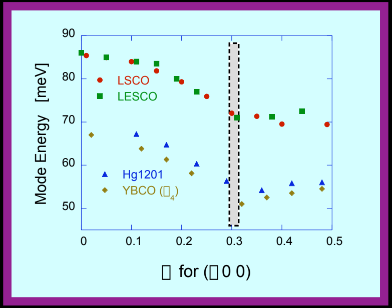
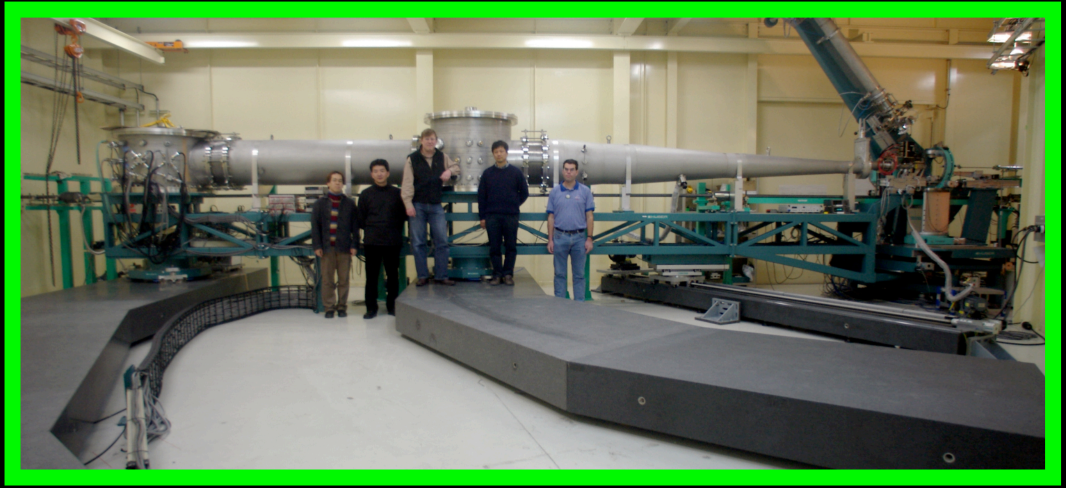
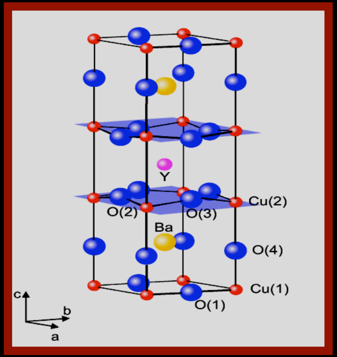
Perspectives In

meV-Resolved Inelastic X-Ray Scattering





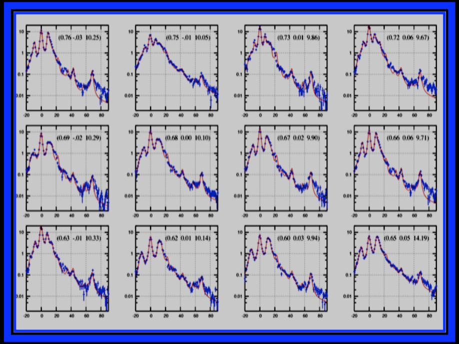


Alfred Q.R. Baron

SPring-8

Materials Dynamics Laboratory, RIKEN
Research & Utilization Division, JASRI

NSLS II Seminar, BNL
24 April 2006



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Contents

Present Spectrometer (BL35XU)
Performance & Results

Considerations for new beamlines.

Note: Emphasis on Instrumentation

Why do meV Inelastic X-Ray Scattering?

Many good points: No Kinematic Constraints (Liquids!)
Nearly No Background
Simple (& Good) Momentum Resolution
Large Scan Range (~ eV or more)

Main Point: Small Samples are Possible

~1 mm^3 comfortable
10^{-1}-10^{-2} mm^3 straightforward
10^{-3}-10^{-4} mm^3 with work
10^{-5}-10^{-6} mm^3 possible
< 10^{-6} mm^3 probable

Neutrons
10^2 to 10^4 mm^3

Systematic & Rapid Response Studies
are feasible in a way never before possible.

Areas of Endeavor

Disordered Materials

“Just” small samples
    “Hot New Samples” (MgB$_2$, Superconducting Diamond)
    Systematic Studies (High-Tcs, CDWs, etc)

Extreme Environments (& usually small): e.g. DAC

Special geometries: Surface sensitivity in extreme grazing incidence

Electronic Excitations (Gaps, Orbitons, etc)
    Not yet successful.

IXS Technical Hurdles

meV Resolution -> $\Delta E/E \sim 10^{-7}$

Small cross-section, low count-rates ->

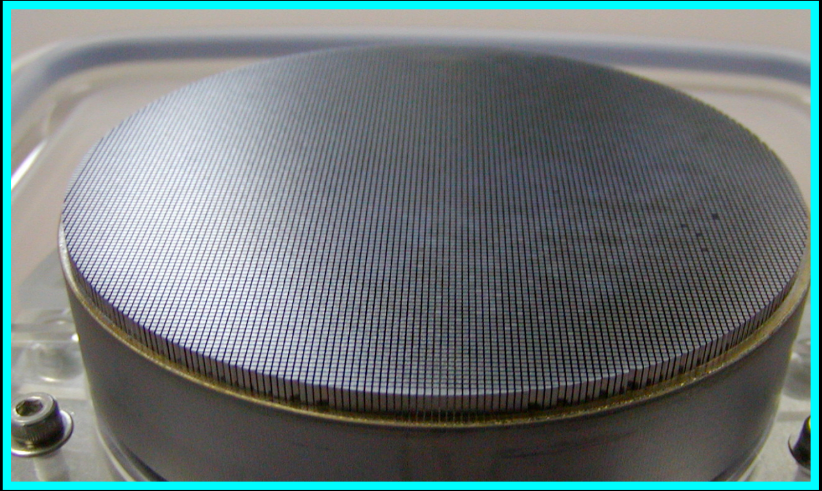
Strong x-ray Source:	**SPring-8**
Highly Efficient Beam Handling:	**Extreme Backscattering**
Highly Parallel Data Collection:	**12-Analyzer Array**

Use thermal expansion for energy scans of monochromator.

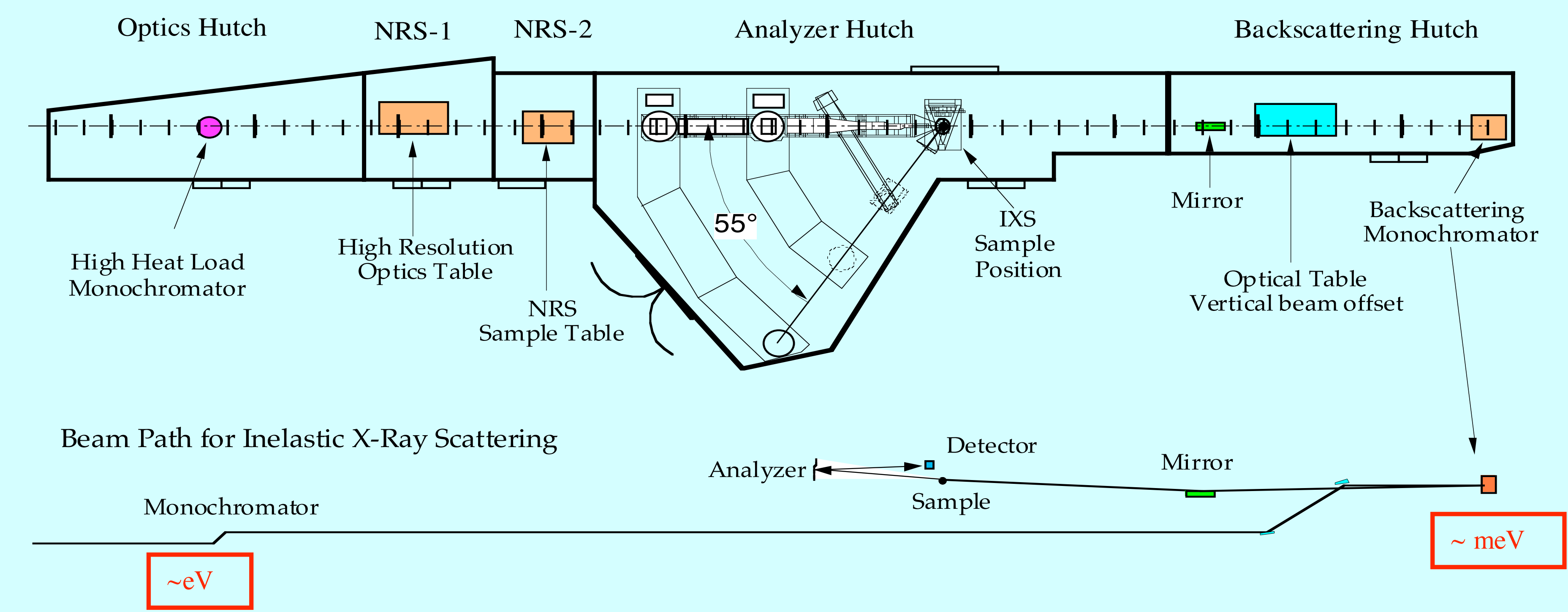
$\Delta d/d = \Delta E/E \sim 2.6$ ppm/K

18 mK / meV at 22 keV

Analyzer Crystals: $\sim 10^4$ independent crystallites on a common substrate to avoid strain.

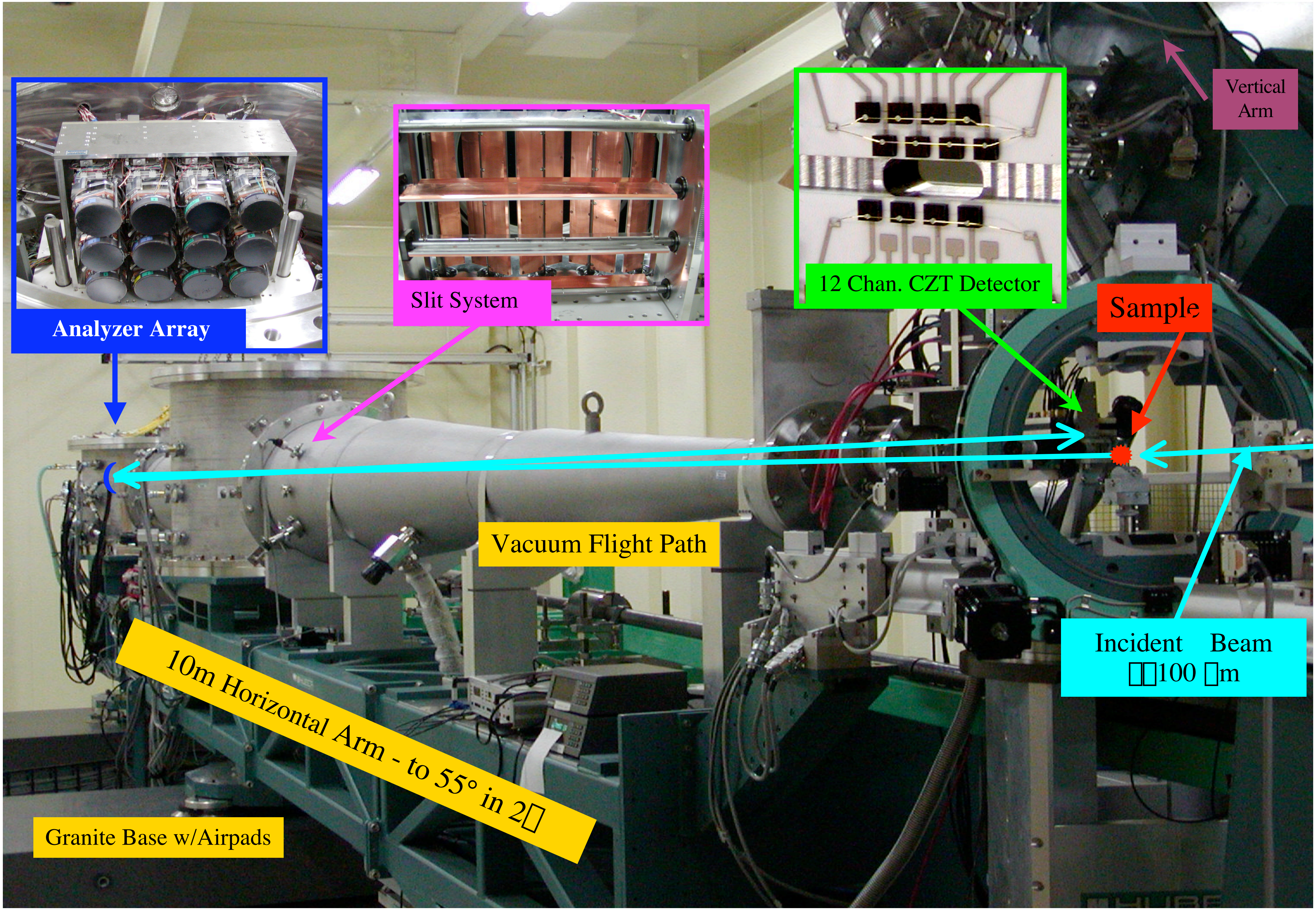


SPring-8 BL35XU



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**	**4.5**
25.702	(13 13 13)	1.0	0.8

Best Beam Spot on Sample: 50 μm V x 70 μm H (FWHM)

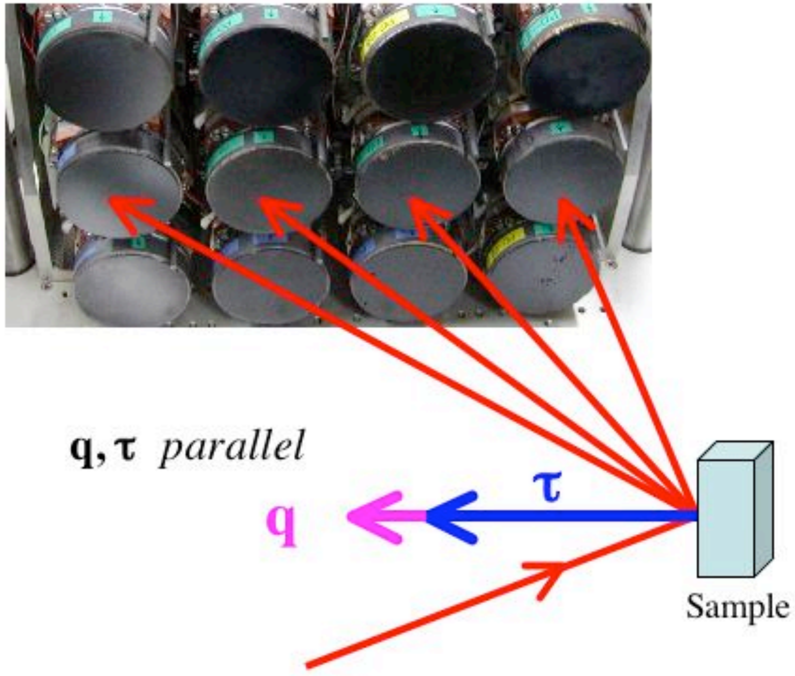

Analyzer Array
Slit System
12 Chan. CZT Detector
Vertical Arm
Sample
Vacuum Flight Path
Incident Beam
□□100 □m
10m Horizontal Arm - to 55° in 2□
Granite Base w/Airpads

Phonon Geometries & Analyzer Array

Q□ = q + □

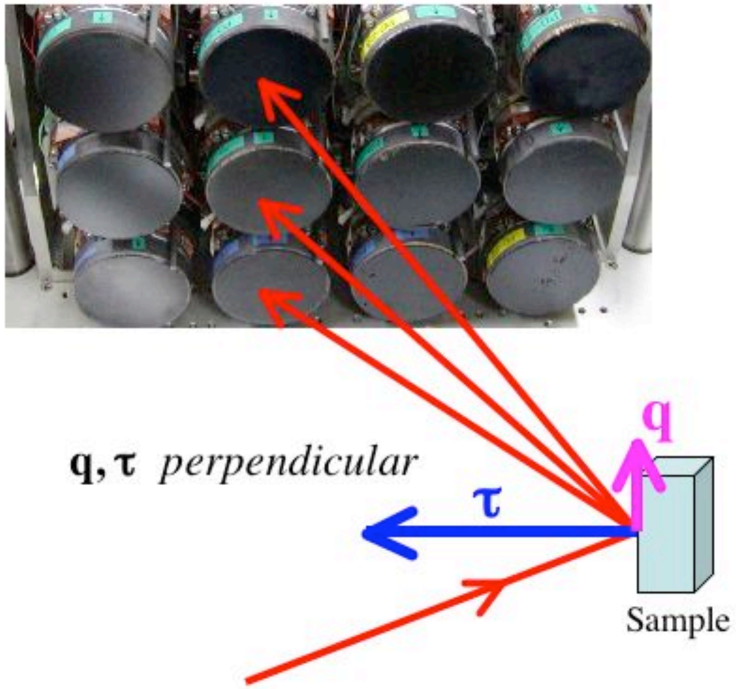
q = reduced momentum transfer in first zone
□ = nearest Bragg point

Longitudinal Geometry



Dispersion measured using
a *horizontal* line of analyzers

Transverse Geometry



Dispersion measured using
a *vertical* line of analyzers

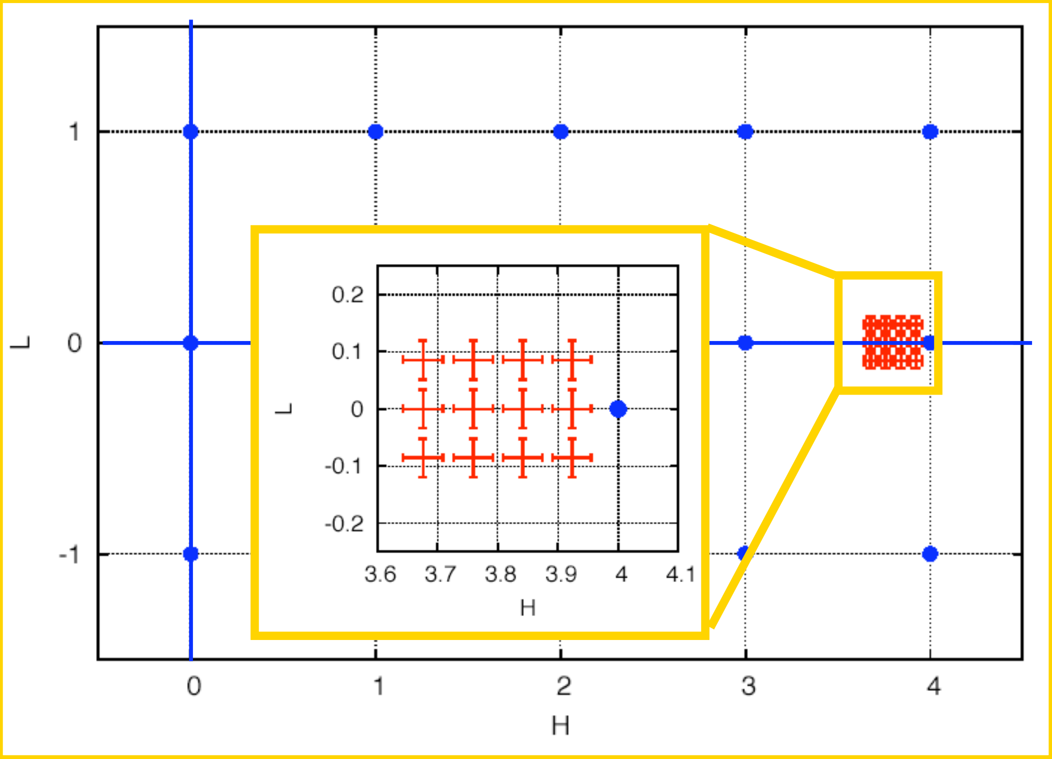
12-Analyzer Array: Example

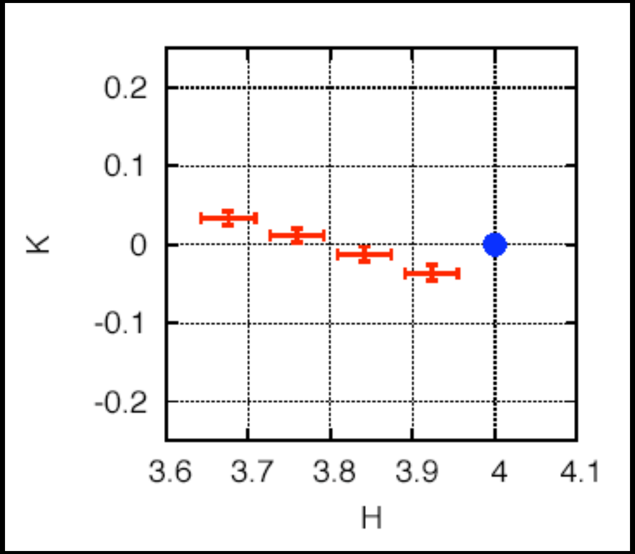
Cubic Sample, a = 4A, 21.7 keV

Scattering Plane: (100)x(010)

Vertical: (001)

Arm near to (400) Bragg Point





Note:

Tilt in (010) due to fixed theta

Spacing/Resolution ~ 0.1 rlu

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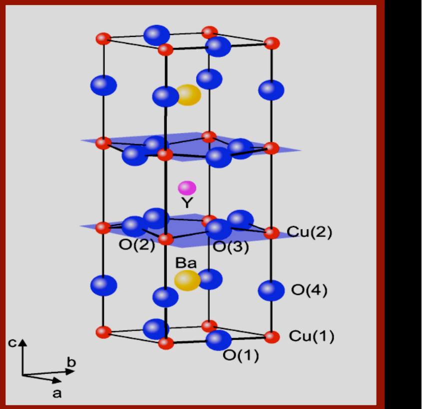
Copper-Oxide Superconductors

Complicated:

- Large Unit Cell (8 atoms minimum, >15 atoms typical)
- Modulated Structures
- Doped & Disordered
- Complicated Electronic Spectroscopy
- Short-range Spin and Electronic Structure



Role of Phonons in High-T_c superconductivity has been re-examined,
based on ARPES data (Lanzara et al, Schachinger, *et al*, Cuk, et al, Devereaux, et al)
& INS (Pintschovious et al, McQueeny et al, Pintschovious&Braden, Braden et al, Mook&Dogan, Reznik et al, etc)

Partial List of Interested Researchers:

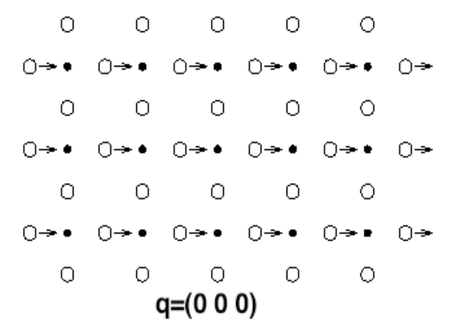
Braden, Cardona, Devereaux, Egami, Gunnarsson, Horsch, Ishihara, Keimer, Khaliulin, Lanzara, McQueeny, Mook, Nagaosa, Pinstchovius, Pyka, Reichardt, Reznik, Rosch, Shen, Sugai, Tachiki, Tranquada …

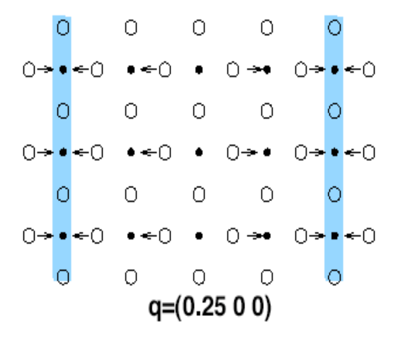
Interesting Phonons in High T_c Materials

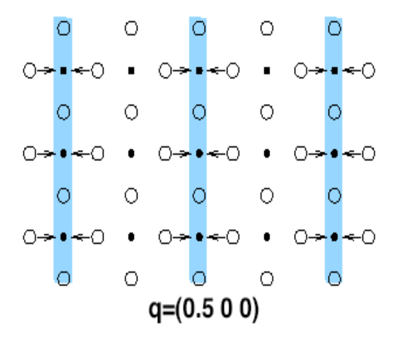
High-Energy "Bond-Stretching" mode

Longitudinal Mode, In-Plane Oxygen Motions

E ~ 60-80 meV, depending on material, Q


q=(0 0 0)


q=(0.25 0 0)


q=(0.5 0 0)

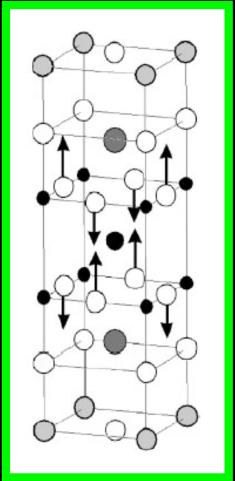
"Half-Breathing"
(0.5,0)= (□,0)

"Breathing"
(0.5,0.5)= (□, □)

Medium-Energy "Buckling" mode

Oxygen motions out of the plane, transverse

E ~ 30 - 45 meV, depending on material



Electron-Phonon Coupling (epc)

(Conventional)

Composite:
Mahan,
Allen,
Ginzburg,
etc

$$H = H_{el} + H_{lat} + H_{ep}$$

Lowest Order Term…

$$H_{ep} = \underset{\substack{n \\ atoms}}{\square} \left[V\left(\mathbf{r} \square \mathbf{R}_n^0 \square \mathbf{u}_n\right) \square V\left(\mathbf{r} \square \mathbf{R}_n^0\right) \right] = \square \underset{\substack{n \\ atoms}}{\square} \mathbf{u}_n \circ \square V\left(\mathbf{r} \square \mathbf{R}_n^0\right)$$

Electron Phonon

$$H_{ep} = \underset{\mathbf{qk}\square}{\square} M_{\mathbf{k},\mathbf{k+q},\square} \; a^{+}_{\mathbf{k+q}} a_{\mathbf{k}} \left(b_{\mathbf{q}\square} + b^{+}_{\square\mathbf{q}\square} \right)$$

Quantize,
Additional Approximations
(Fröhlich Hamitonian)

$$M_{\mathbf{k},\mathbf{k+q},\square} = \left(\frac{\hbar}{2M\square_{\mathbf{q},\square}} \right)^{1/2} \left\langle \mathbf{k} + \mathbf{q} \middle| \tilde{\square}_{\mathbf{q},\square} \circ \square V \middle| \mathbf{k} \right\rangle$$

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Phonon Self-Energy: “Π”

General Formulation for Superconductors

(Papers by Allen & collaborators, 1970s)

Π = Phonon self-energy
Ω_q = Phonon freq. w/o ep coupling
ω_q = Phonon freq. w/coupling.
γ_q = Phonon line-width (hwhm)
N(0) = Electron DOS @ Fermi Surface

$$\Pi = -2\sum_{\mathbf{k}} \frac{f_{\mathbf{k}} - f_{\mathbf{k+q}}}{\varepsilon_{\mathbf{k+q}} - \varepsilon_{\mathbf{k}} - \hbar\omega - i\eta} \left| M_{\mathbf{k},\mathbf{k+q},\nu} \right|^2$$

$$\gamma_q = \frac{\Omega_q}{\omega_q} \mathrm{Im}\left\{\Pi\left(\mathbf{q},\omega_{\mathbf{q}}\right)\right\}$$
$$= 2\pi\; N(0)\; \omega_q^{\,2}\; \lambda_q$$

Linewidth

$$\omega_q^{\,2} = \Omega_q^{\,2} \square 2\Omega_q \,\mathrm{Re}\left\{\square\square\left(\mathbf{q},\omega_{\mathbf{q}}\right)\right\}$$

$$\Rightarrow \quad \omega_q^{\,2} = \Omega_q^{\,2}\left(1 \square \square_q \square_q\right)$$

Frequency

Classical Indications of Electron-Phonon Coupling (epc)

1. Reduced Phonon Lifetime (Broadening)
2. Electronic Screening (Softening or Kohn Anomalies)
3. Influenced by Occupation Factors (T-Dependence)

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Electron-Phonon Coupling & Superconductivity

Conventional (BCS/Eliashberg) Superconductors

The Electron-Phonon Coupling Constant, λ,
is directly proportional to the phonon linewidth

$$\lambda_q = \frac{\gamma_q/\omega_q}{2\pi N(0)\omega_q}$$

$$\lambda = \sum_q \lambda_q$$

Approximate relationship due to McMillan:

$$T_c = \frac{\overline{\omega}_{\log}}{1.3}\exp\left[\frac{-1.04(1+\lambda)}{\lambda-\mu^*(1+0.62\lambda)}\right]$$

μ* = effective coulomb interaction
typically ~ 0.1

λ<<1 -> "Weak coupling"
λ >~1 -> "Strong coupling"

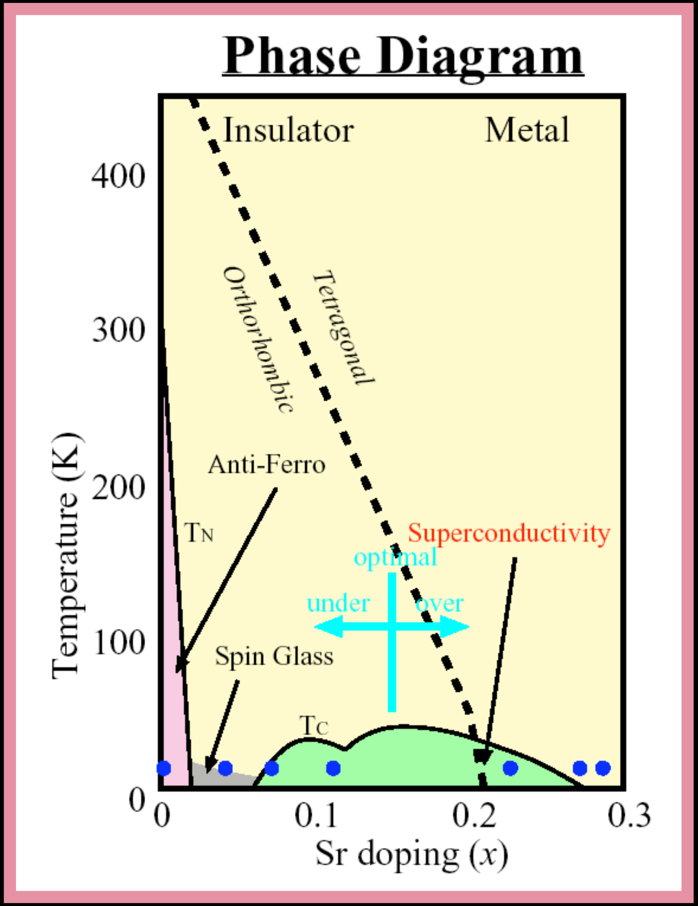
"Elaishberg Spectra Function" $\alpha^2F(\omega) = \frac{1}{2}\sum_q \lambda_q\,\omega_q\,\delta(\omega-\omega_q)$

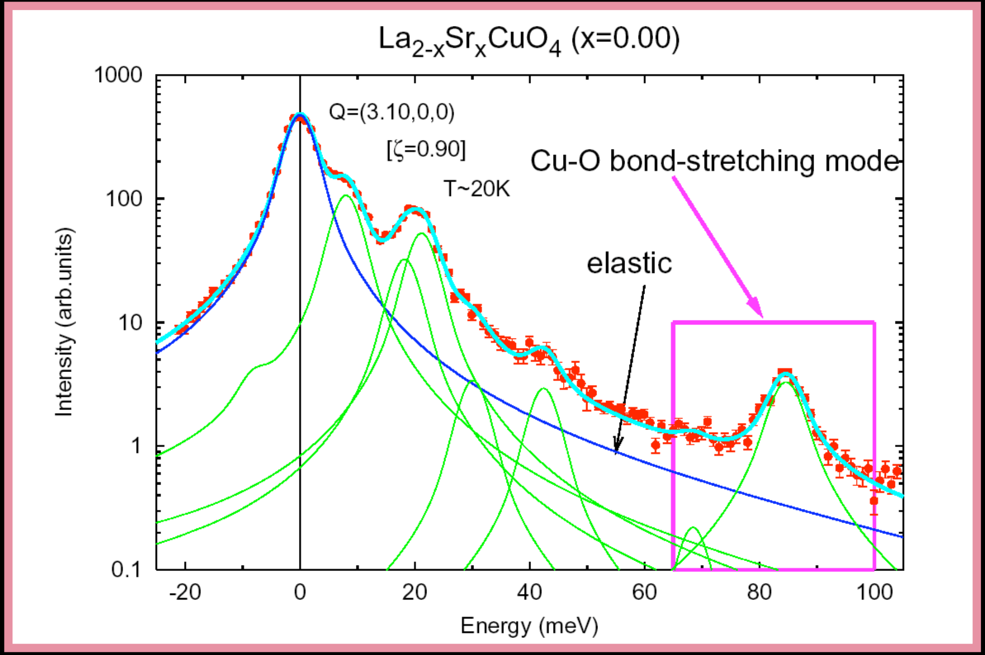
How to see electron-phonon coupling?

1. Broadening:	Usually small, ~ 0.1 meV Other sources (anharmonicity) exist. But: can be clear if big enough!
2. Softening:	Relationship to epc somewhat complicated (KK) But clearer experimentally in some cases (abrupt change = Kohn Anomaly)
3. Direct T-dep:	If the phonon spectrum changes going through T_c, this is a rather clear indication of epc.

Note: 1&2 require theory (or doping dependence)
3 is, perhaps, clearer, but rarer

IXS Study of $La_{2-x}Sr_xCuO_4$





Fukuda, Ikeuchi, *et al*,

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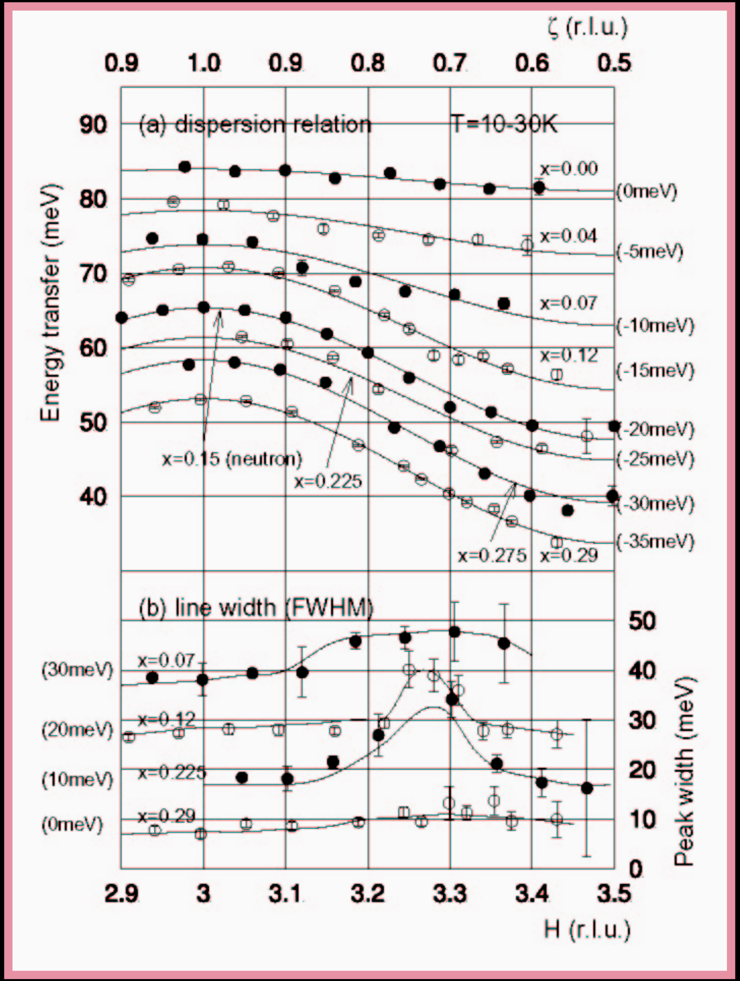
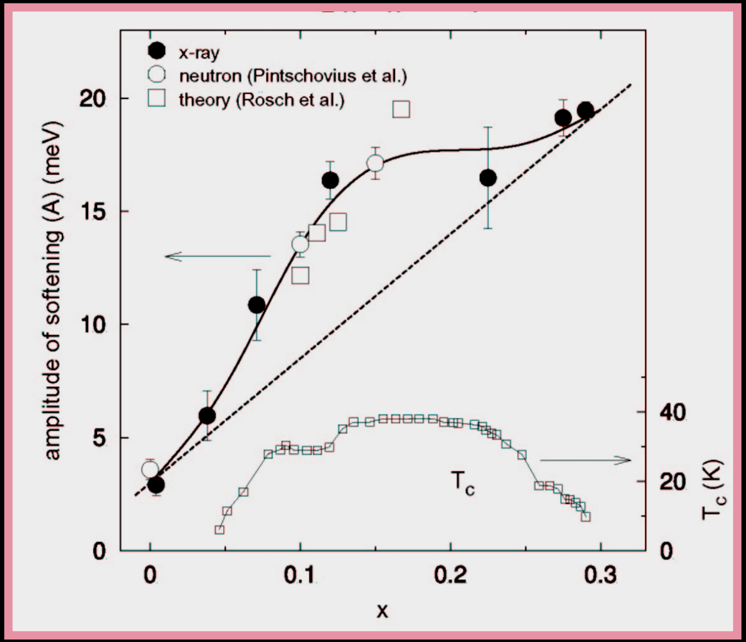
Doping Dependence of Softening in $La_{2-x}Sr_xCuO_4$

x = 0 to 0.29



Fukuda, Mizuki, *et al*, prb 71 (2005) 060501(R)



Deviation from linearity in x seems to correlate with superconductivity

Broadening at □~ 0.3 in superconducting region

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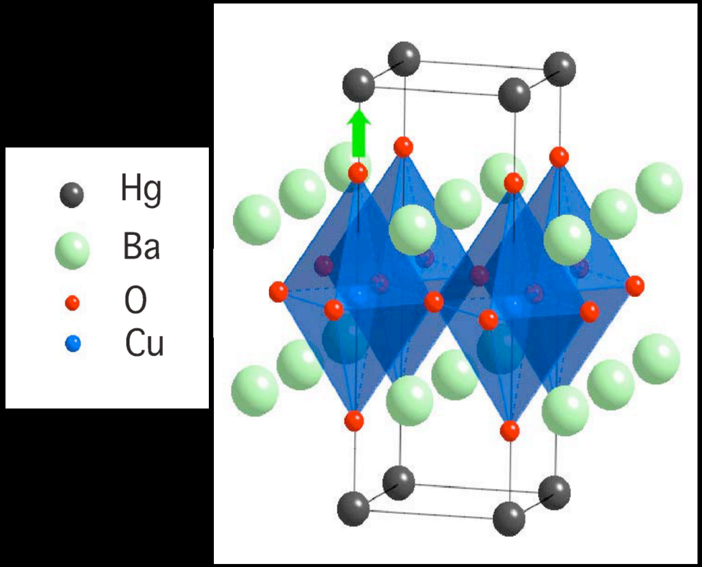
$HgBa_2CuO_{4+\delta}$



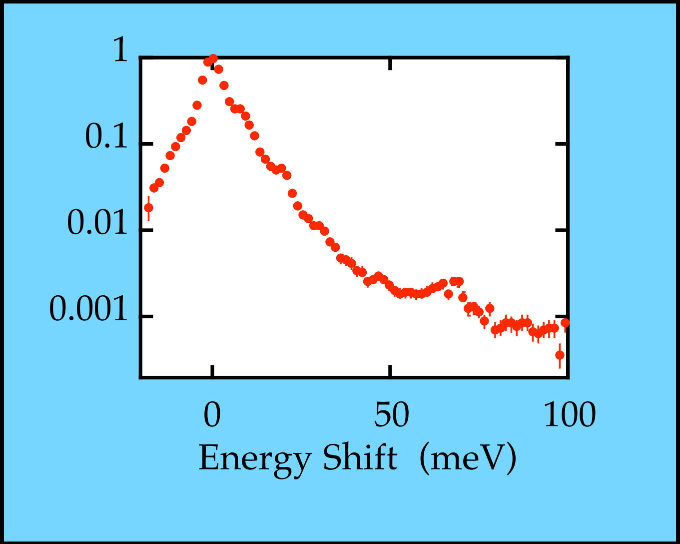
Simple Structure
Tetragonal, 8 atoms/cell

High T_c ~ 98K

However: Crystals are small
This Work: 0.3x0.3x0.2 mm^3

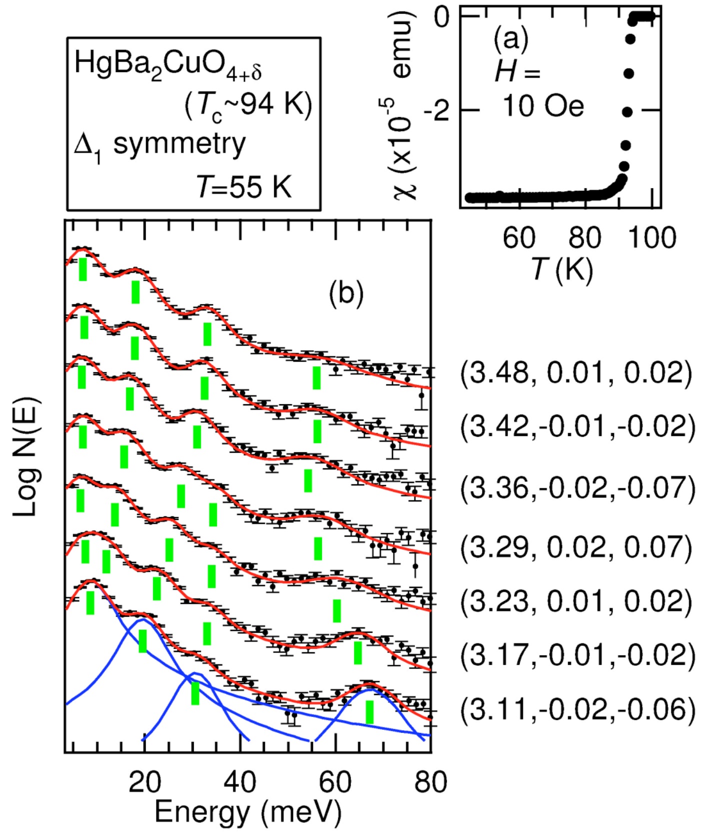
And absorbing (Hg and Ba)
l_{abs} ~18 um at 15.8 keV

But possible…

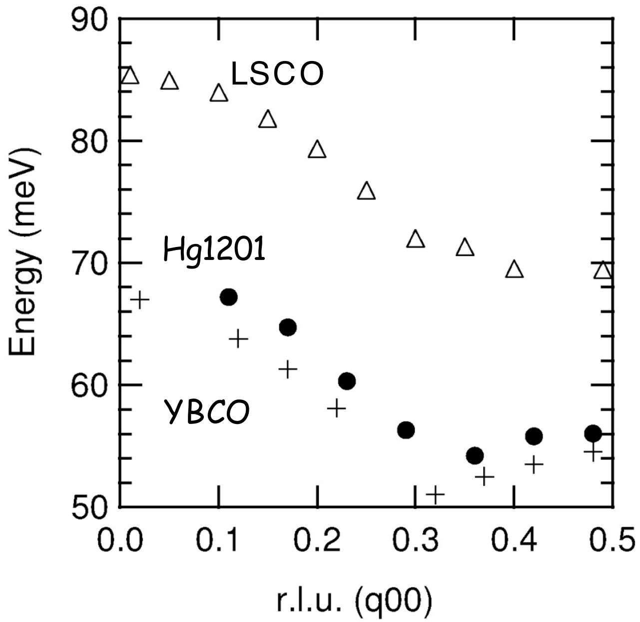


Elastic background…surface quality?

Softening in Hg1201



Elastic background subtracted.

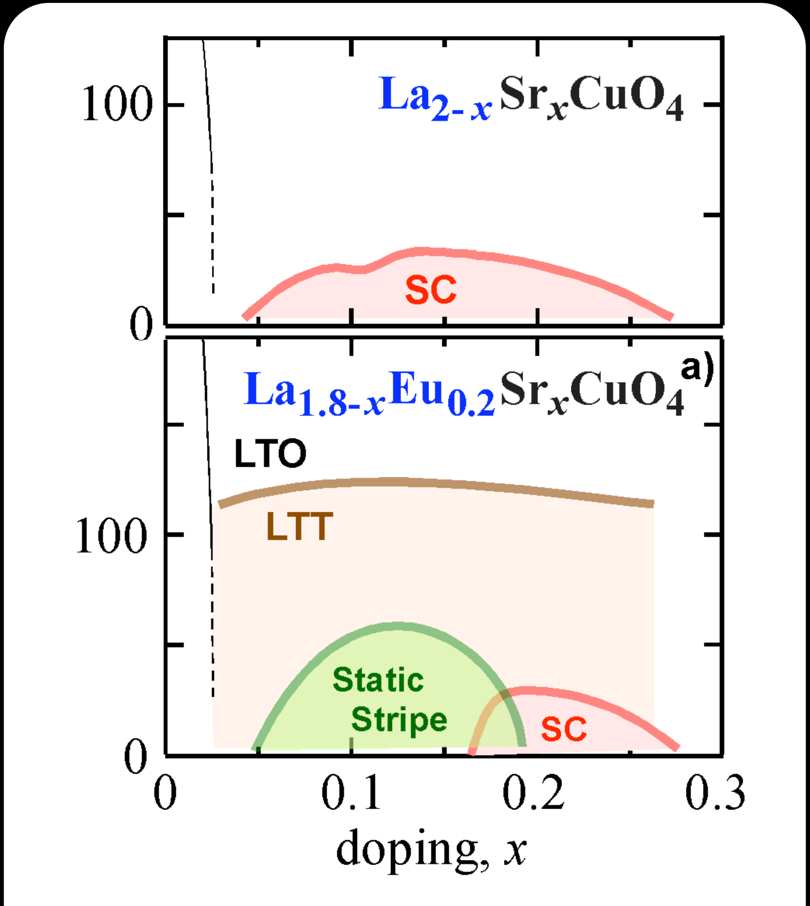


Uchiyama, Baron, et al, PRL 92, 197005

LSCO (INS) Pintschovius & Braden, *PRB* **60**, (1999) R15039.

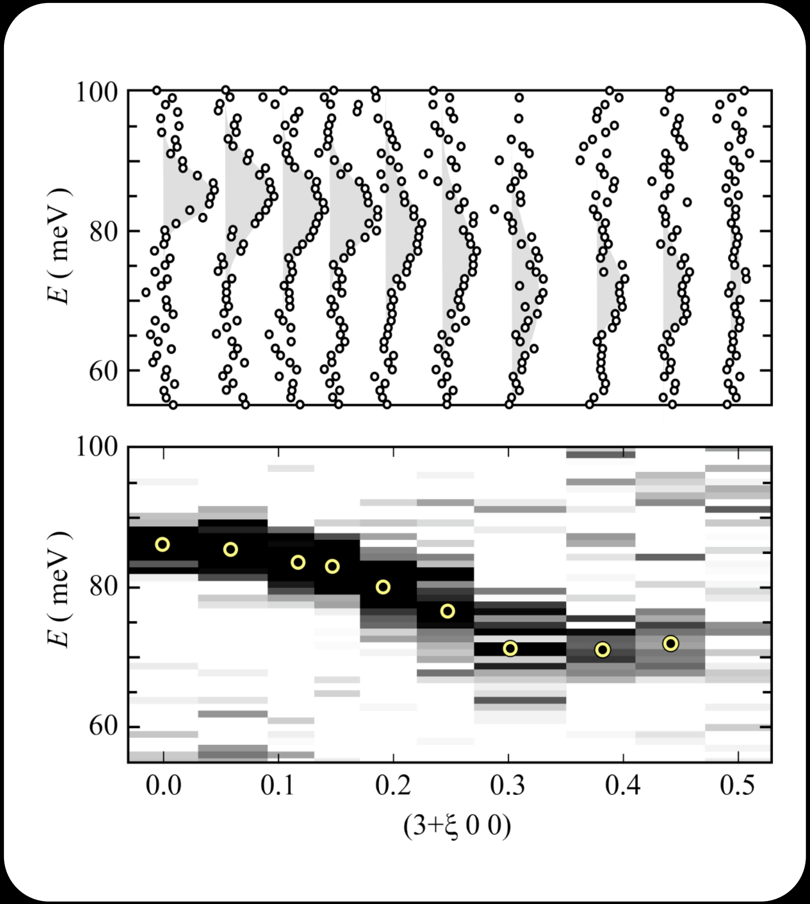
YBCO (INS) Pintschovius, Private Comm.

Effect of Stripes?



[a] H.-H.Klauss *et al.*, PRL85, 4590('00).

x = 0.16



Sasagawa, et al, 28aTD-2

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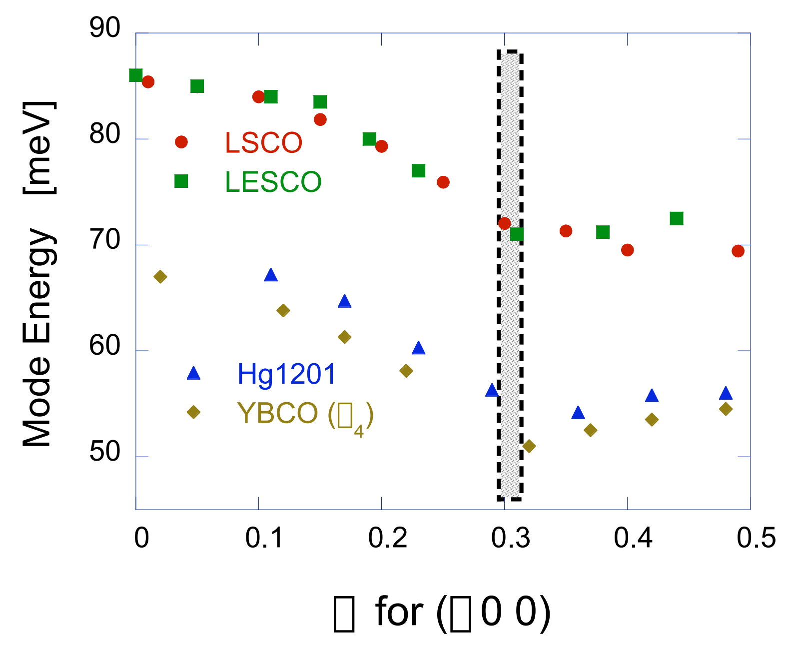
Bond Stretching Mode in Four Hole-Doped Samples

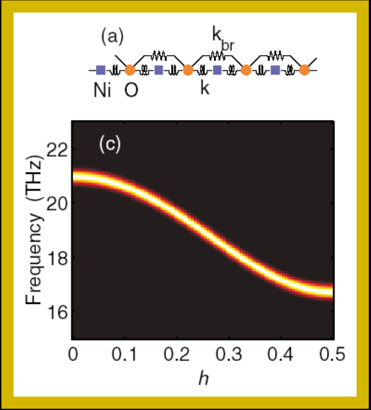


YBCO is INS
Pintschovius et al

"Anomalous" Softening is the default.

At x~0.3 have local minimum, and broadening.

Many Explanations of the Softening…



1. Next-nearest neighbor oxygen interaction (No kink)

   Pinstchovius (Tranquada…)

2. T-J models seems to give reasonable agreement for LSCO x~0.15

   Rosch & Gunnarsson, Khaliullin & Horsch

3. Vertex Correction in t-J Model (Kink)

   Ishihara & Nagaosa

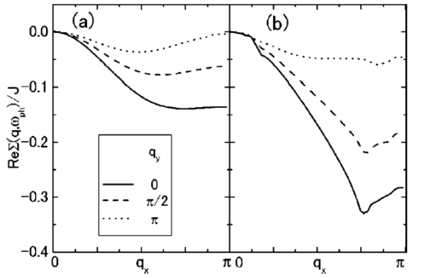


FIG. 11. Real part of the phonon self-energy without the vertex correction (a), and with the vertex correction (b). Parameters are chosen to be $t/J=3$, $t_1/J=-1.5$, $\tilde{g}_{off}/J=0.1$, $\omega_{ph}/J=0.5$, and $x=0.1$.

4. Ab-Initio Calc shows softening (No Kink)

   -> FS Nesting? Bohnen, Heid, et al

5. Disorder & effect on band structure as seen by ARPES?

   Janowitz et al.

Direct (Simple) T-Dependence

For low-energy phonons, that are coupled to the electron system, reducing T can remove the epc, and lead to abrupt line narrowing.

$$\hbar\square_{q} < 2\square$$

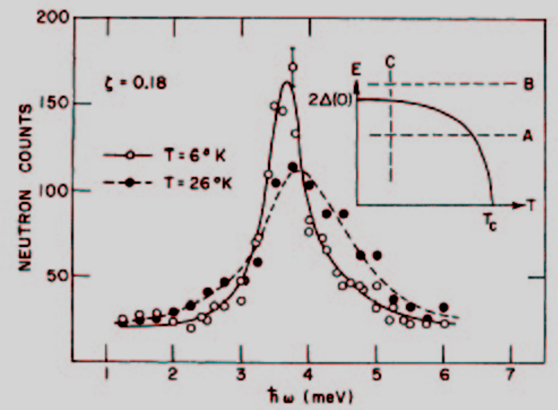


FIG. 1. The linewidth of low-energy $[\zeta\zeta 0]$ T_1 acoustic phonons broadens appreciably with increasing temperature. The inset shows graphically the effect of the energy conversion condition $\hbar\omega_p \geq 2\Delta(T)$ upon phonon damping with increasing temperature (scans A and B) and with phonon energy at a fixed temperature (scan C).

Nb_3Sn Axe and Shirane, prl, (1973)

More complicated, but also clear effects, in TWINNED YBCO

Bucking Mode

Pycka, *et al*, prl, 1993 ,
Reznik, *et al*, prl 1995

&

Stretching Mode

Pintschovius, et al. prb 2004

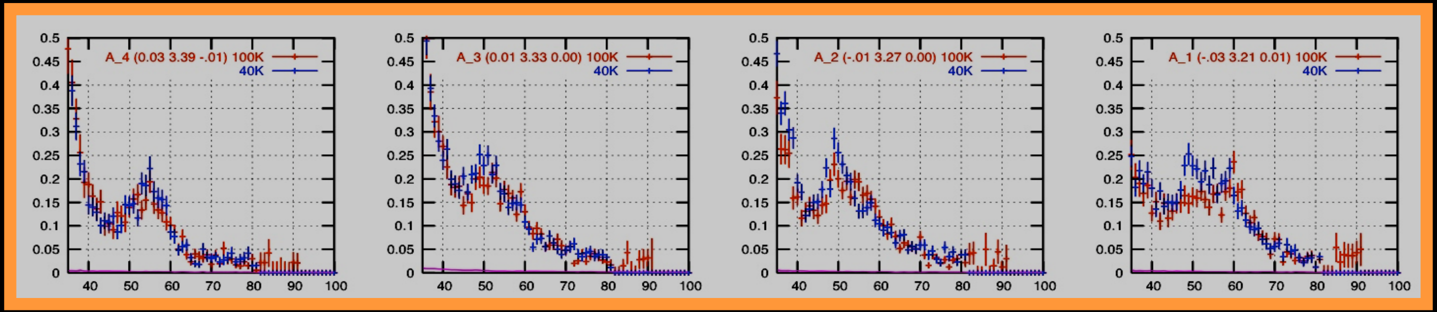
Toward T-Dependence using IXS

First Experiments with Bond-Stretching mode and (999)/3 meV resolution failed

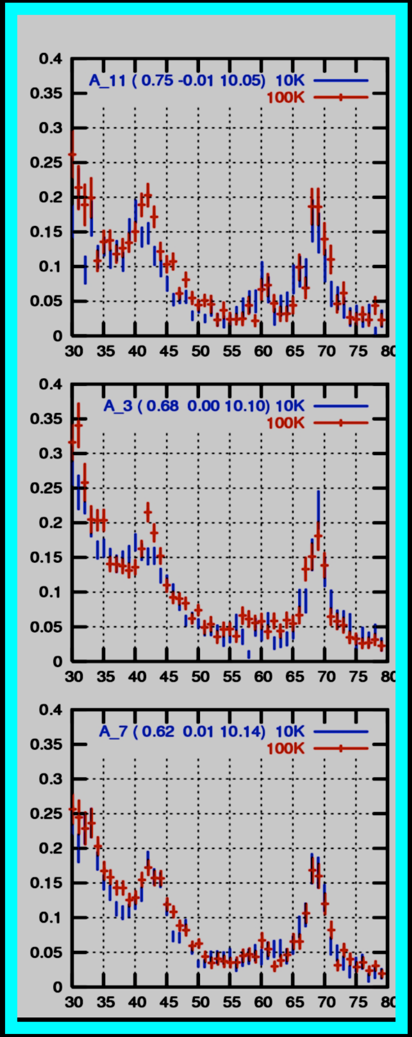
Test (888)/6 meV in December… Maybe OK

Buckling Mode: Successful…

Qualitative agreement with Reznik & Pycka



Bond Stretching Mode



Buckling Mode

New Instruments

Present instruments (ESRF, APS, SPring-8) are *highly*

Beamtime & Flux Limited

-> Additional facilities & additional flux are immediately useful.

High probability of new applications:

- New Geometries (Grazing Incidence, Phonon Microscopy)
- *More* Extreme Sample Environments (P, T, H, D).
- **Electronic Excitations?**
- Time Resolved (Stroboscopic) Studies?
- Non-Equilibrium Dynamics?
- Field-Induced Phase Transitions?
- (Biological materials?)

Do NOT forget liquids & glasses!

Low AND High Energy Photons

10 keV and 20 keV

	Characteristic	Low Energy (10 keV)	High Energy (20 keV)
Just □	Flux / meV	+	
	High Momentum Resolution	+	
	High Momentum Transfer		+
Materials	Scattering/Absorption		+
	High Refraction (Surfaces)	+	
	High Transmission	- (High Z)	+
Optics	Large ΔE/E for fixed ΔE	+	
	~ meV Resolution Optics	?	+
	~ 10 meV Resolution Optics	+?	
	Silicon Detector	+	

Main Comment: Both are good.

Low Energy (High Resolution) Optics

Silicon (Symmetric) Bragg reflections are limited:

$$\left.\frac{\Delta E}{E}\right|_{hkl} = \frac{1}{N} = \frac{d}{\overline{\Lambda}_{ext}}$$

N = Number of planes

$\overline{\Lambda}_{ext}$ = Average Extinction Length

No weak reflections with large d

=> ΔE>10 meV @ E<10 keV

meV Monochromator:

~10x30 urad²

Asymmetric Reflections

meV Analyzer:

10x10 mrad²

A Problem

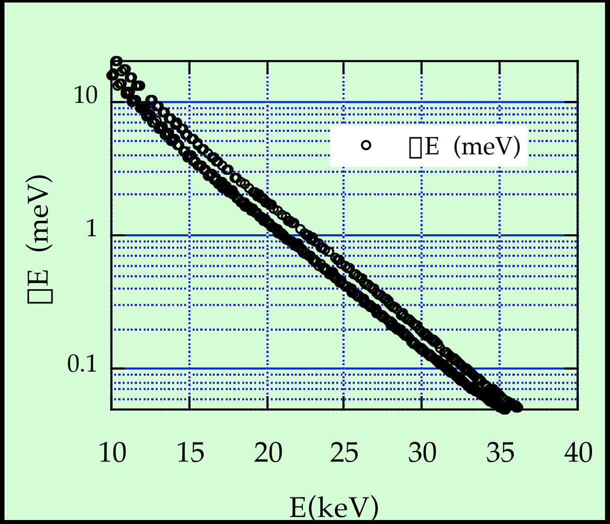
Analyzer Alternatives:

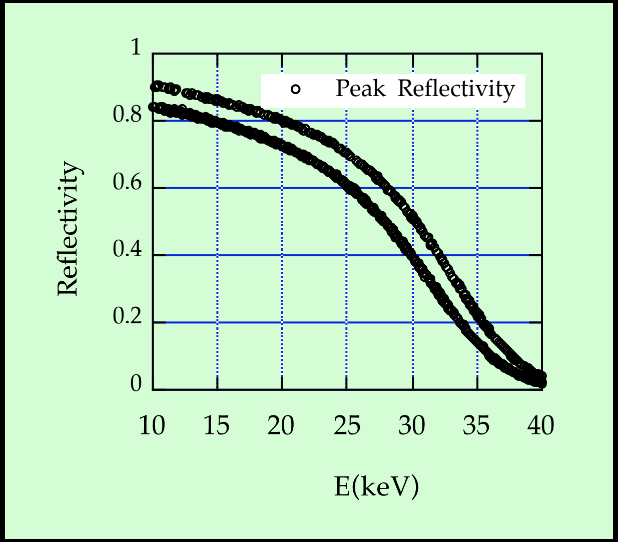
Not Silicon

Not Symmetric

(Not Bragg Reflections?

Nuclear Analyzer: Chumakov, Baron et al, prl, 1996)

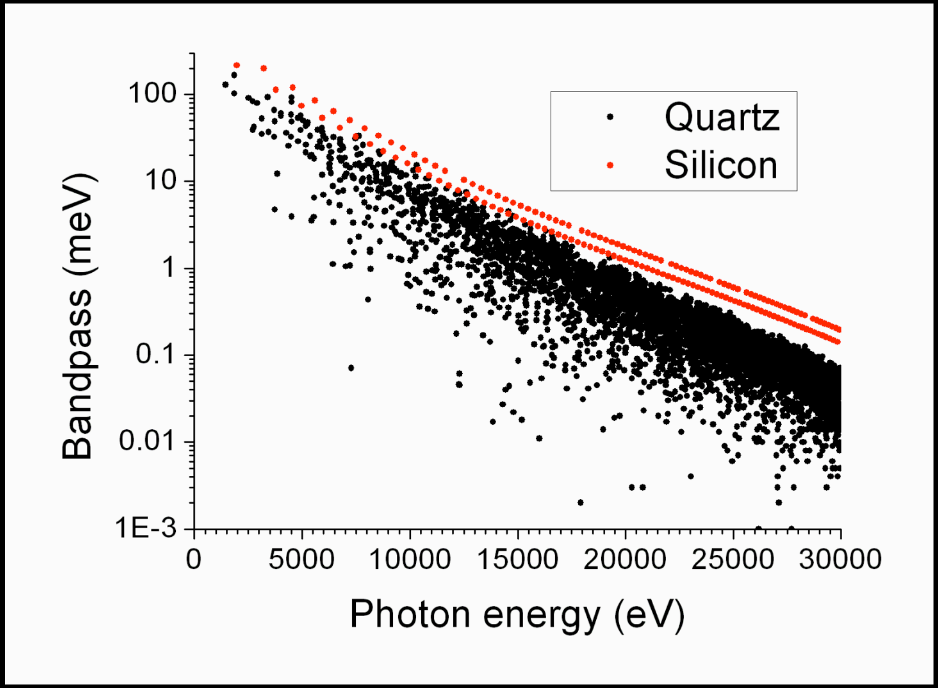


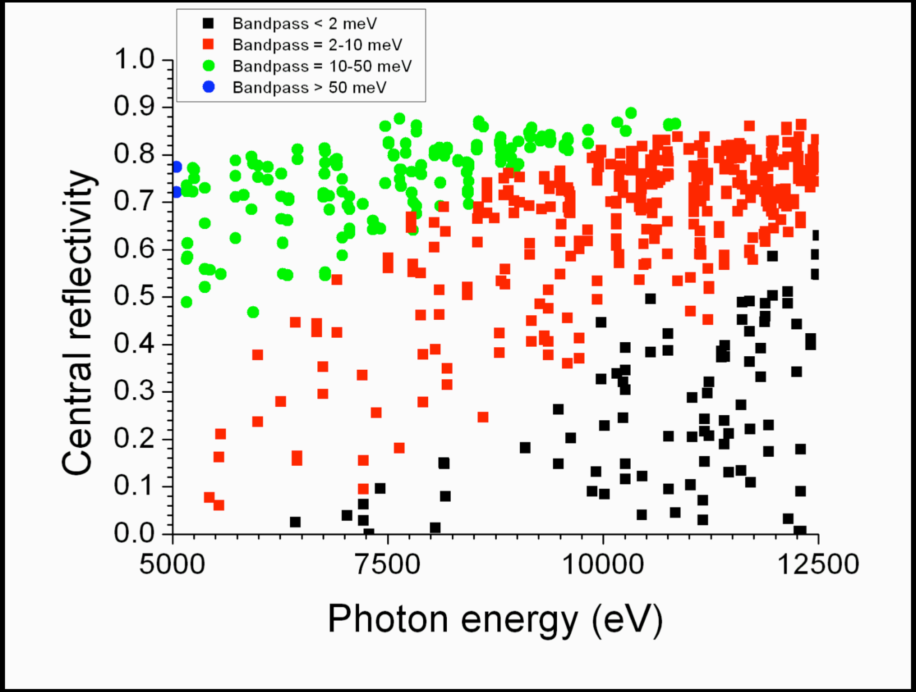


Quartz - SiO_2

Chiral Trigonal Lattice: $P3_121$

Hexagonal (a=4.9149, c=5.405) with 9 atoms/unit cell





-> Many Reflections

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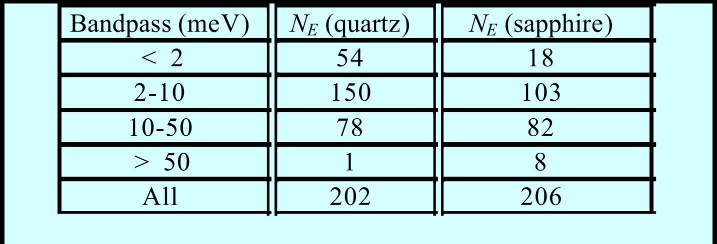
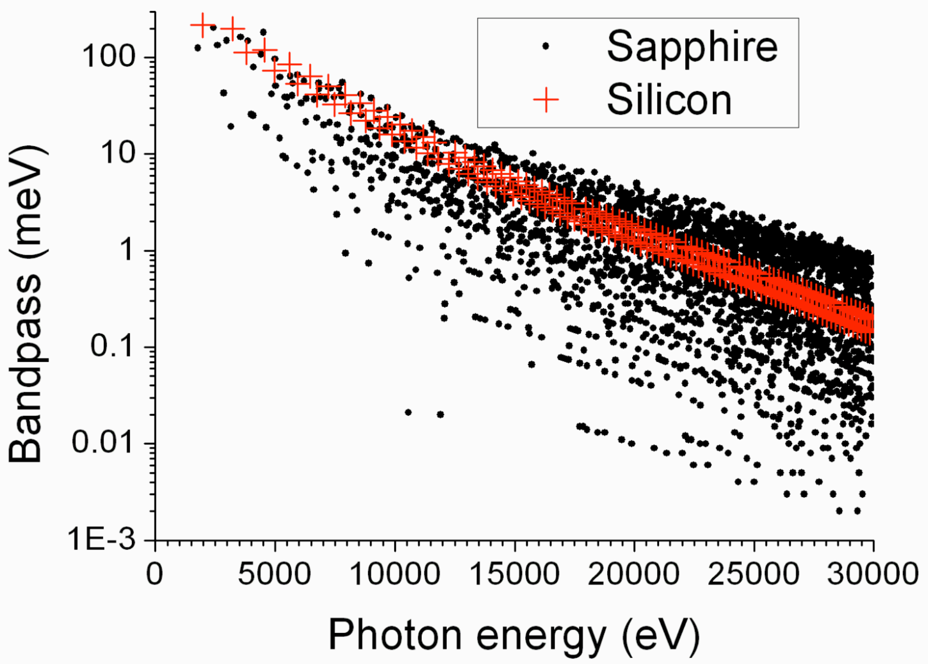
Quartz vs Sapphire

1. Quartz has a relatively low Debye temperature
   -> Many narrow bandwidth reflections at low energy
   -> Reflectivity drops fast at high energy



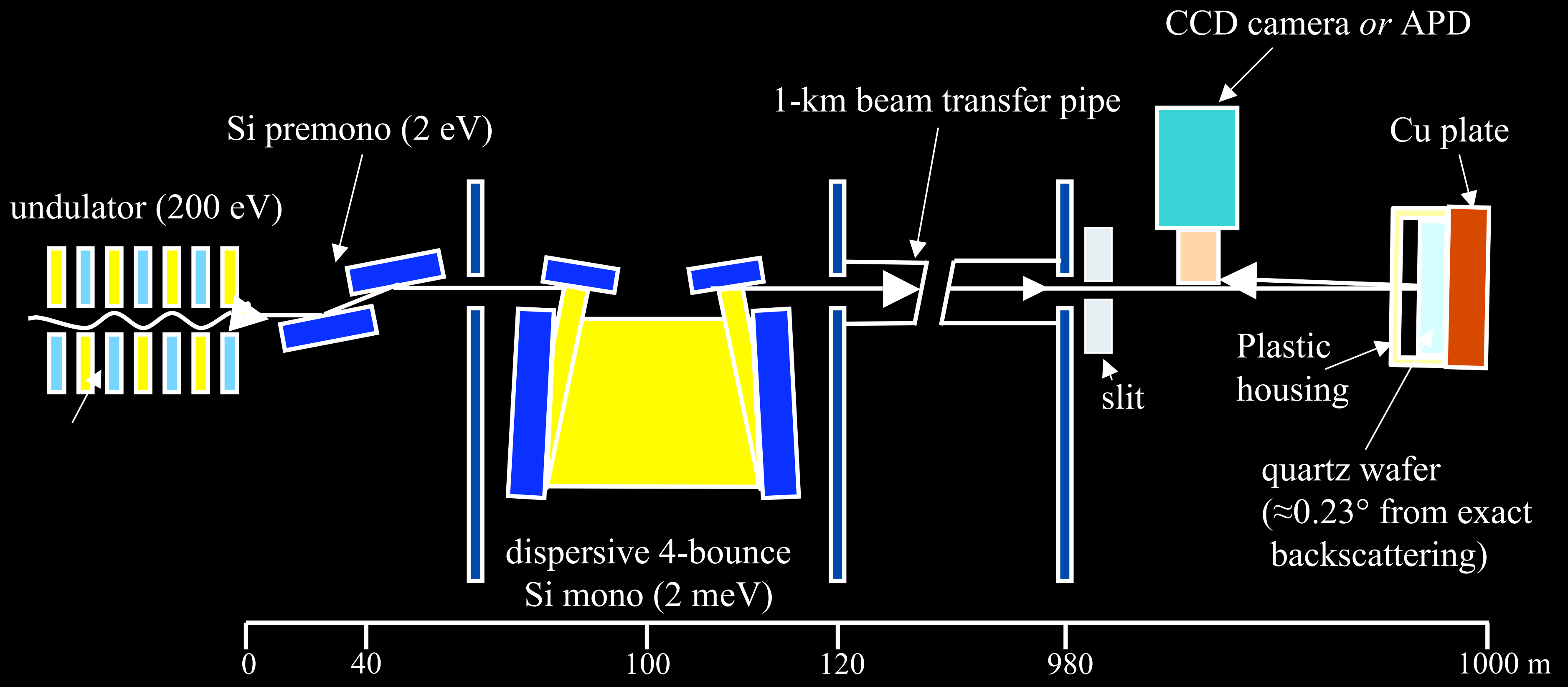
Bandpass (meV)	N_E (quartz)	N_E (sapphire)
< 2	54	18
2-10	150	103
10-50	78	82
> 50	1	8
All	202	206

5-12 keV



2. Crystal Quality: Large area, high-quality crystals readily available. (Crystal Oscillators)

Backscattering Topography at the 1 km Beamline (BL29XUL)



Large Area (10x10 mm^2), Highly Parallel (10x10 urad^2), Highly Monochromatic (2 meV at 10 keV)

AQRB, April 2006

Backscattering Topographs of Quartz

Quartz (7 -4 -3 4) Reflection

E = 9978 eV

$\square_{abs}$ = 210 um,   Av. $\square_{ext}$ = 290 um

Bandwidths (nominal)

2.0 meV incident

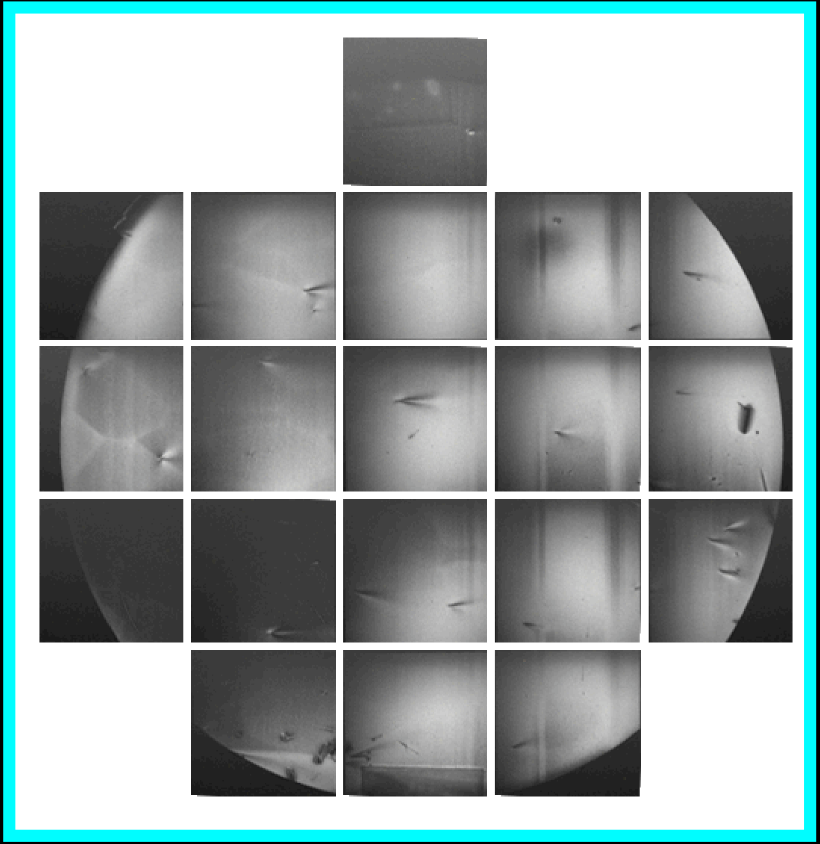
1.9 meV for quartz

Backscattering Sensitivity:

~ $2 \times 10^{-7} = \square d/d$ (sensitive)

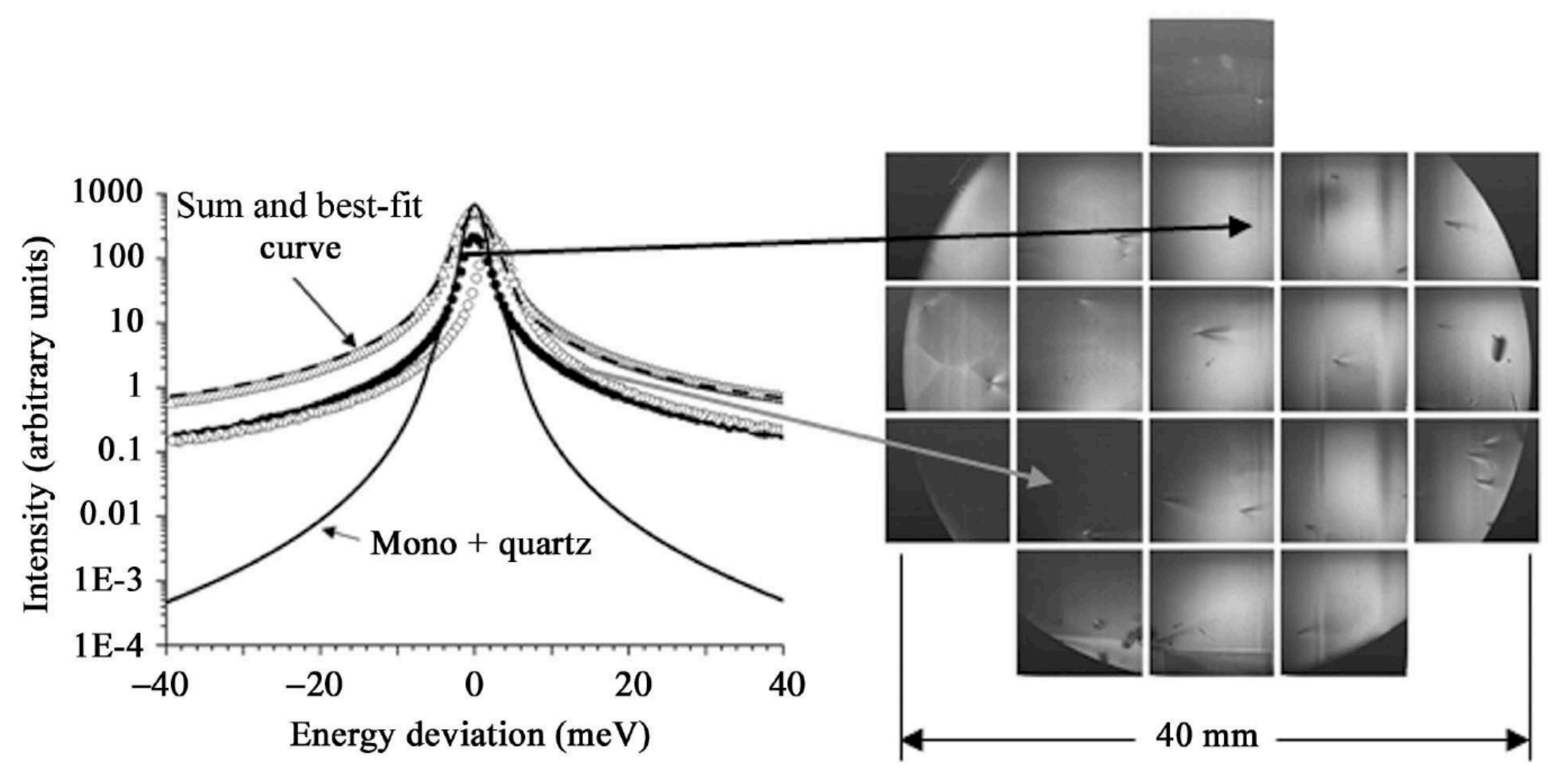
~ 50 urad (not sensitive)

Note: $\square_{abs} \sim \square_{ext}$



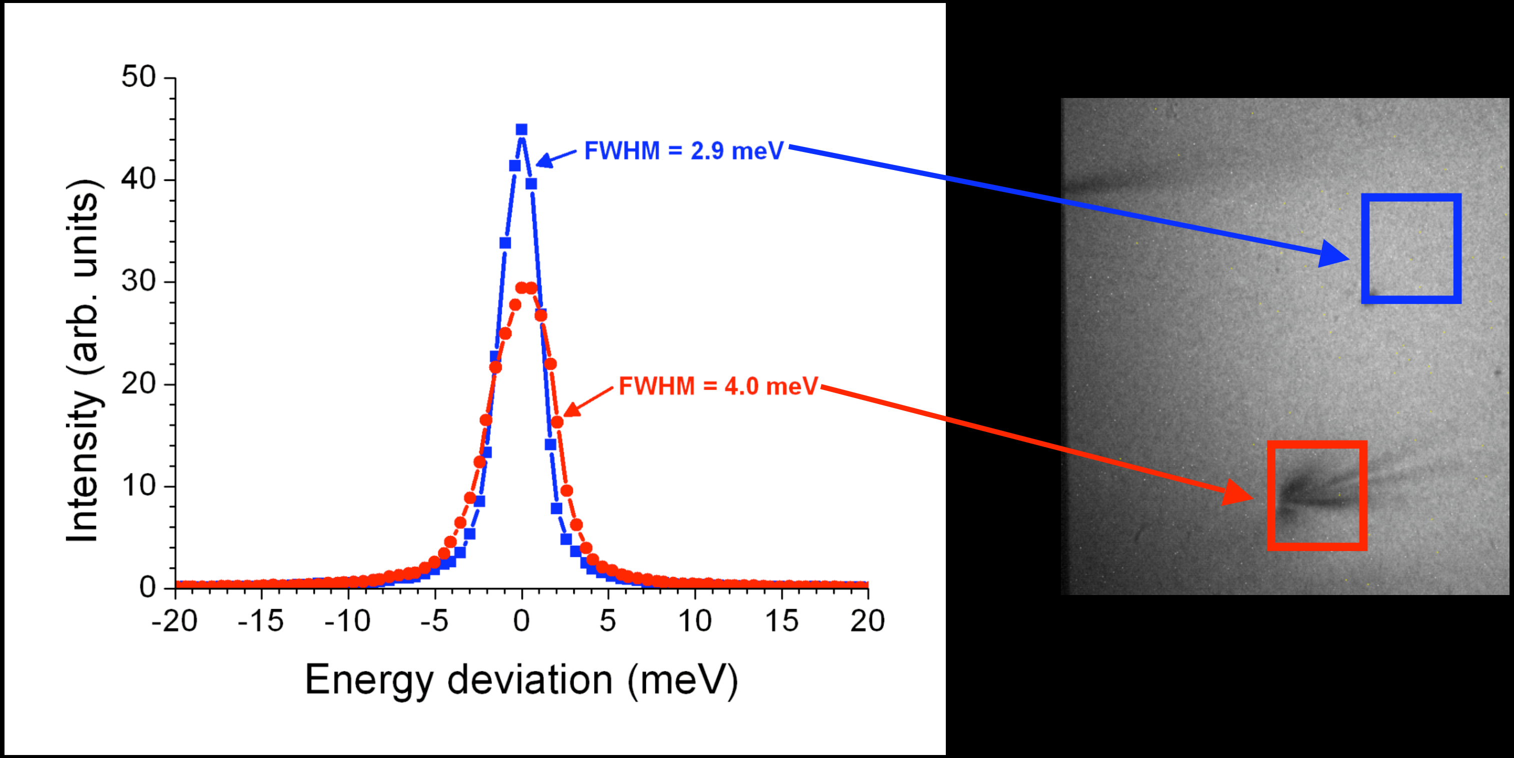
J.P. Sutter, *et al*, JSR

Bandwidth (FWHM) Good, But Some Tail



Measured bandwidth (10 cm^2) **3.7 meV**

Resolution Worsens in Defect



Optimized Long Undulator Beamlines

Planned Facilities: NSLS II (15m?), PETRA III (1x20m)

SPring-8: 3 x 30m Straights

Electron Optics:

Require e beam size small enough for minimum gap of undulator.

SP-8 Standard 4.5 m ID allows 6.5 mm gap ($\square_{ymin}$=5.6m, not opt.).

SP-8 Standard Long ID allows 12 mm gap ($\square_{ymin}$= 14.1m, opt,)

Undulator:

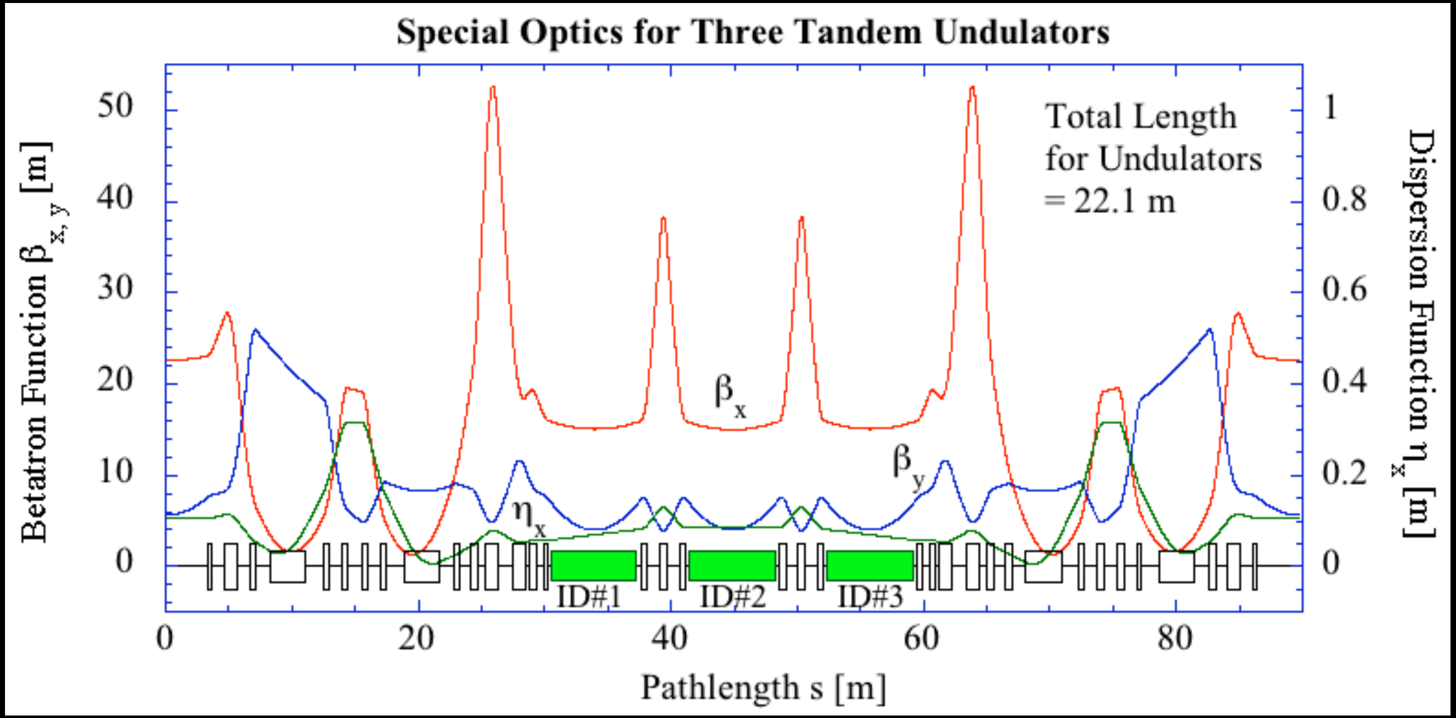
Generally best performance if use lower harmonics:

Highest Brilliance

Highest Flux/Power Load

For a long undulator: “Diffraction Limit” superceded by the “e Energy Spread Limit ” (0.1%)

Electron Beam Parameters

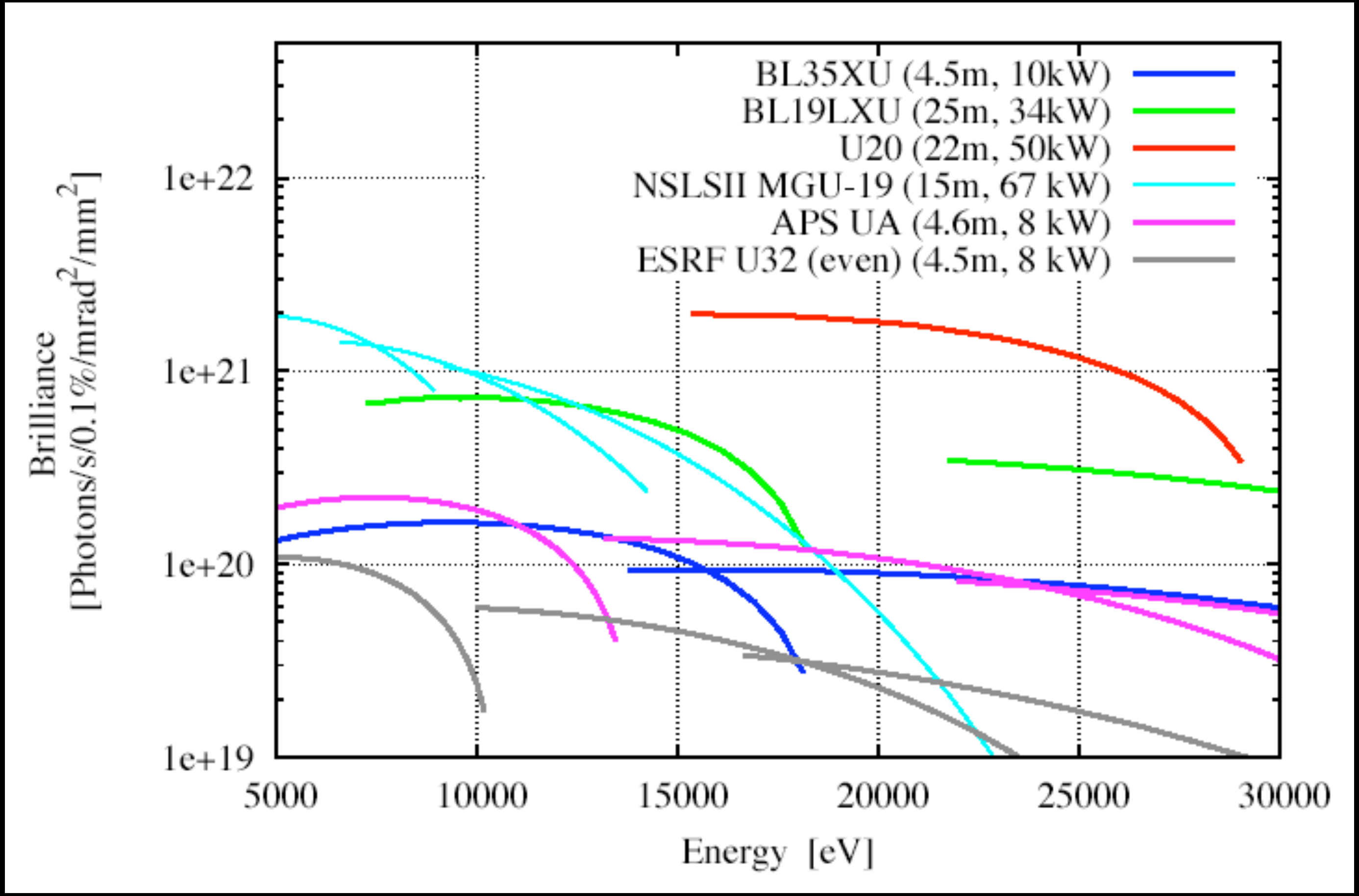


Preliminary Electron Optics, Feb 06

K. Soutome and H. Tanaka

With Intermediate Focusing, 30m Straight -> 22m Undulator
(3 x 7.3 m sections + ~0.2m to phase undulators)

Brilliance



Calculations Using SPECTRA
(Tanaka & Kitamura)

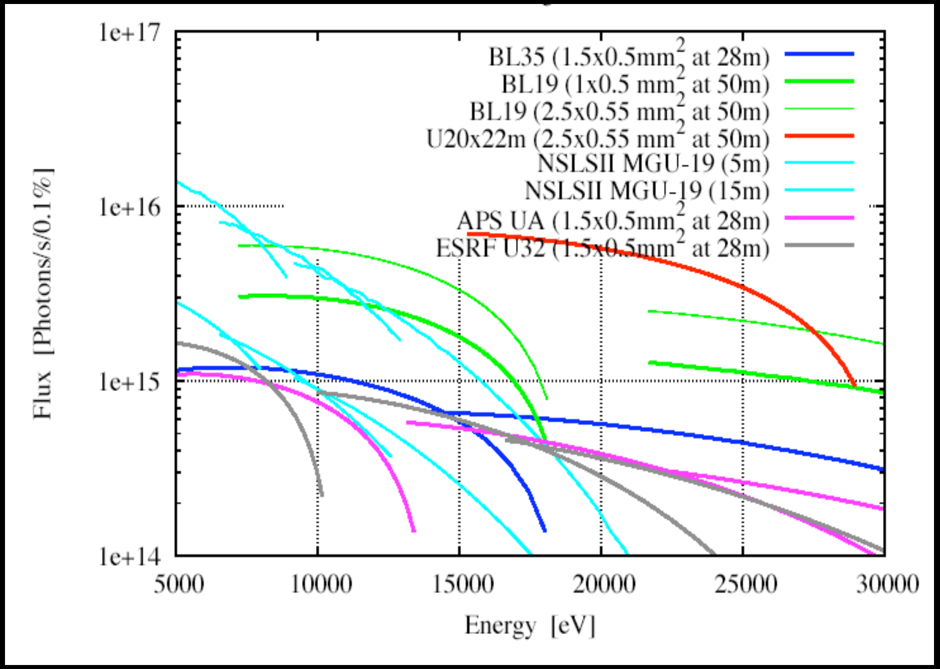
Note: SPring-8 results are most reliable.
Note: ESRF for high beta - so "artificially" low.
Note: Undulators always being improved.

Note: NSLS II *Superconducting* U14 x 5m, K_{max}=2.28 => >40 kW!!

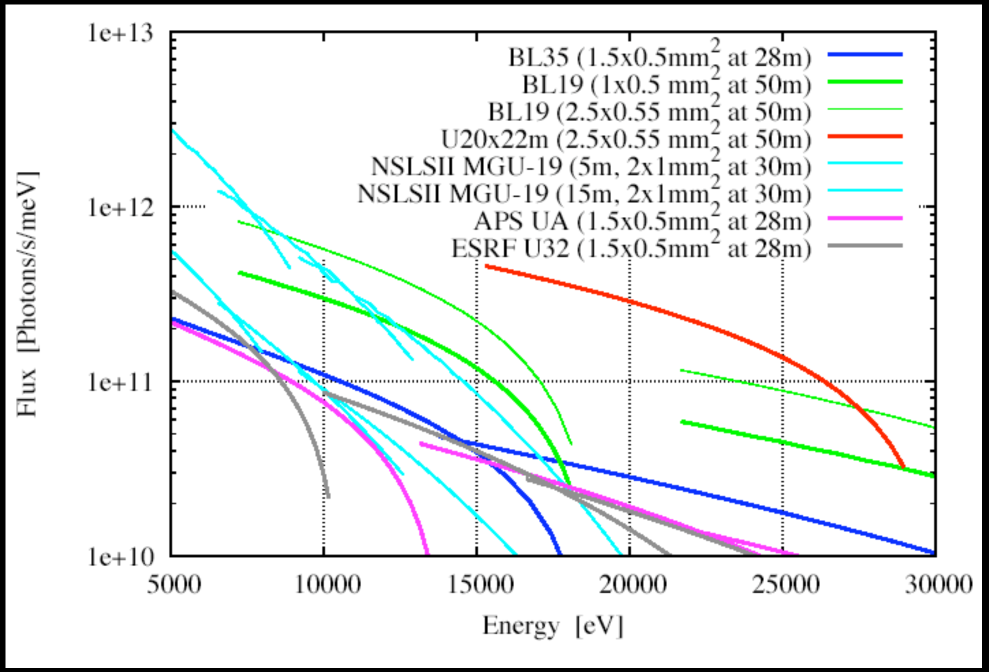
AQRB, April 2006

Flux

Photons/s/0.1%BW

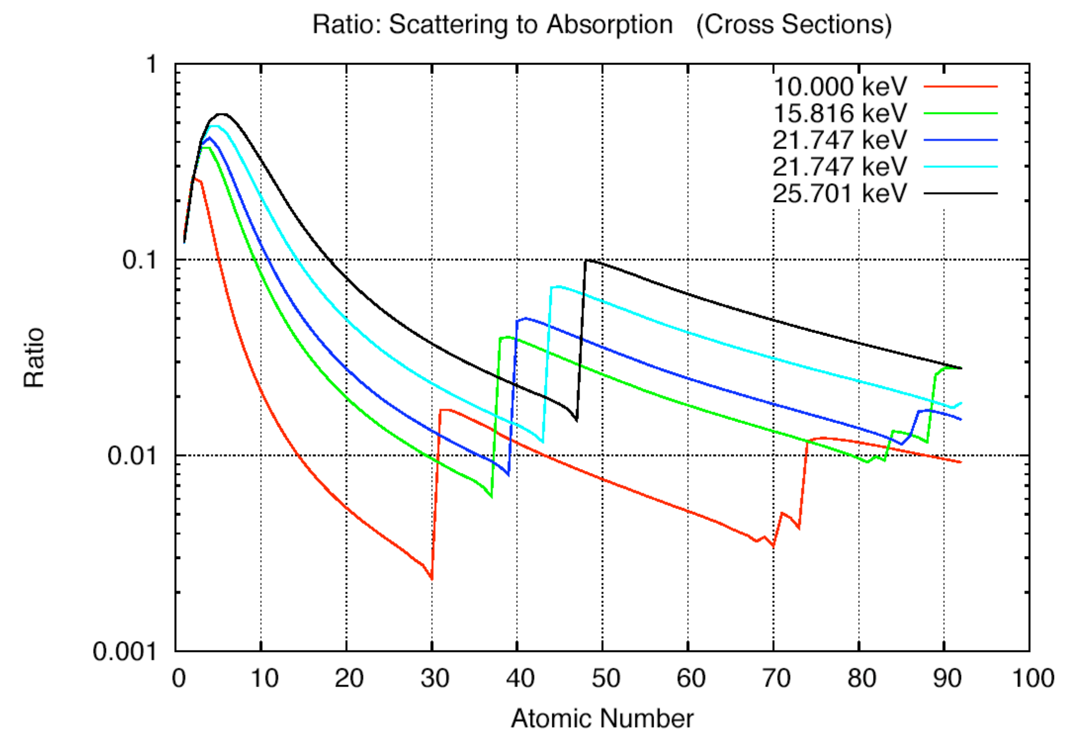


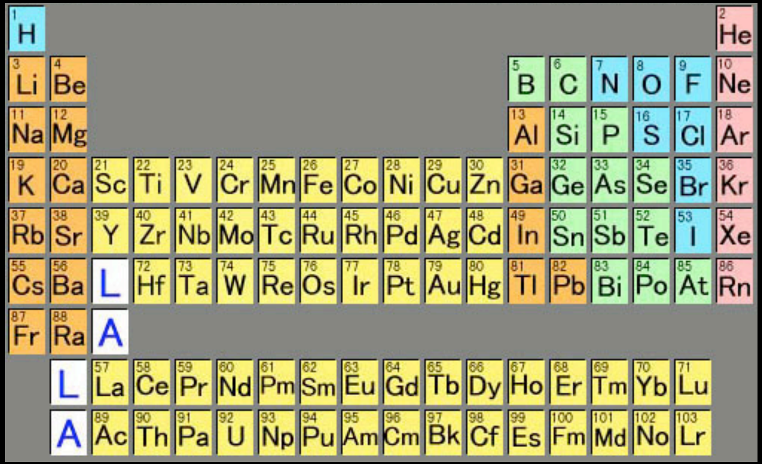
Photons/s/meV



Note: Slit size dependent! Attempt to choose reasonable values.

Scattering vs Absorption





1 H																	2 He
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
55 Cs	56 Ba	L	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
87 Fr	88 Ra	A															
	L	57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu	
	A	89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr	

$$\square_{Scatt} = (Z + f')^2 \; r_e^{\,2}$$

$$\square_{Abs} = 2 \; r_e \; \square \; f''$$

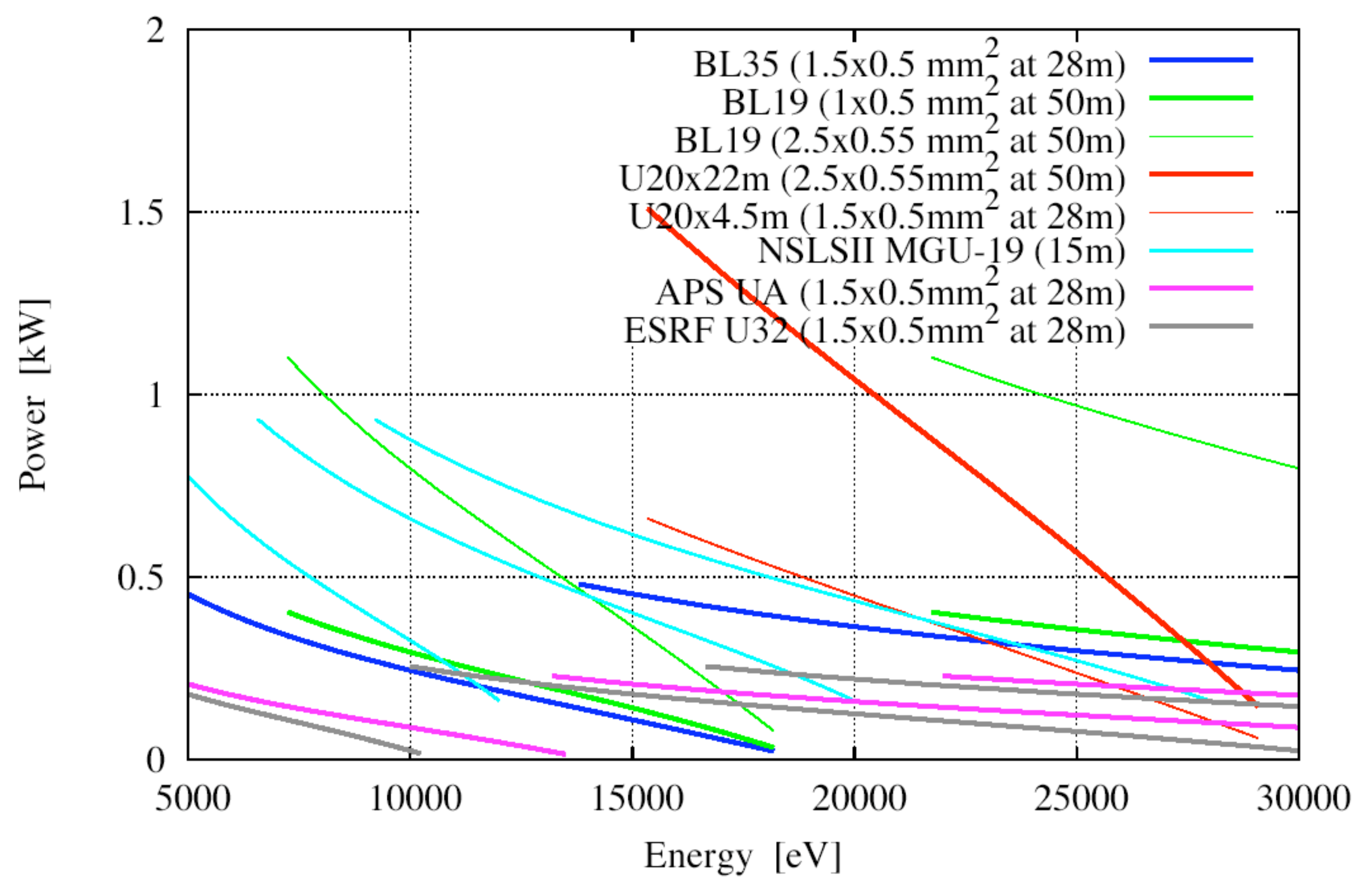
Lighter elements and higher energies generally favorable.

(note: heaviest element in a sample usually dominates)

ID Parameters

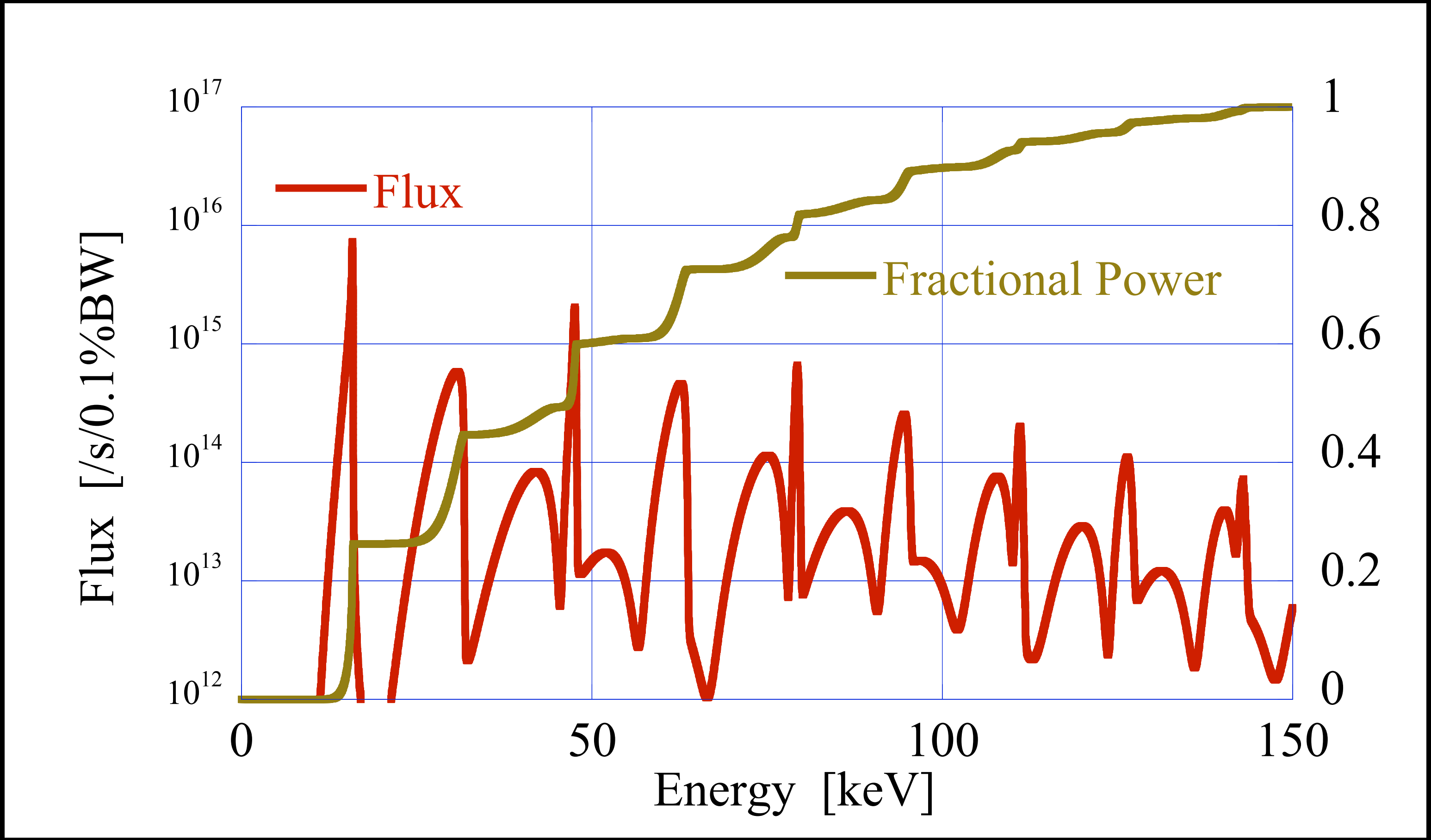
	BL35XU	BL19LXU	New BL
Period (mm)	32	32	20
Length (m)	4.5	25	22
Periods	140	781	1100
B_0 (T) / G_{min} (mm)			1.15 / 6.5
Gap (mm)	9 – 50 (8-50)	12 – 50 (8-50)	6.5 - 50
Kmax	2.3 (2.5)	1.76 (2.6)	1.4
Max Power (kW)	10 (13)	35 (67)	50
Bmax (T)	0.77 (0.85)	0.58 (0.87)	0.75
1st Harmonic	5.2 - 16	7.5 – 17+	15.3 – 27+
3rd Harmonic	15.5-29	22.4 – 30+	46. - ??
Emittance (nm-rad)	3.4		
Coupling (%)	0.2		
Energy Spread (%)	0.1		
Horizontal Beta (m)	22.6	21.7	16
Vertical Beta (m)	5.6	14.1	5
Horizontal Disp. (m)	0.11	0.1	0.1
Horizontal Size (μm)	298	290	254
Vertical Size (μm)	6.2	9.8	5.8
Horizontal Div. (μrad)	12.3	12.5	14.6
Vertical Div. (μrad)	1.1	0.7	1.2

Power in Central Cone



> 500 W is a problem for cryogenic silicon (SPring-8)

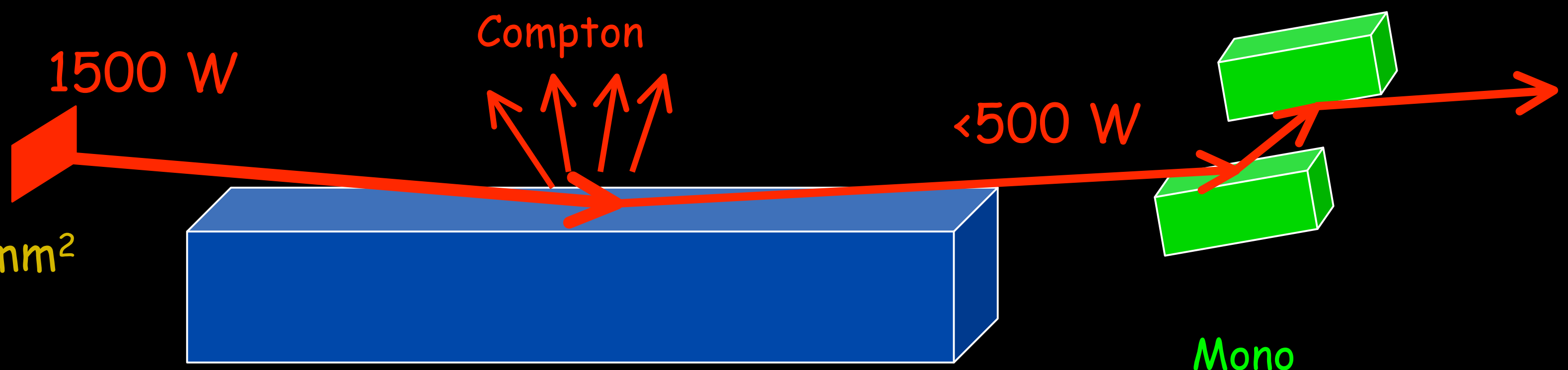
Spectrum at Minimum Gap



Spectrum through 0.55 mm x 2.5 mm slit at 50m

Power Load In Central Cone: 1.5 kW

Strategy: Use LN_2 cooled mirror *and* LN_2 cooled Mono



~300 W
Compton

1500 W

<500 W

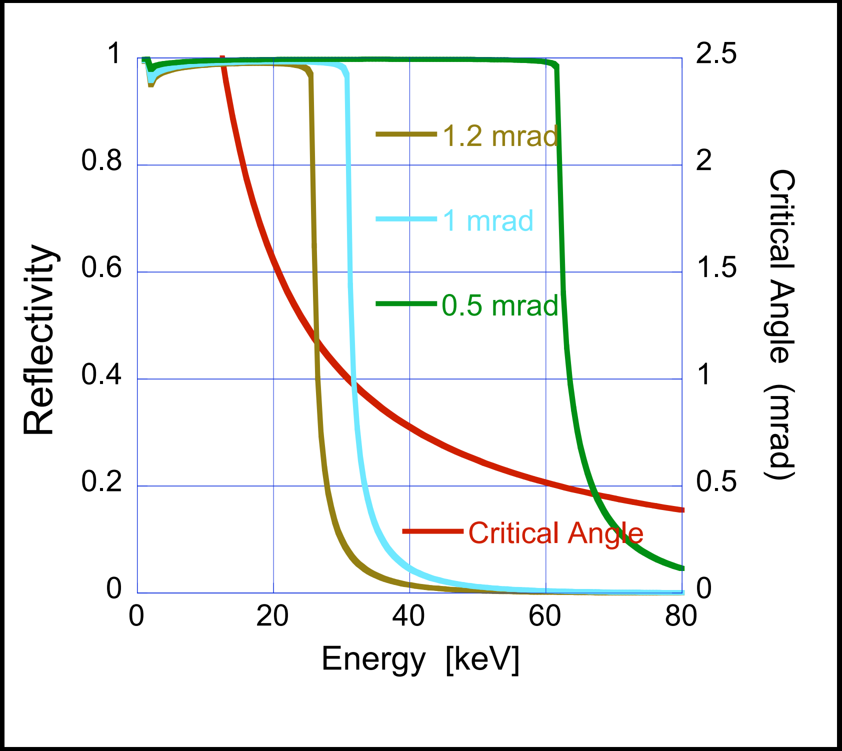
2 W

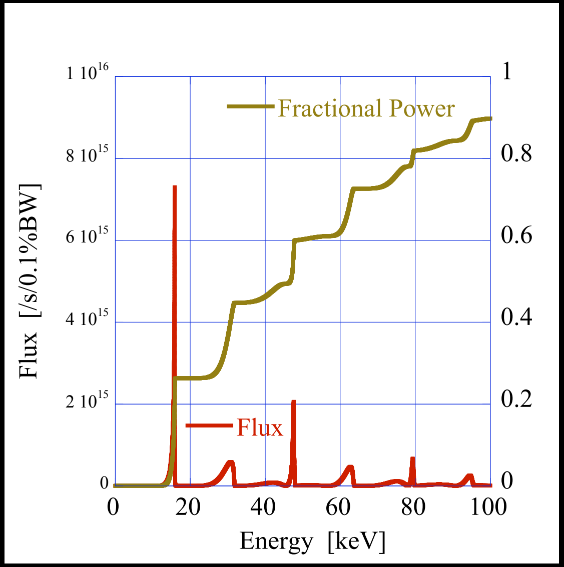
~2.5x0.55 mm^2
At 50 m

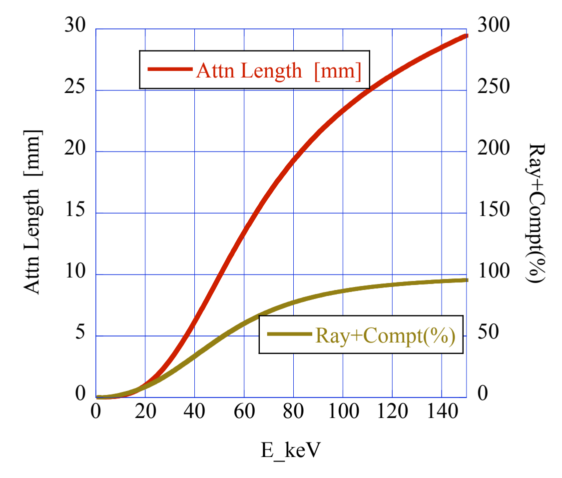
Si Mirror, 1 m long, θ_g = 1.1 mrad
Absorbed Power: ~700 W
(Horizontal Scattering ?)
(Mounting!)
(Compton Absorber Required)

Mono
Slightly improved cooling
(400W Now Possible
-> 500W)

Mirror Calcs







>2/3 of the power is at high energies

Choose grazing Angle ~ 1.1 mrad

AQRB, April 2006

The Spectrometer

Monochromator: In-Line or Backscattering?

Mis-Matched -> In Line
Otherwise -> ?

Sample Area: Microfocus mandatory. Nanofocus?
Extreme environments?
Unusual geometries? Grazing incidence & ?

Spectrometer Arm: How big an arm? (10m? 5m? 2m ?)

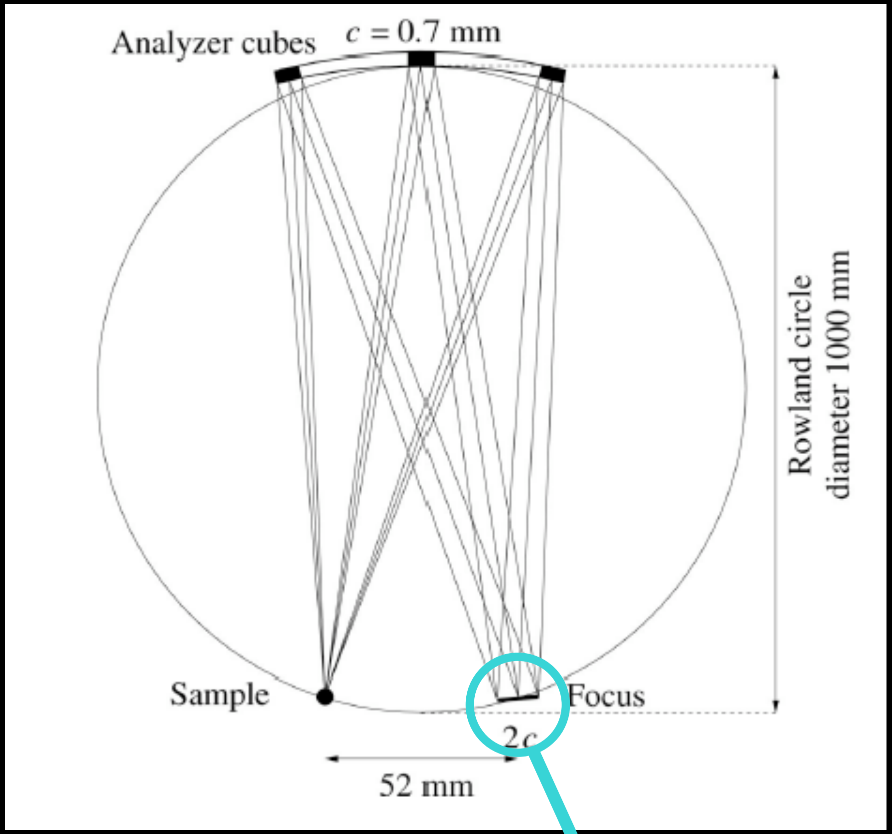
How large a diameter analyzer (5-10 mrad?)

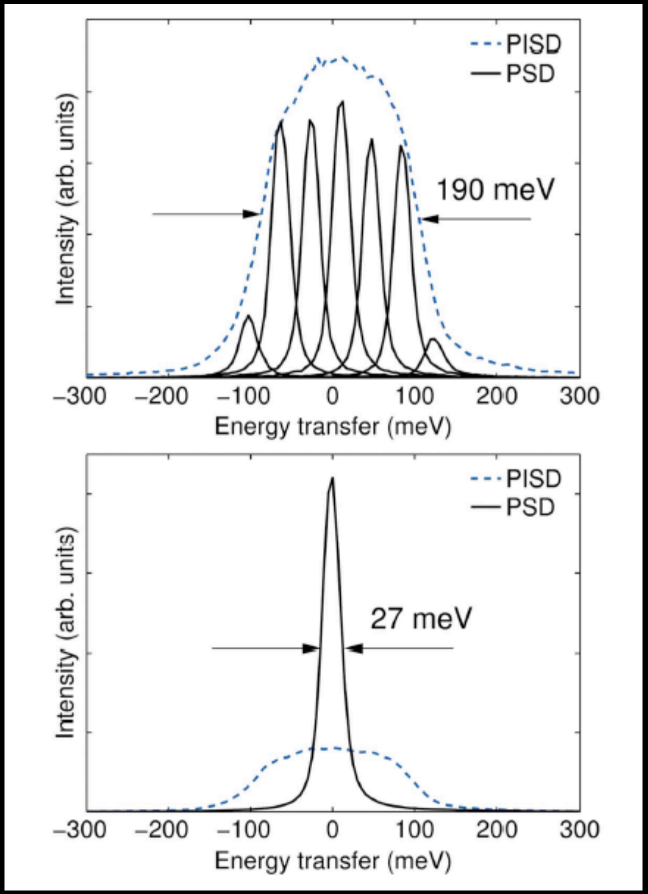
How many analyzers? 3x5 ? 4x5?

Dispersion compensation PSD? Not for meV?

"Dispersion Compensation"

Huotari et al, JSR 2005





Position Sensitive Detector

190 meV -> 27 meV
R = 1 m, Si(555) 9.9 keV

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Simplest Case Mathematics

(Negligible Source Size, Perfectly on Rowland Circle, Not Too Close to Backscattering)

From Braggs Law: □= □□ □□

□= □E/E due to "Divergence"
□= Deviation from Backscattering
□□ = Angular Divergence

Monolithic Large Detector :
c = analyzer crystallite size

□□ = c/R

c ~ 1 mm

Area Detector :
p = pixel size

□□ = p/2R

p ~ 0.1 mm

Geometrical Gains of x20 possible:

Reduce the radius for a fixed energy resolution.
Improve the resolution for a fixed radius.

ESRF Showed: □= 26 mrad, □= 27 meV/9.9 keV = 2.7 $\times 10^{-6}$, (p ~ 0.1 mm)

For ~meV resolution….?

1 meV @ 20 keV:

□= 5x10^{-8}
p = 0.1 mm
R= 5 m

□$_{max}$ = 5 mrad
(detector offset 50 mm)

(at 10 keV things a factor of 2 easier -> R=2.5m or □$_{max}$=10 mrad)

Most interesting if you want **MANY (>>10)** analyzers
or are severely constrained to a small arm.

Note Also:

Rowland circle requirement is VERY severe.
Detector efficiency an issue at higher energy (Silicon!).
(Source size contribution possible)

Question: If the source is moved out of the focus, can the Roland circle be relaxed?

Collaborators - Beamline

BL35XU

Design: T. Ishikawa[1,2], K. Takeshita[1], S.Goto[1], T. Matsushita[1]
Commissioning: Y. Tanaka[2], D. Ishikawa[2], H. Thiess[1] T. Mochizuki[1]
Operation: S. Tsutsui[1], J. Sutter[1], D. Ishikawa[2]
Upgrades: J. Sutter[1], S. Tsutsui[1]

Quartz: J. Sutter[1], T. Ishikawa[1,2]

New Beamline Design: H. Tanaka[1], T. Tanaka[1], T. Mochizuki[1], S. Goto[1]
S. Takahashi[1], T. Uruga[1], H. Ohashi[1]

[1] SPring-8/JASRI [2] SPring-8/RIKEN

Collaborators - High T_c Materials

Hg1201	H. Uchiyama[1], S. Tsutsui[2], Y. Tanaka[3], W. Hu[1], A. Yamamoto[1], S. Tajima[1] & Y . Endoh[5]
YBCO	T. Fukuda[4], J. Sutter[4], S. Tsutsui[2], J. Mizuki[5], H. Uchiyama[1], T. Masui[1], S. Tajima[1], & Y. Endoh[5]
LSCO	T. Fukuda[4], J. Sutter[1], J. Mizuki[4], K. Ikeuchi[5], K. Yamada[5], S. Tsutsui[2]
LESCO	T. Sasagawa[6], M. Misawa[6], H. Takagi[6]
Bi2212	H. Uchiyama[1], S. Tsutsui[2]

[1]ISTEC-SRL-> Osaka U., Rikukyou U. [2]SPring-8/JASRI [3]SPring-8/RIKEN
[4]SPring-8/JAEA [5]IMR/Tohoku U. [6]Univ. Tokyo & CREST/JST