high flux

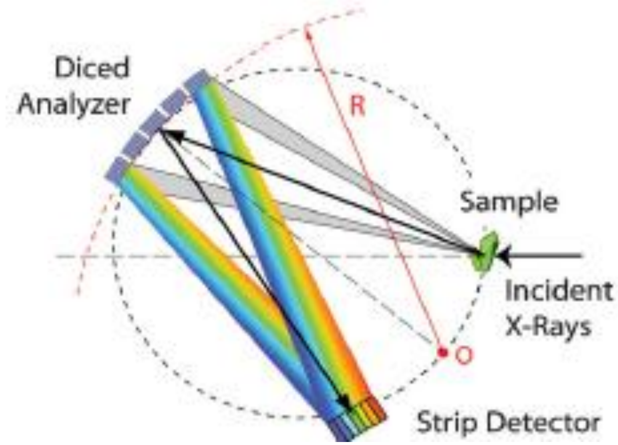
KB mirror to provide horizontal and vertical focusing

MERIX Spectrometer for analyze and
detect the photons
Used in
30 ID, APS
9ID, APS (2007)
ID16, ESRF (2009)
11XU, SPring-8 (2001)



MERIX Spectrometer

MERIXS & Total Energy Resolution



2 m detector arm Moves horizontally and vertically
Rowland circle geometry

Analyzer is situated in He gas tank
To minimize the absorption and background scattering

10 cm spherically bend, diced Ge(733) Bragg reflection at Cu K edge

Energy Resolution contribution:

- Intrinsic Darwin width
- Johan error in horizontal and vertical direction (increases quadratically with r/R)
- Finite beam footprint on the sample
- Finite block size (reduced linearly with size by using diced analyzer)

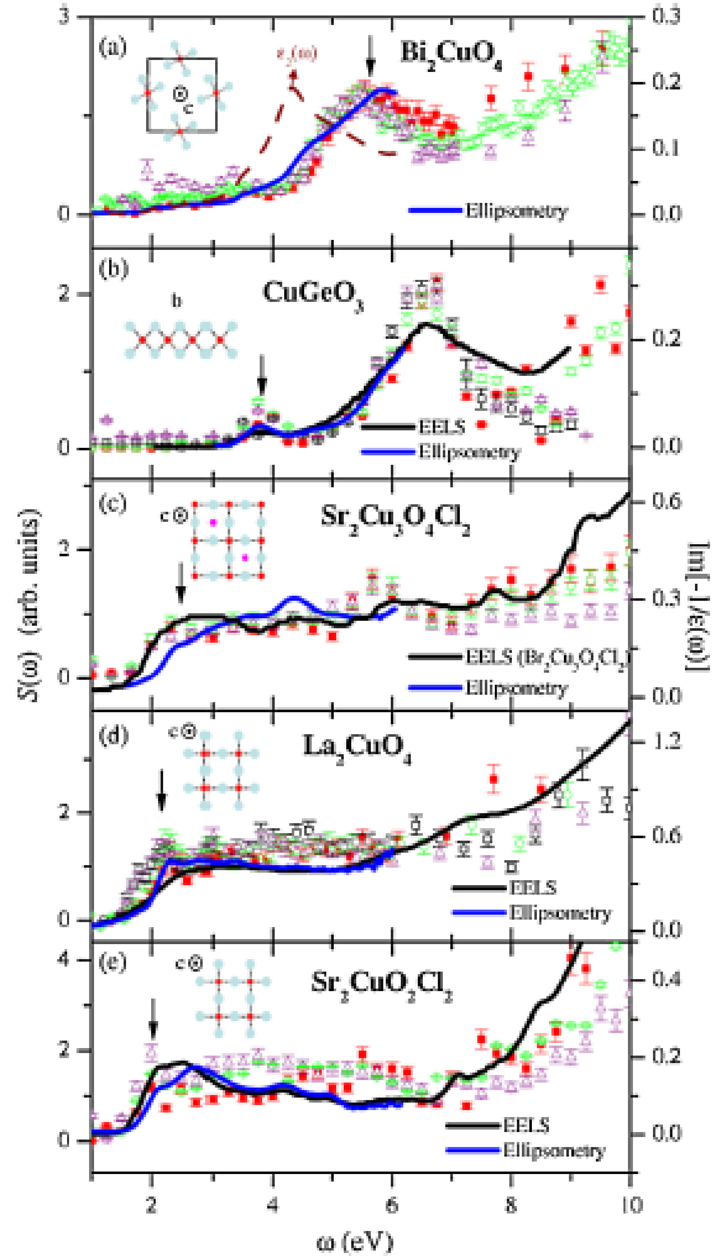
Total resolution at Cu K edge (FWHM)~ 83 meV
For other transition metal K-edge ≤ 100 meV

Improving resolution by Saphire (Al_2O_3), Quartz (SiO_2), and $LiNbO_3$ is under progress

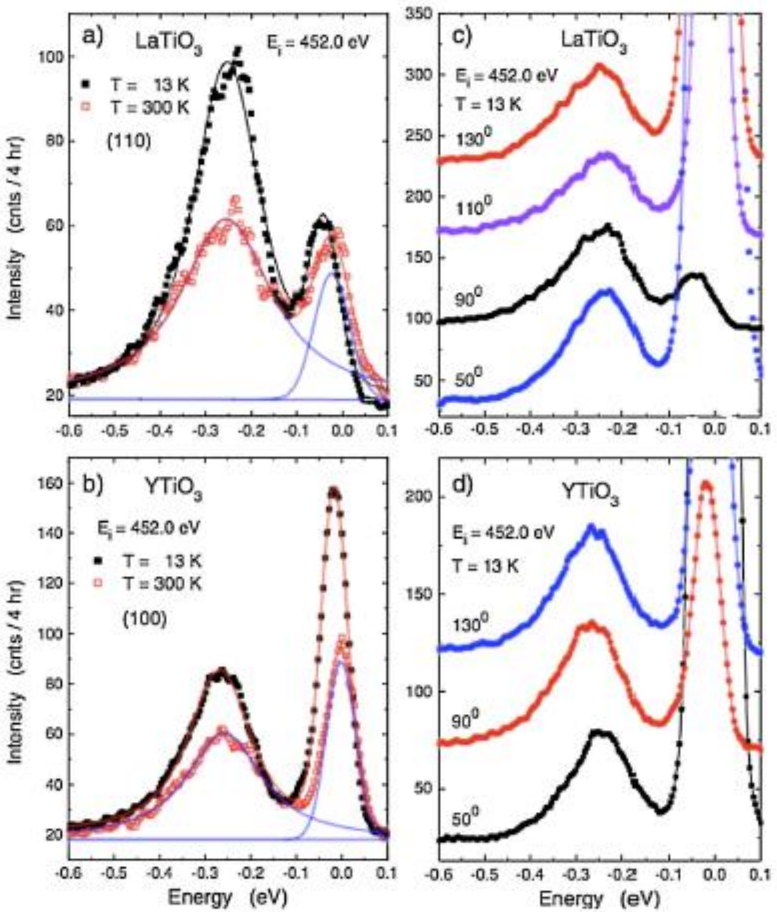
Typical Results

Comparison of S(ω) with the measured Im[-1/ε(ω)] for various cuprates,

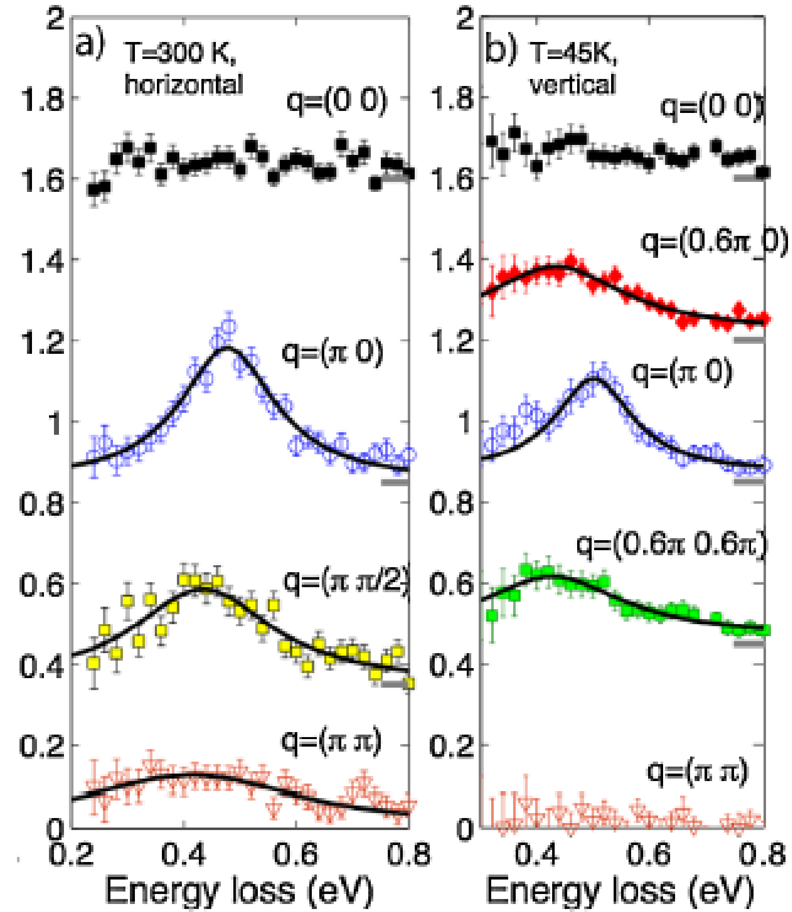
In comparison with EELS



Typical Results



Ti L3-edge RIXS spectra of Titanates



Cu K-edge RIXS spectra of La2CuO4
High Tc Cuprate superconductor

Inelastic X-ray Scattering

Rep. Prog. Phys. 63 (2000) 171–232. Printed in the UK

Phonon spectroscopy by inelastic x-ray scattering

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The present synchrotron sources with brilliant x-ray beams, due to high photon fluxes, small source sizes and high collimation, have revolutionized x-ray physics.

Enormous progress has been initiated in all established x-ray methods, with the aim of the development of new types of spectroscopy. This is particularly true for the spectroscopy of the dynamics in condensed matter. Meanwhile, there are two powerful x-ray methods with very high-energy resolution available for the study of low energetic excitations like phonons.

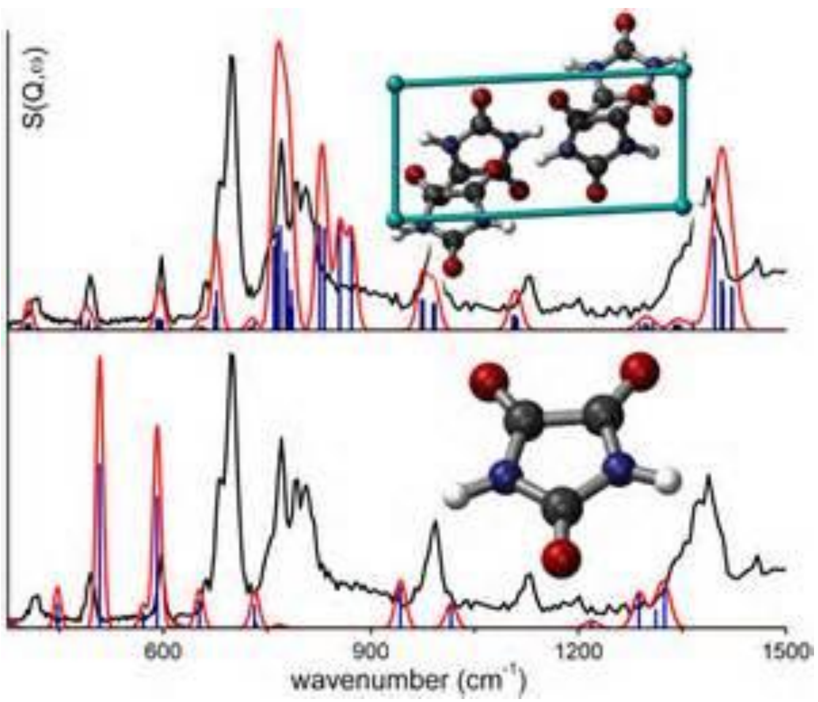
This review summarizes the developments of these methods focusing on these instrumental developments of the spectrometers using either crystal optics in close-to-backscattering geometry or nuclear resonant techniques.

Applications to measurements of phonon dispersion curves and of phonon density of states

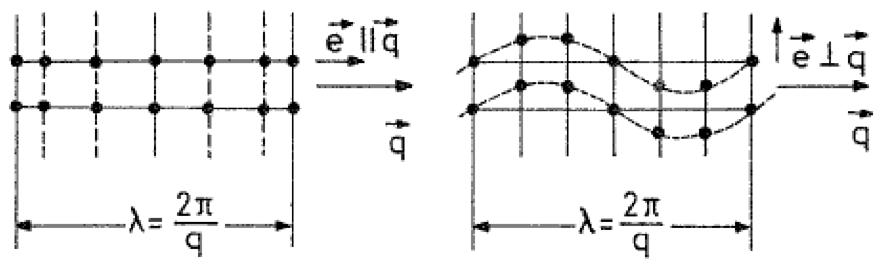
IXS for Phonon

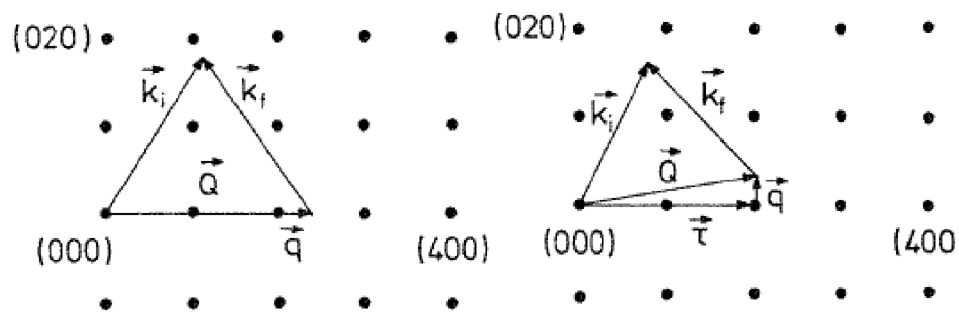
Measuring dynamical properties give very important physical and chemical properties of materials like specific heat capacity, heat transport, electron transport, sounds, etc

In molecules
specific vibration modes
for each bond can be measured by Raman



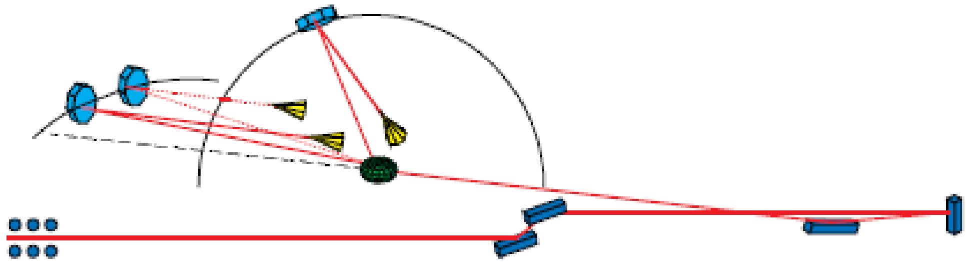
In Solids
The phonon mode has dispersion
with momentum
The dispersions can be measured by NIS or XIS



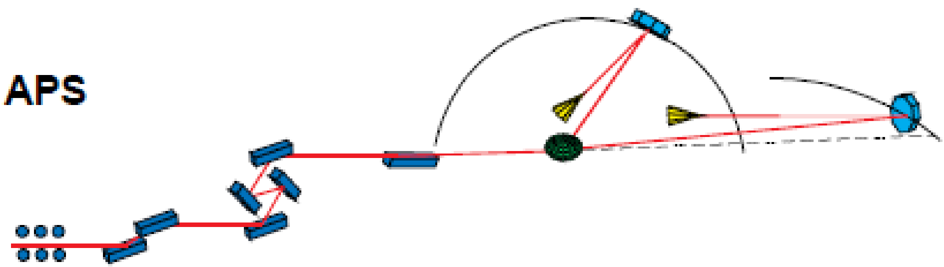


Beamline Layout

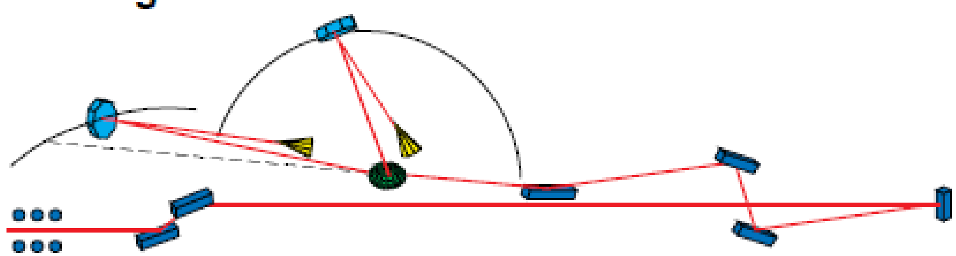
ESRF



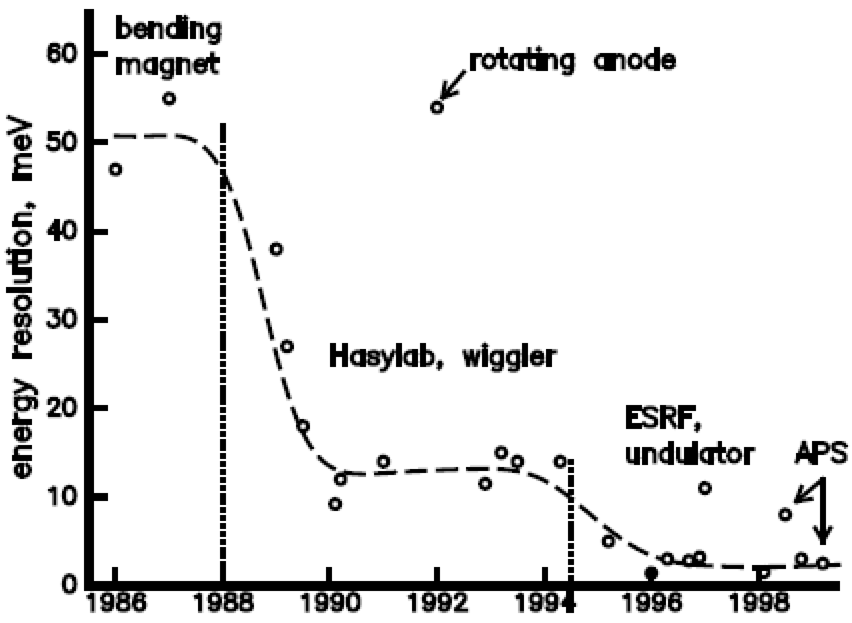
APS

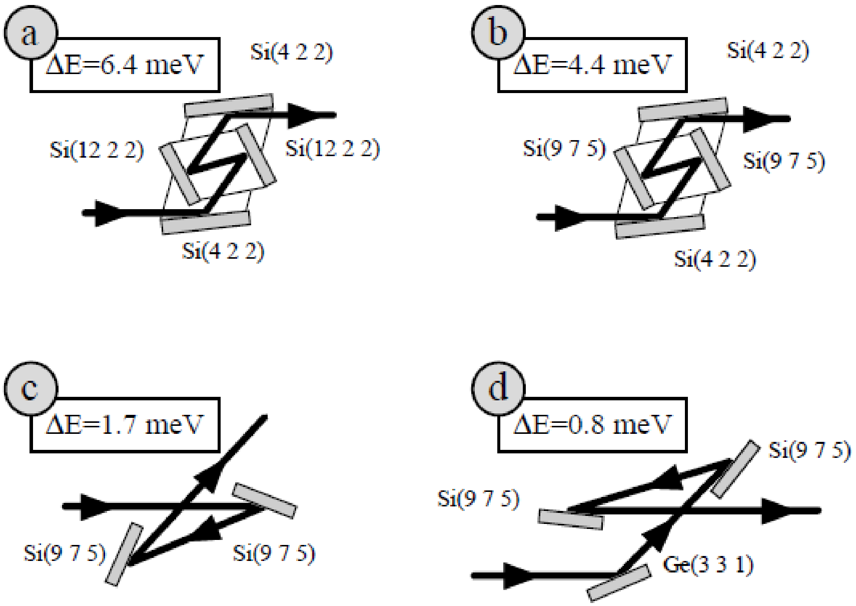


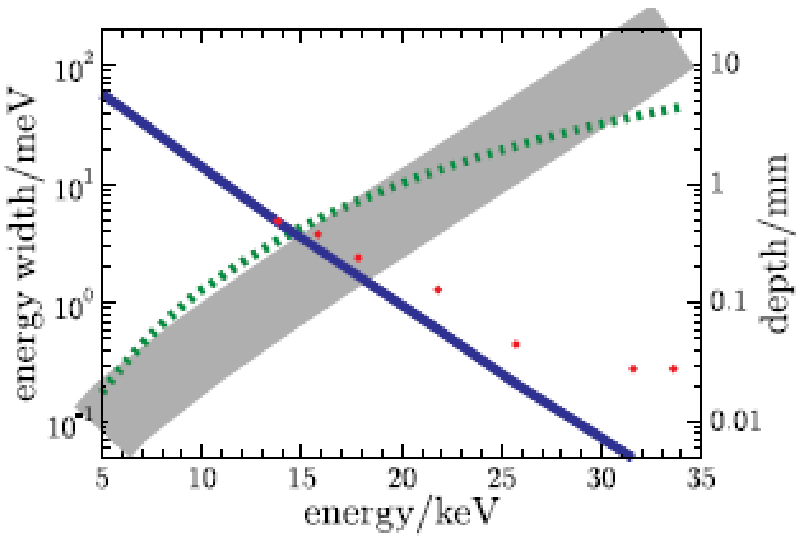
SPring8



High Energy Resolution







Experimental Condition

Table 2. Comparison of backscattering spectrometers at HASYLAB, at the ESRF, at APS and SPring-8.

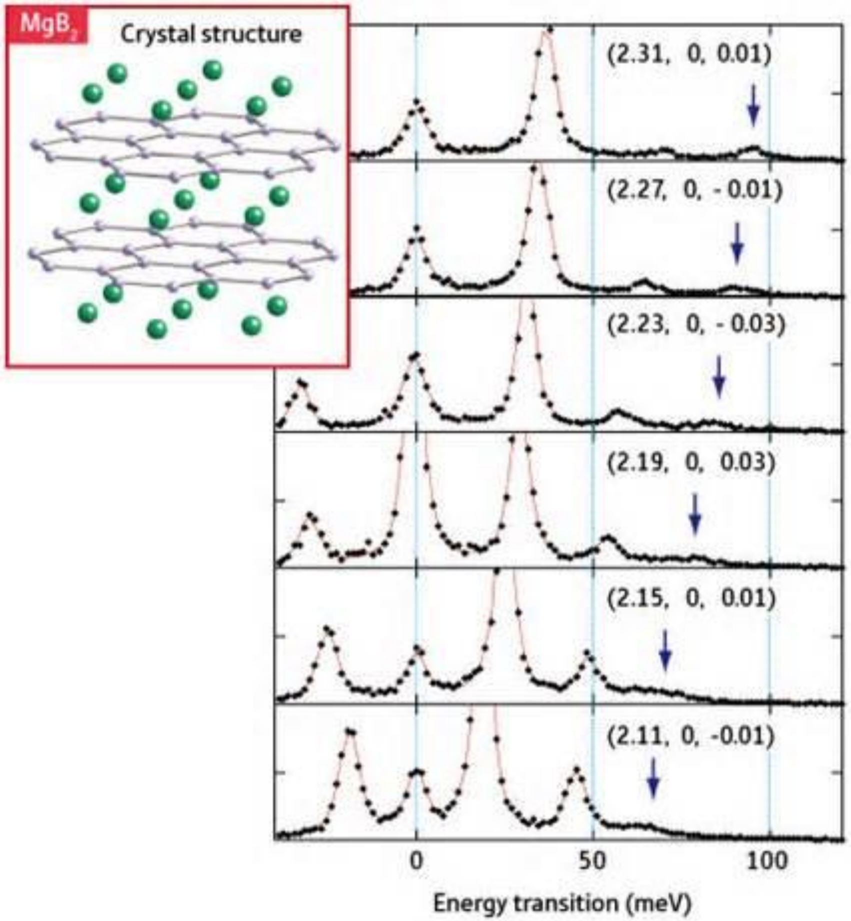
Location	INELAX HASYLAB Hamburg, Germany	ID16 ESRF Grenoble, France	3ID APS Chicago, USA	BL35XU SPring-8 Kansai, Japan
Operational	1987	1994	1997	2000
		Source characteristics		
Ring energy	4.5 GeV	6.0 GeV	7.0 GeV	8.0 GeV
Ring current	140 mA	200 mA	100 mA	100 mA
Insertion device	wiggler	2 undulators	undulator	undulator
Length	2.6 m	1.6 m each	2.5 m	4.5 m
Divergence:				
vertical	0.12 mrad	24 μrad	12 μrad	11 μrad
horizontal	2.6 mrad	40 μrad	42 μrad	35 μrad
		X-ray optics		
Pre-monochromator	Si (1 1 1) 2-crystal setting room temperature	Si (1 1 1) channel-cut at 90–120 K	diamond 2-crystal setting room temperature	Si (1 1 1) 2-crystal setting cryogenically cooled
Main monochromator	Si (h h h) backscattering spherically bent	Si (h h h) backscattering flat focusing mirror	Si (4 4 0) + (15 11 3) nested channel-cut focusing mirror	Si (h h h) backscattering flat focusing mirror
Focus spot	3×1 mm^2	500×300 μm^2	600×500 μm^2	100×150 μm^2
Analyser	Si (h h h) spherically bent	Si (h h h) spherically bent	Si (h h h) spherically bent	Si (h h h) spherically bent

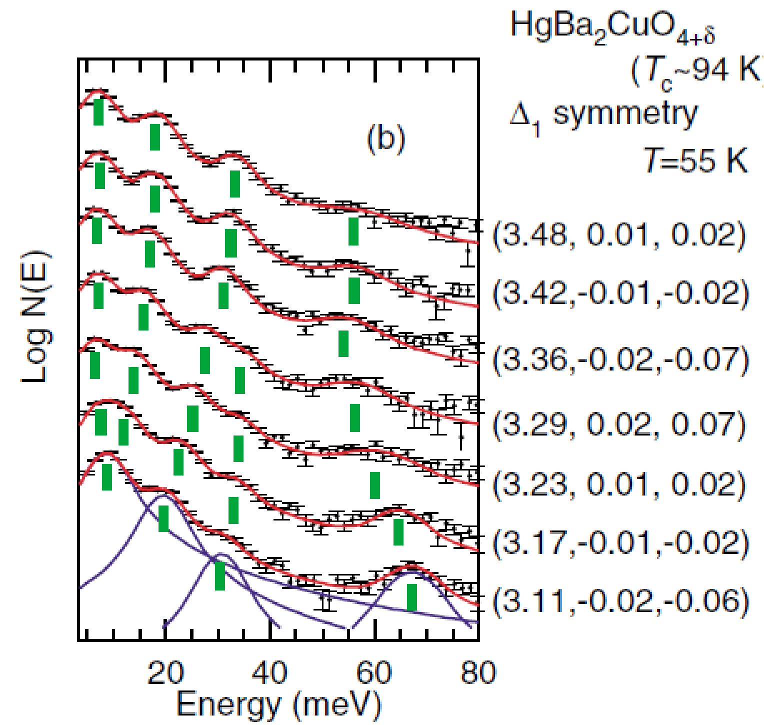
Examples

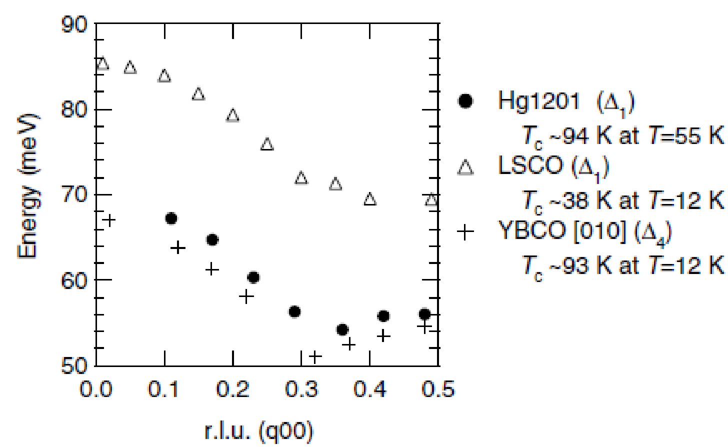
VOLUME 92, NUMBER 19 PHYSICAL REVIEW LETTERS week ending 14 MAY 2004

Softening of Cu-O Bond Stretching Phonons in Tetragonal $HgBa_2CuO_{4+\delta}$

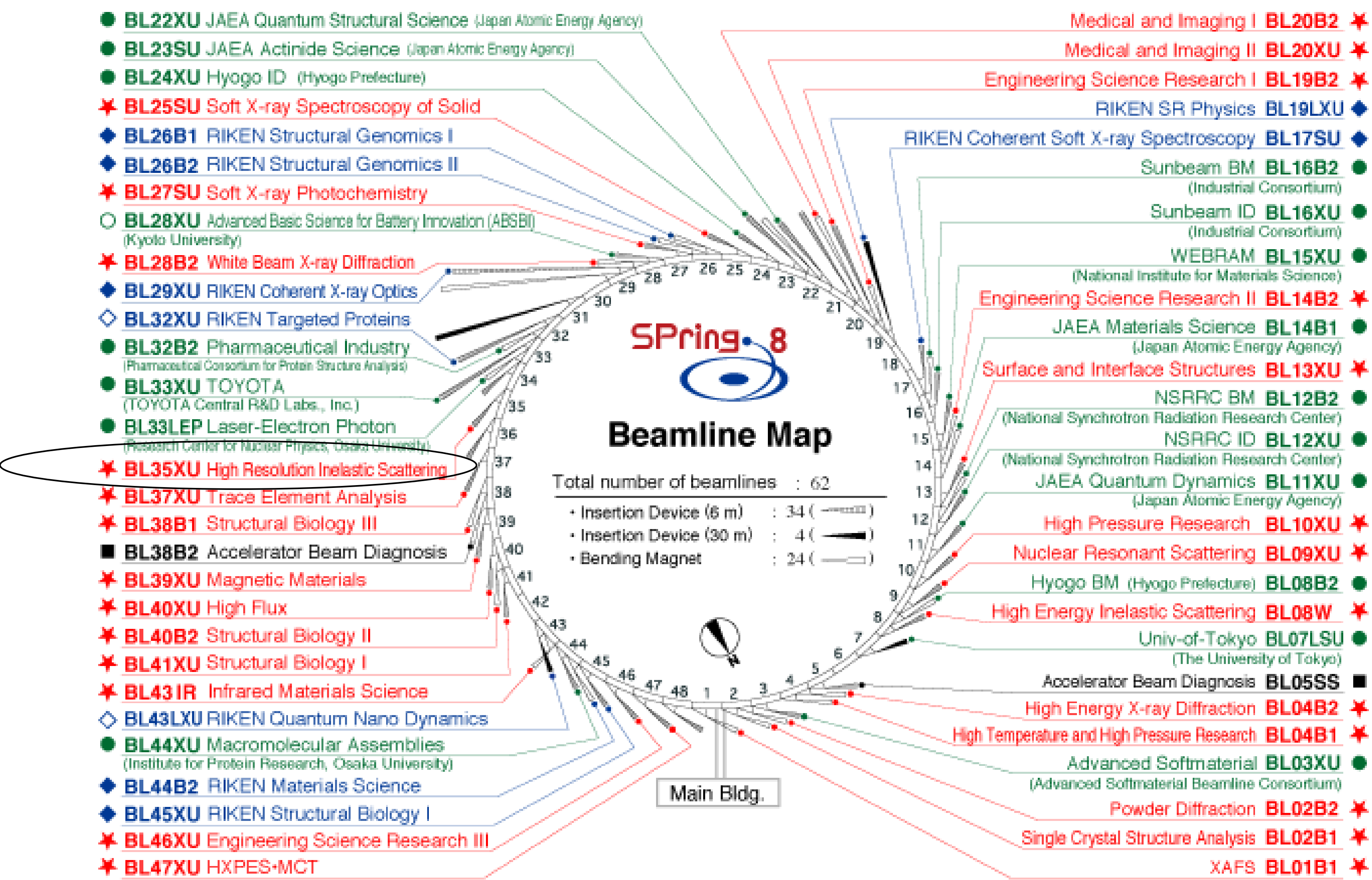
H. Uchiyama,[1] A. Q. R. Baron,[2] S. Tsutsui,[2] Y. Tanaka,[3] W.-Z. Hu,[1] A. Yamamoto,[1] S. Tajima,[1] and Y. Endoh[4]



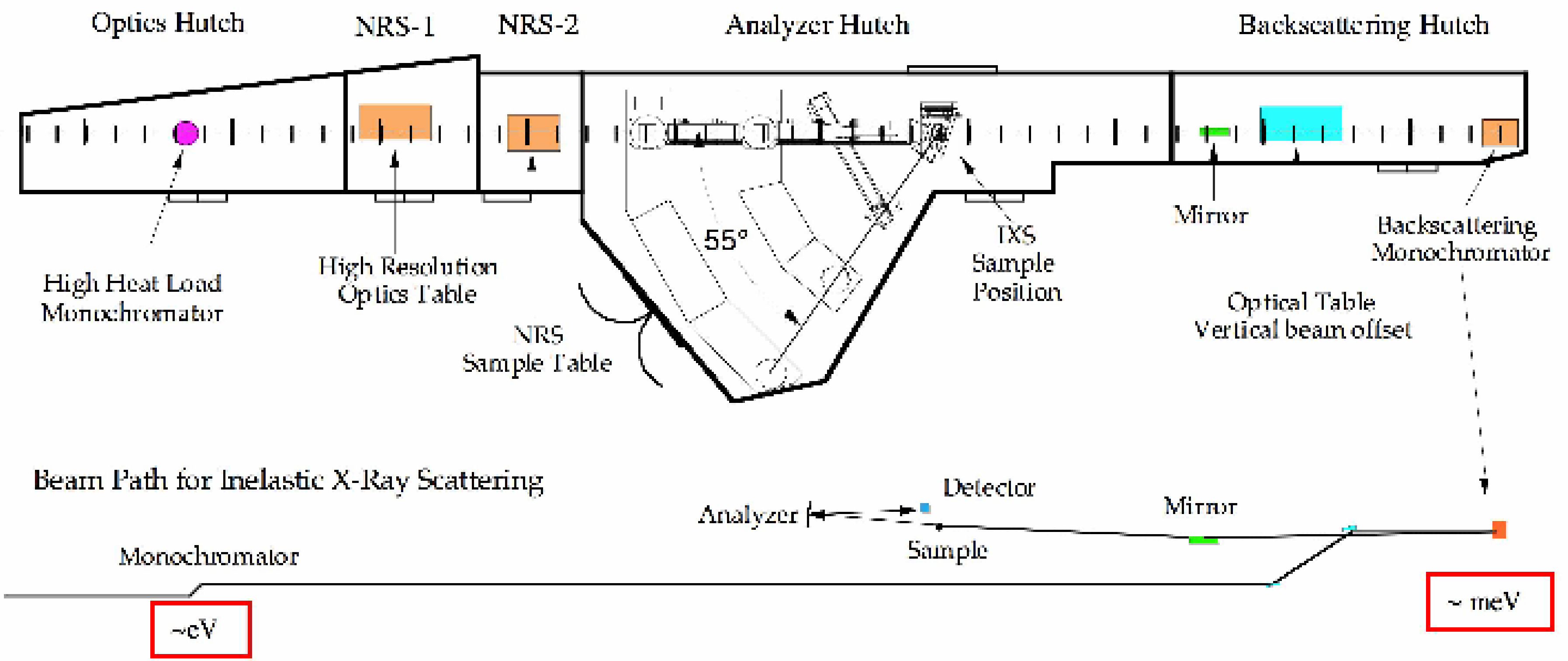




BL35- Inelastic X-ray Scattering

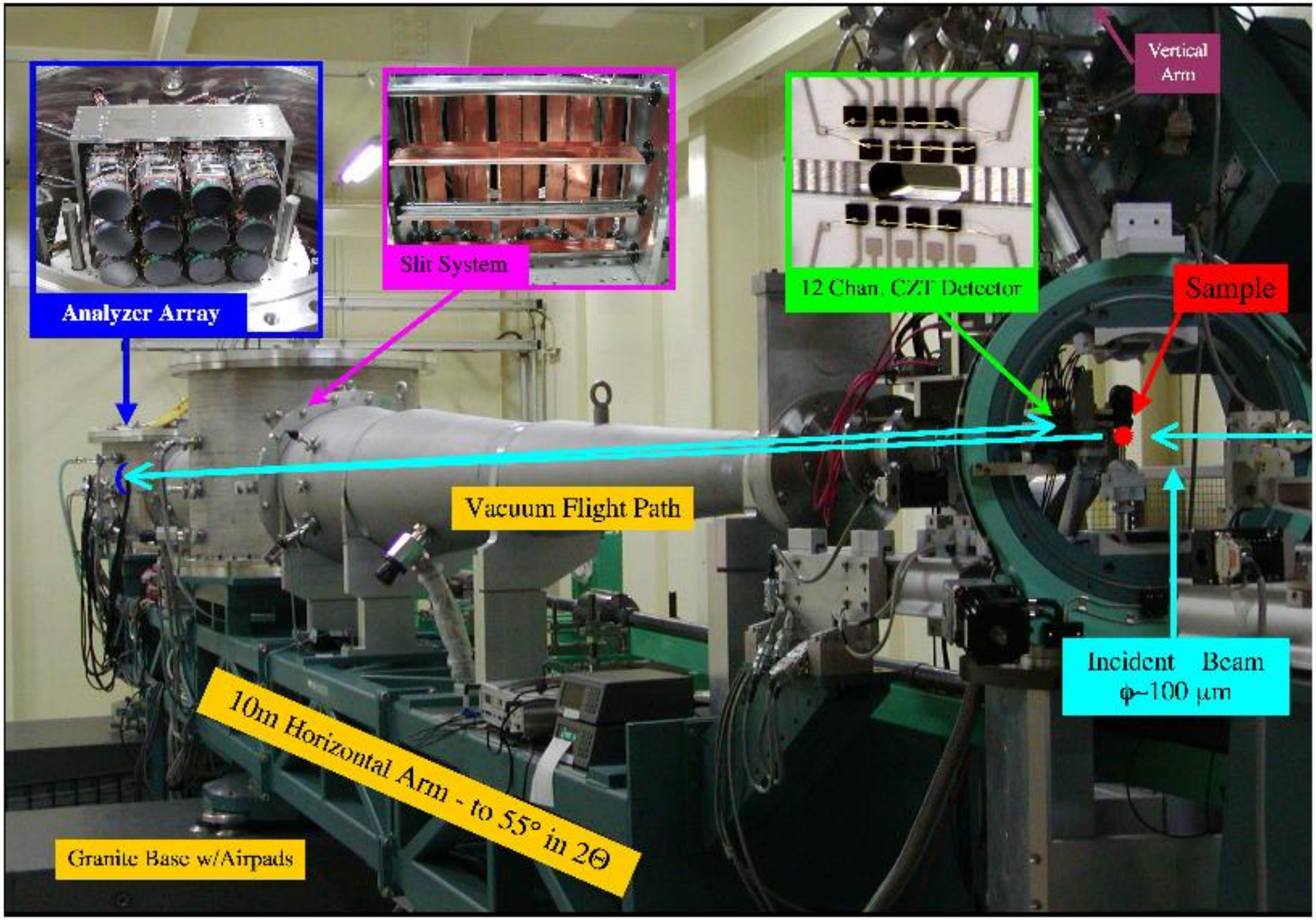


BL35XU Layout

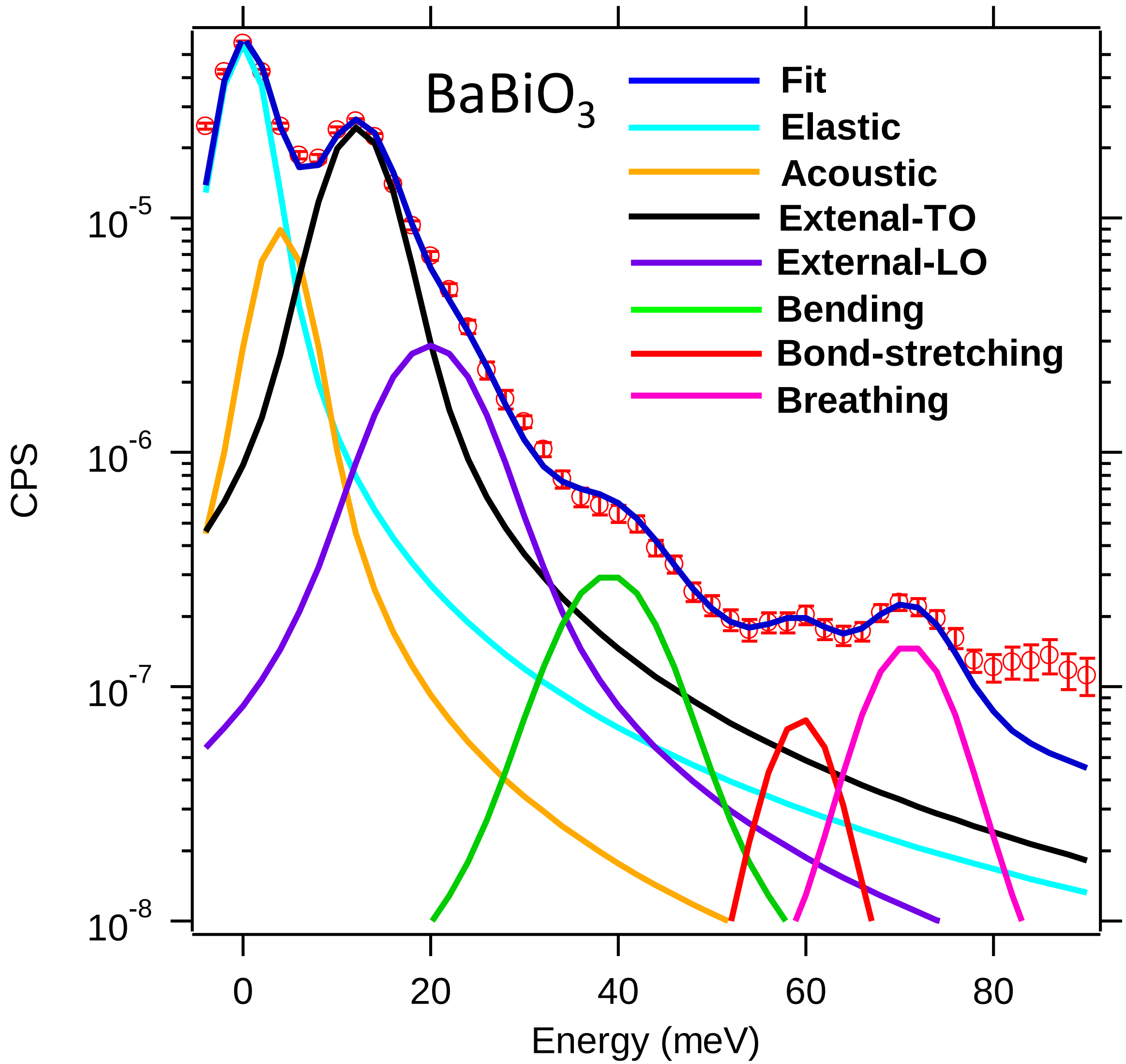


Energy (keV)	Si Order	Resolution (meV)	Flux at Sample (GHz)
15.816	**(8 8 8)**	**6**	**30**
17.794	**(9 9 9)**	**3**	**10**
21.747	**(11 11 11)**	**1.5**	**3.5**
25.702	(13 13 13)	1.0	0.8

Experimental Station

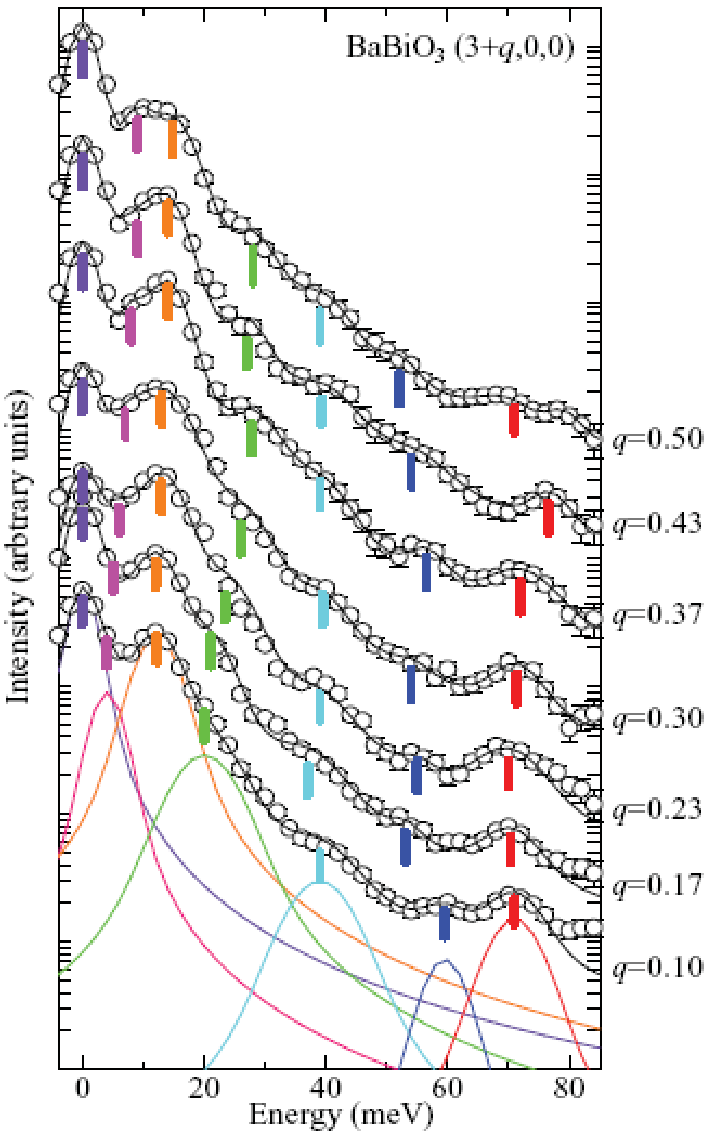


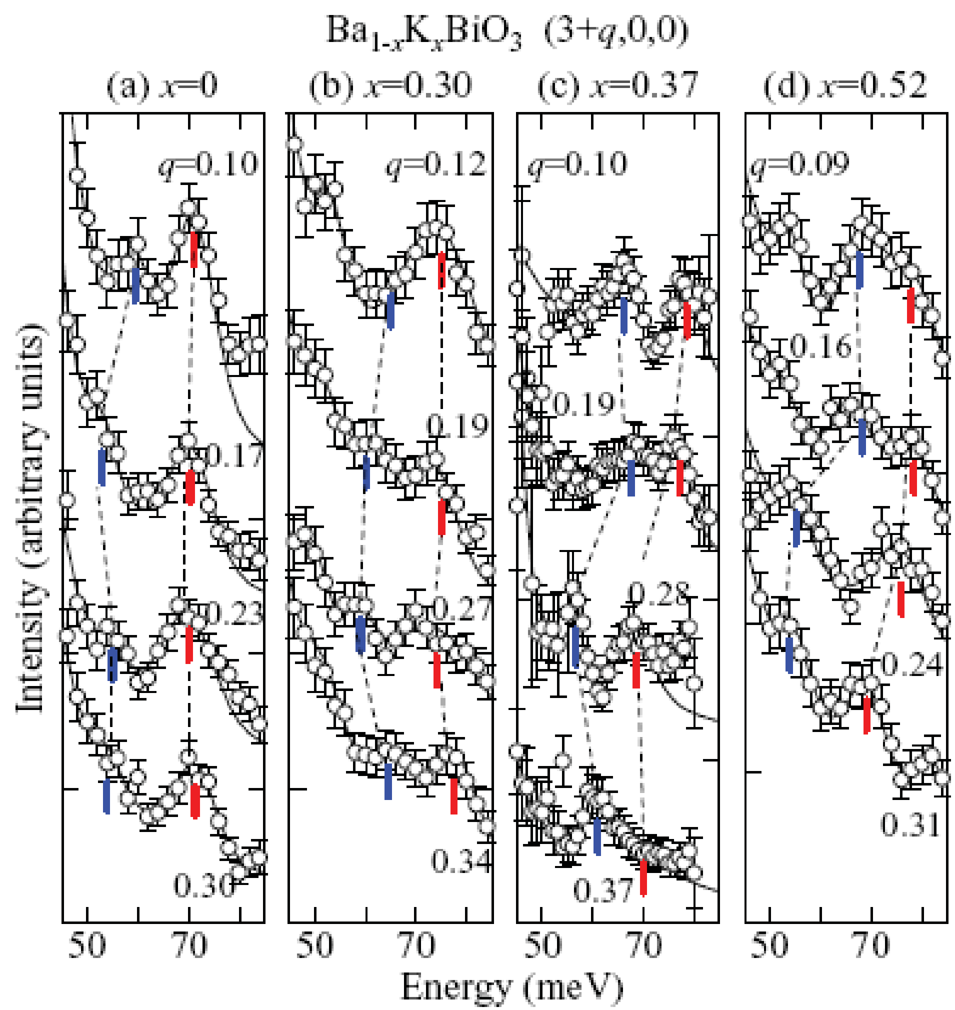
Phonon Spectrum

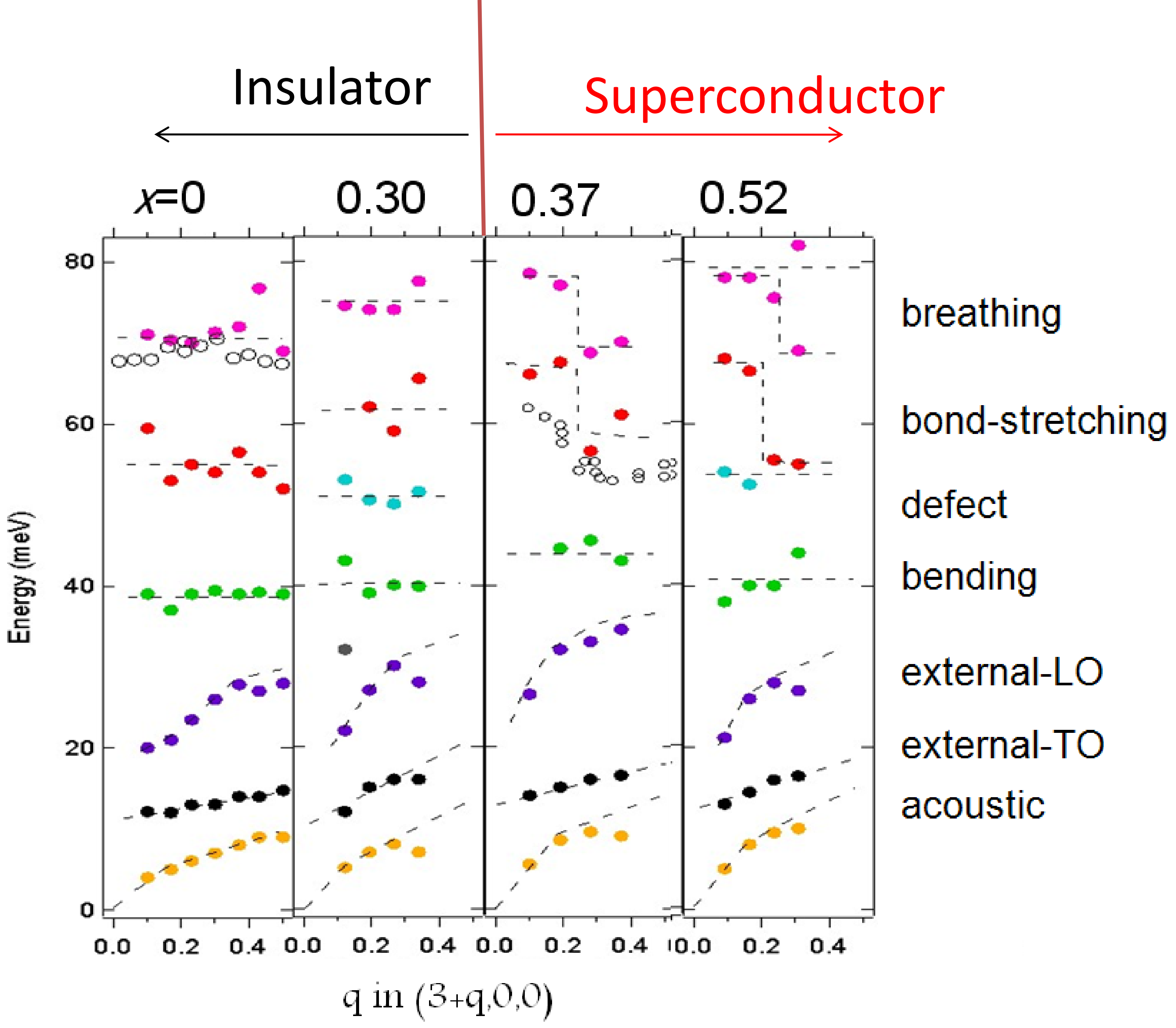


$$I(\omega) = I_0 \left\{ \frac{2\eta}{\pi} \frac{\Gamma}{\Gamma^2 + 4(\omega - \omega_0)^2} + \frac{2(1-\eta)}{\Gamma} \left(\frac{\ln 2}{\pi}\right)^{0.5} \exp\left(-4 \ln 2 \left(\frac{\omega - \omega_0}{\Gamma}\right)^2\right) \right\}$$

Momentum & Doping dependence






Insulator
Superconductor
x=0
0.30
0.37
0.52
Energy (meV)
80
60
40
20
0
0.0 0.2 0.4
q in (3+q,0,0)
breathing
bond-stretching
defect
bending
external-LO
external-TO
acoustic

Summarize

Many attempts has been done in the recent years to develop RIXS and IXS experimental techniques for measuring the low energy excitations

Using synchrotron radiation and technological improvement in the beamline and spectrometers now it is possible to reach very high resolution measurement of this excitations in wide range of momentum

Different beamline in modern synchrotrons are active for these measurements

For variety of complicated physical systems like manganites, high-Tc superconductors, strongly correlated systems, etc the techniques have been successful to give many good microscopic Information for understanding the physical phenomenon

Progress is continuing to obtain better resolution and experimental data and for unconventional new materials