20 Minute Introduction to IXS

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Set the scene for discussion

Basically Impossible

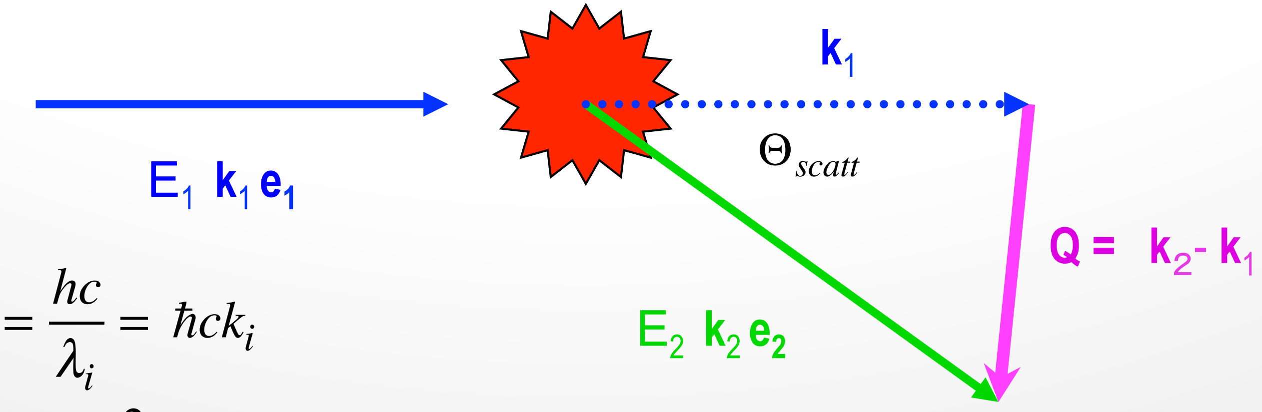
Mention some useful common points – essentially for reference

Bring up methodological aspects – what might be changed

Touch on some possible scientific areas of interest
to be addressed/expanded upon

Followed with contributions from participants:
(DeBeer -> Emission Spectroscopy, HR-RIXS, NIS)
Monaco -> NRIXS & HR- RIXS
Cramer -> NIS
Chumakov -> NIS vs IXS, Nuclear Analyzer (?)
Sinha -> Phonon form factor

Inelastic X-Ray Scattering



$$E_i = \hbar\omega_i = \frac{hc}{\lambda_i} = \hbar c k_i$$

$$k_i = |\mathbf{k}_i| = \frac{2\pi}{\lambda_i}$$

Two Main Quantities:

Energy Transfer

$$E = E_1 - E_2 = \hbar\omega$$

Note: For Resonant Scattering
E_1 and E_2 and Polarization
are also important

Momentum Transfer

$$\mathbf{Q} = \mathbf{k}_2 - \mathbf{k}_1$$

$$|\mathbf{Q}| \approx \frac{4\pi}{\lambda_i}\sin\left(\frac{\Theta_{scatt}}{2}\right)$$

Periodicity $d = \frac{2\pi}{|\mathbf{Q}|}$

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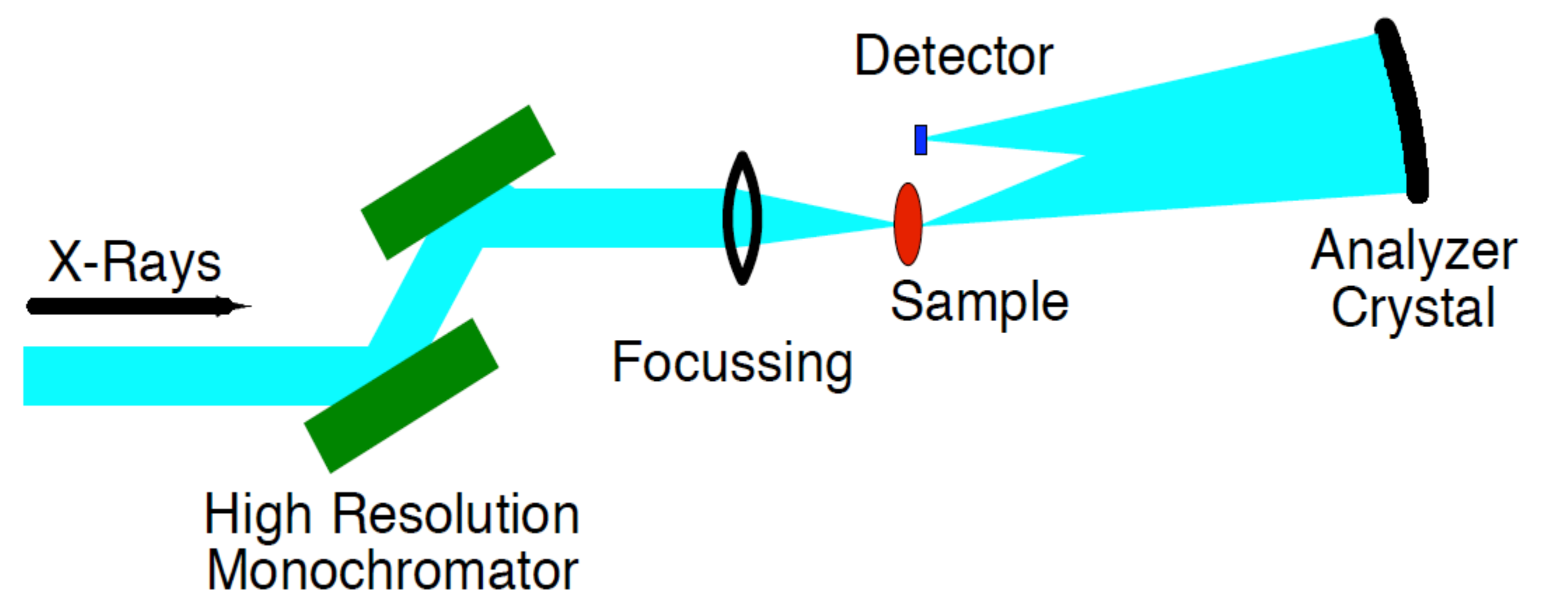
The IXS Spectrometer
An Optics Problem

Main Components

Monochromator:
Modestly Difficult
Accepts 15x40 μrad^2

Sample Stages
Straightforward
Only Need Space

Analyzer:
Large Solid Angle
Difficult



Common Features

Photon In -> Photon Out

Measure intensity as a function of energy transfer (Mostly…)

Measure at fixed (range of) scattering angles -> **Q** typically well defined

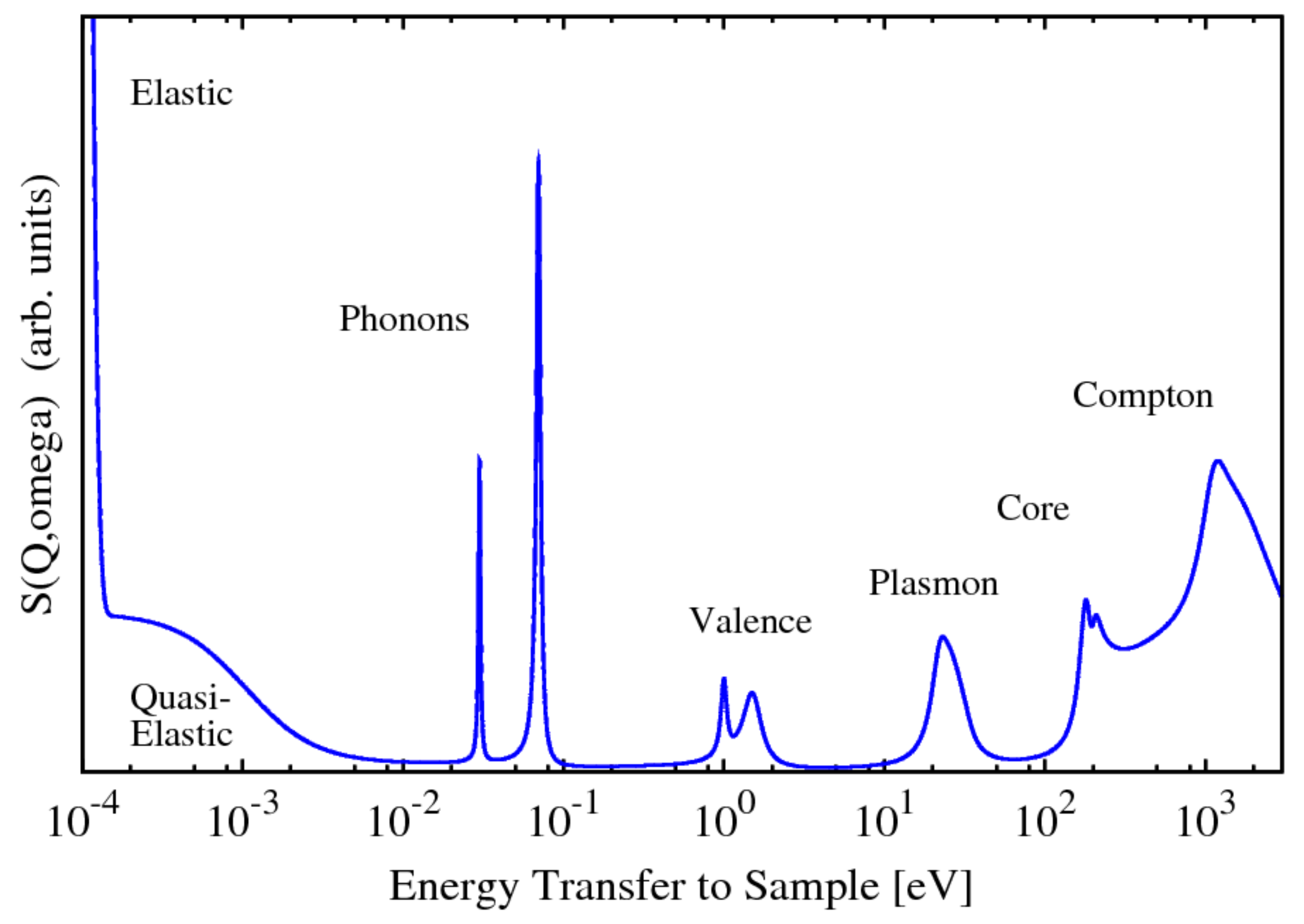
Dividing Features

Energy Resolution & Transfer
Resonant vs. Non-Resonant (Cross section, Resolution, Q)
Electronic vs. Nuclear

Momentum (**Q**) Resolution and Transfer

Sample Geometries (layers, surfaces, micro-crystals, liquids, allowable sample volume)
Sample Environments (T, P, H, E, …)

The IXS Zoo



Elastic
Quasi-
Elastic
Phonons
Valence
Plasmon
Core
Compton
S(Q,omega) (arb. units)
10^{-4}
10^{-3}
10^{-2}
10^{-1}
10^{0}
10^{1}
10^{2}
10^{3}
Energy Transfer to Sample [eV]

The IXS Zoo II

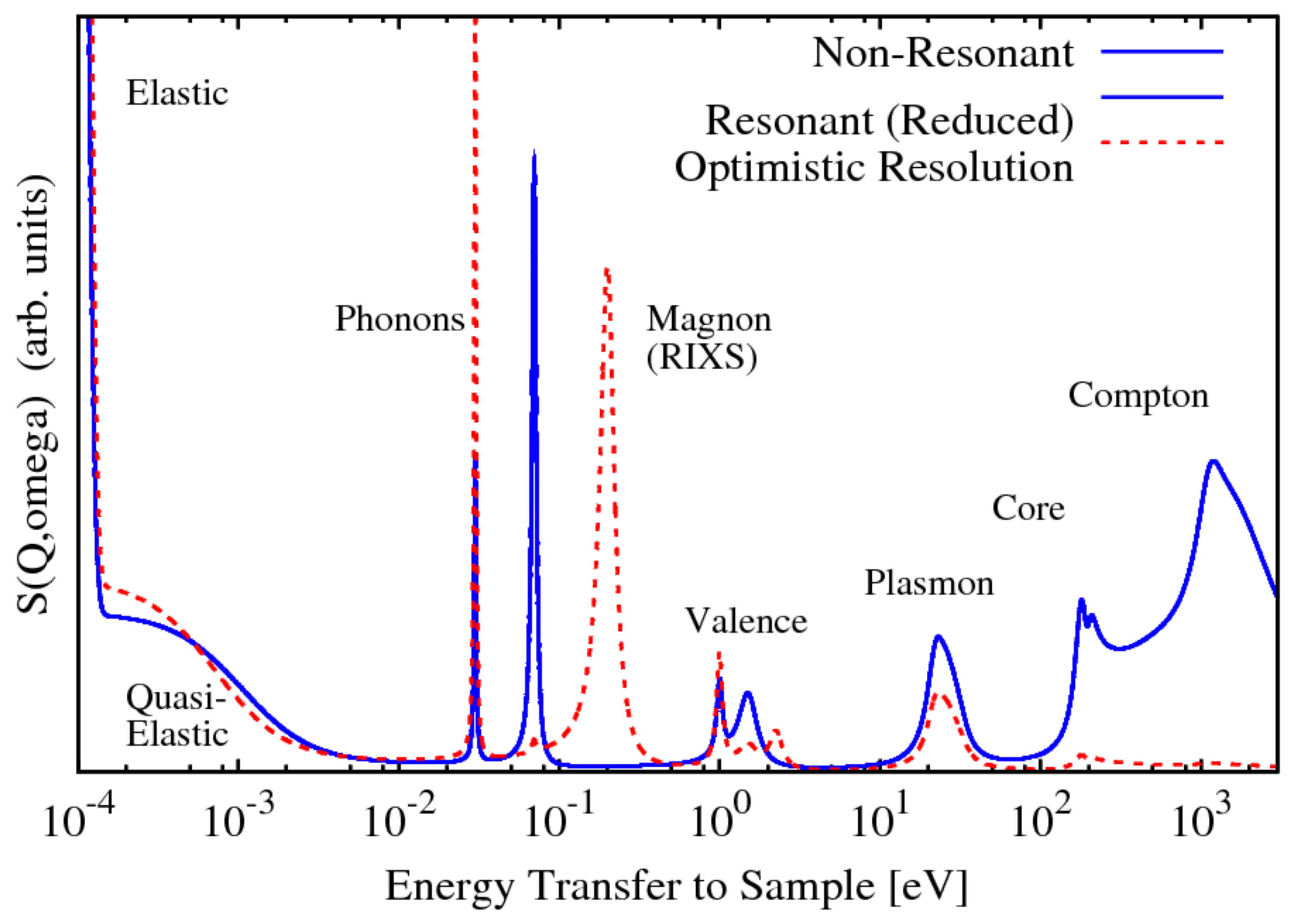


Table Of IXS Techniques/Applications

Technique	Comment	Energy Scale	Information
X-Ray Raman	IXS version of xafs High resolution case?	E_{in}>4 keV ΔE~500-1000 eV Res: ~eV	Chemical Bonding
X-Ray Emission	High resolution case?	E_{in}>4 keV ΔE~1-100 eV Res: ~eV	Chemical Bonding
RIXS Resonant IXS	High Rate Somewhat Complicated	E_{in} >4 keV ΔE ~ 1-50 eV Res: ~0.1-1 eV	Electronic & Magnetic Structure
HR - RIXS High Res. Resonant IXS	To Be Developed Analyzer a problem	E_{in} ~ >4 keV ΔE ~ 0.1-10 eV Res: 1-20 meV?	Electronic & Magnetic Structure
NRIXS Non-Resonant IXS	Low Rate Simpler Interpretation	E_{in}~10 keV ΔE ~ <1-50 eV Res: 20-200 meV	Electronic Structure
IXS High-Resolution IXS	Larger Instrument Also: Nuclear Analyzer	E_{in}~16-26 keV ΔE ~ <1-100 meV Res: 1-3 meV, (<~0.1meV?)	Phonon Dispersion Liquid Excitations
NIS Nuclear IXS	Atom Specific Via Mossbauer Nuclei	E_{in} ~ 14-25 keV ΔE ~ 1-100 meV Resolution: .~0.5 meV	Element Specific Phonon Density of States (DOS)

Note: ΔE = Typical Energy Transfer (Not Resolution)

Relevance to the XFEL-O

XFEL-O Characteristics

Hard X-Ray: 5 – 25 keV (higher?) (~6% scan range)

Extreme flux in a small bandwidth: ~5x10^{14}/s in 5 meV

3 to 4 orders of magnitude more photons/s/meV than 3rd gen

1 to 10 times 3rd gen flux into Si (111) but into a few meV

1-2 MHz Pulse rate, pulse width ~0.5 ps (pump probe)

Mostly Coherent: small focal spot sizes possible (?)

"3 to 4 orders of magnitude more photons/s/meV than 3rd gen"

-> Incredible potential for experiments

Flux/count-rate multiplier
New geometries (standing waves?)

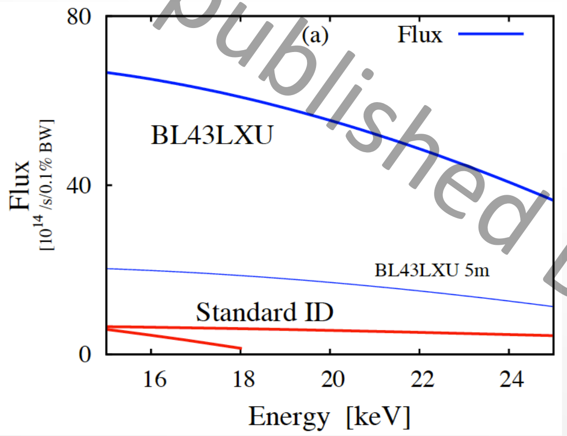
"1 to 10 times 3rd gen flux into Si (111) but into a few meV"

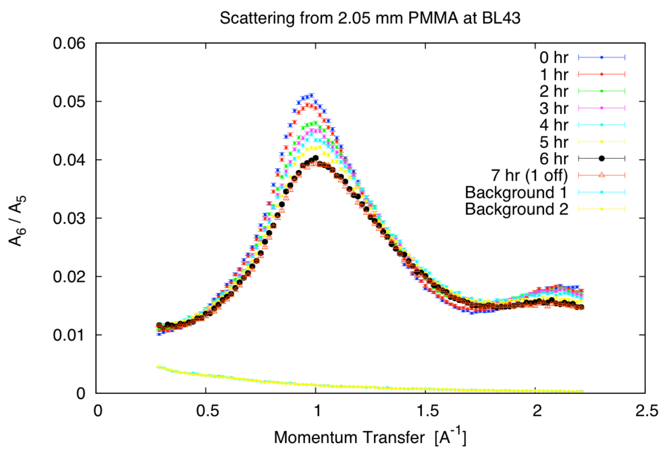
-> Many (most) samples will not survive very long, esp. w/focusing

Optics/Detectors downstream of the sample *must be efficient*

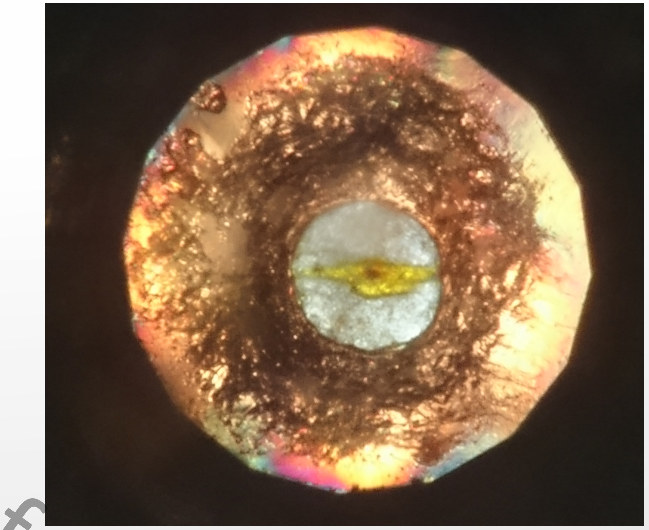
and/or step through the sample
and/or choose the right sample

Radiation damage with meV beams

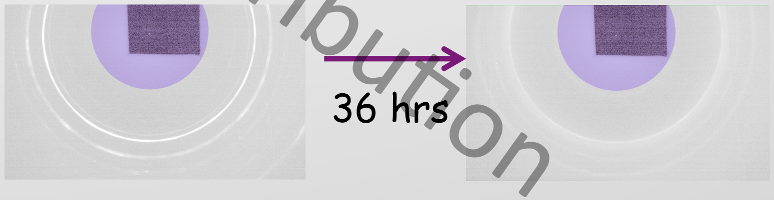


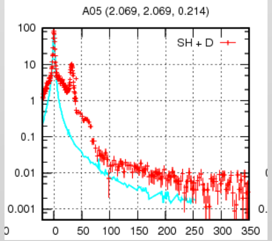


PMMA Elastic Standard
~20 GHz/0.8 meV @ 21.7 keV
Into 0.04 x 0.05 mm^2 spot
Notable damage after an hour



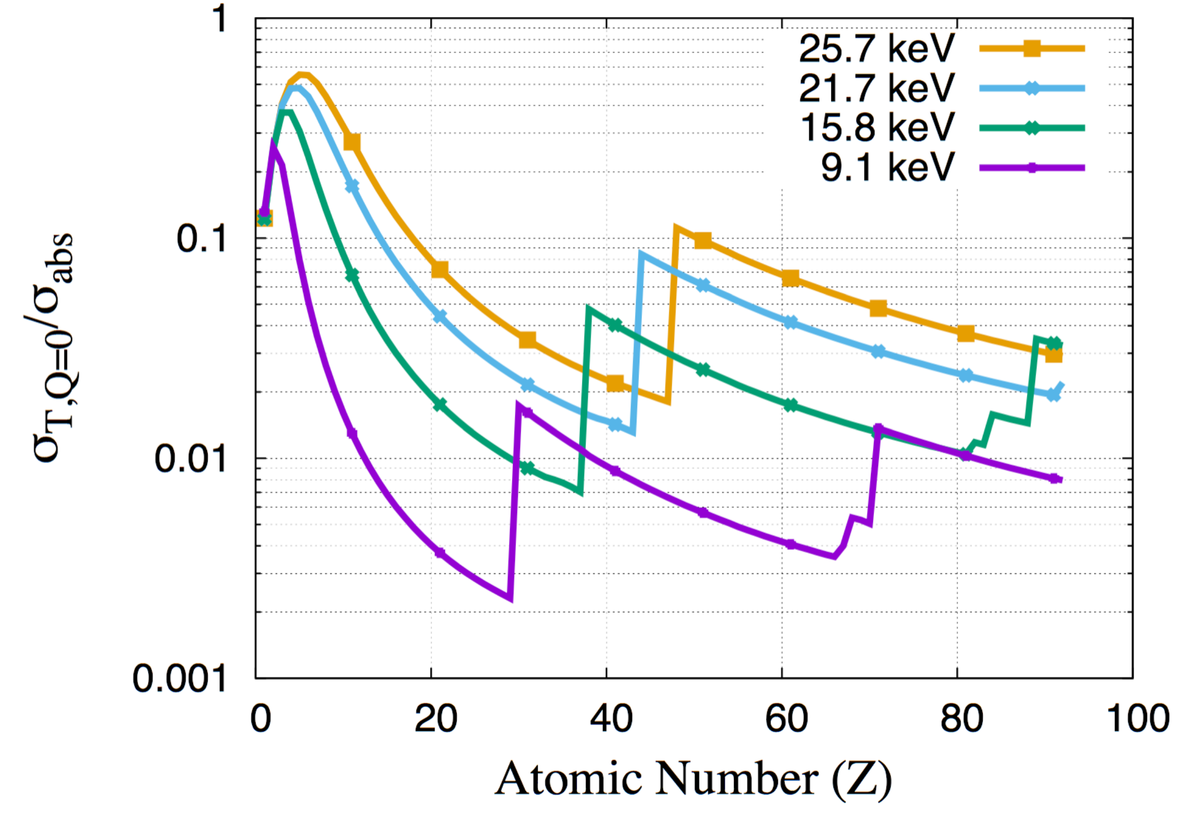
H_2S in DAC (Test at 10 GPa)
~40 GHz / 1.8 meV @ 17.8 keV
0.016 x0.017 mm^2 (with weak tail)





Absolute Energy Matters

(generally higher is better)



Plot shows relative signal for a thicker (10, 100 um) sample.

Radiation damage scales similarly (x factor of energy – so slower)

Higher energy also better for penetration into sample environments, & Q range

Higher energy worse for Q Resolution

Formal Dynamic Structure Factor

Measured Intensity is proportional to
the Dynamic Structure Factor S(**Q**,ω)

$$I_{scattered} \propto \frac{d^2\sigma}{d\Omega d\omega} = r_e^{\,2}\left(e_2^{\,*}\bullet e_1\right)^2 \frac{\omega_2}{\omega_1} S(\mathbf{Q},\omega_1,\omega_2)$$

$$S(\mathbf{Q},\omega) = \tfrac{1}{2\pi\hbar}\int dt\, e^{-i\omega t}\int d\mathbf{r}\int d\mathbf{r}'\;\; e^{i\mathbf{Q}\bullet(\mathbf{r}-\mathbf{r}')}\left\langle \rho(\mathbf{r}',t)\rho(\mathbf{r},t=0)\right\rangle$$

Dynamical Density Correlation
(FT of the Dynamical Pair Dist.)

$$= \; N\sum_{\mathbf{q}}\;\sum_{Modes}\left|\sum_{\substack{d\\ Atoms\\ /Cell}} \frac{f_d(\mathbf{Q})}{\sqrt{2M_d}}\; e^{-W_d(\mathbf{Q})}\mathbf{Q}\bullet\mathbf{e}_{\mathbf{q}jd}\; e^{i\mathbf{Q}\bullet\mathbf{x}_d}\right|^2 \delta_{(\mathbf{Q}-\mathbf{q}),\tau}F_{\mathbf{q}j}(\omega)$$

Phonons

$$= \sum_{\lambda,\lambda'} p_\lambda \left|\langle\lambda'|\sum_{\substack{electrons\\ j}} e^{i\mathbf{Q}\bullet\mathbf{r}_j}|\lambda\rangle\right|^2 \delta\left(E_{\lambda}'-E_\lambda-\hbar\omega\right)$$

Coupling to excitations

$$= \frac{1}{\pi}\frac{1}{1-e^{-\hbar\omega/k_BT}}\mathrm{Im}\left\{-\chi(\mathbf{Q},\omega)\right\} \quad = \quad \frac{1}{\pi}\frac{1}{1-e^{-\hbar\omega/k_BT}}\frac{1}{v(\mathbf{Q})}\mathrm{Im}\left\{-\varepsilon^{-1}(\mathbf{Q},\omega)\right\}$$

Generalized
Dielectric Response

Phonon Spectroscopy

$S(\mathbf{Q},\omega)$ for $\mathbf{Q}>0$

Normal modes of vibration (3N modes/Q in periodic materials)
For simpler materials & specific examples of complexity:
  Well explored experimentally (60's on)
  Calculations have now largely caught up to that work

Frontiers in Phonon Physics

- Coupling to meso/macro scale order & disorder. — Ferroelectrics…
- Phonon-Phonon scattering / anharmonicity — Many, Relaxors
- Coupling to magnetism & orbital order — Multiferroics etc
- Low dimensional systems
- Precision investigation for epc in complex materials — Many Superconductors etc

Frontiers for BOTH experiment and calculation

IXS Offers: Clean data, Small samples, Good Q Resolution

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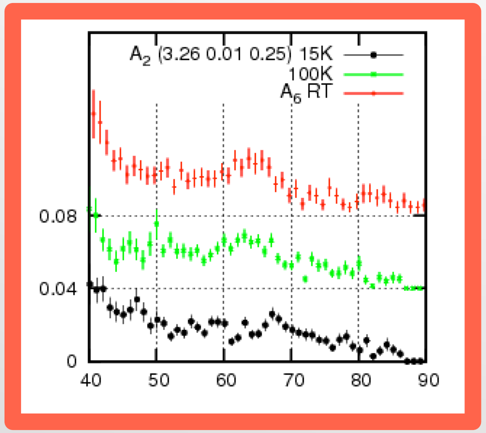
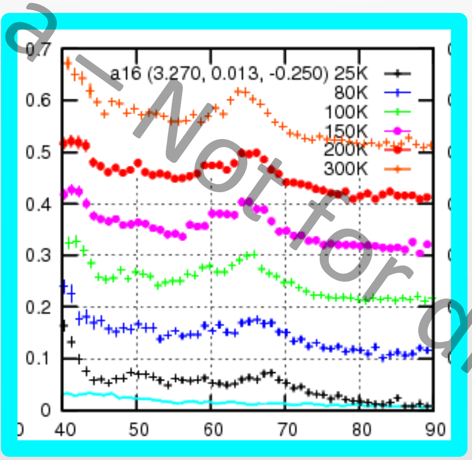
Precision Phonons in Complex Materials

There remain a number of cases (large unit cell, mixed heavy/light atoms) where phonon measurements can benefit from improved flux/Q resolution



2007, BL35XU
40+ hours/trace



BL43LXU (1.5 IDs)
~16-24 hours/trace

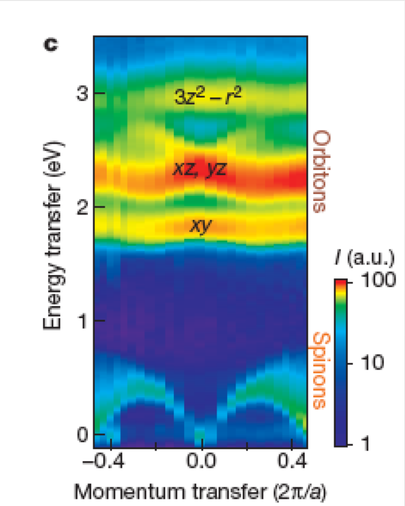
Immediate Question on Presentation
Is there fine structure?

NRIXS

Non-Resonant Inelastic X-Ray Scattering

Typically ~100 meV resolution, targeting low energy (eV-scale)
electronic excitations: d-d, orbitons, crystal field?, mixed?
subtle band-structure effects, gaps (plasmons?)
Large Q acceptance to get intensity (50x50 vs 5x5 mrad^2)

Improve resolution to allow better spectra, closer to the elastic line,
and to avoid phonons (note 2-phonon will limit smallest cross section)

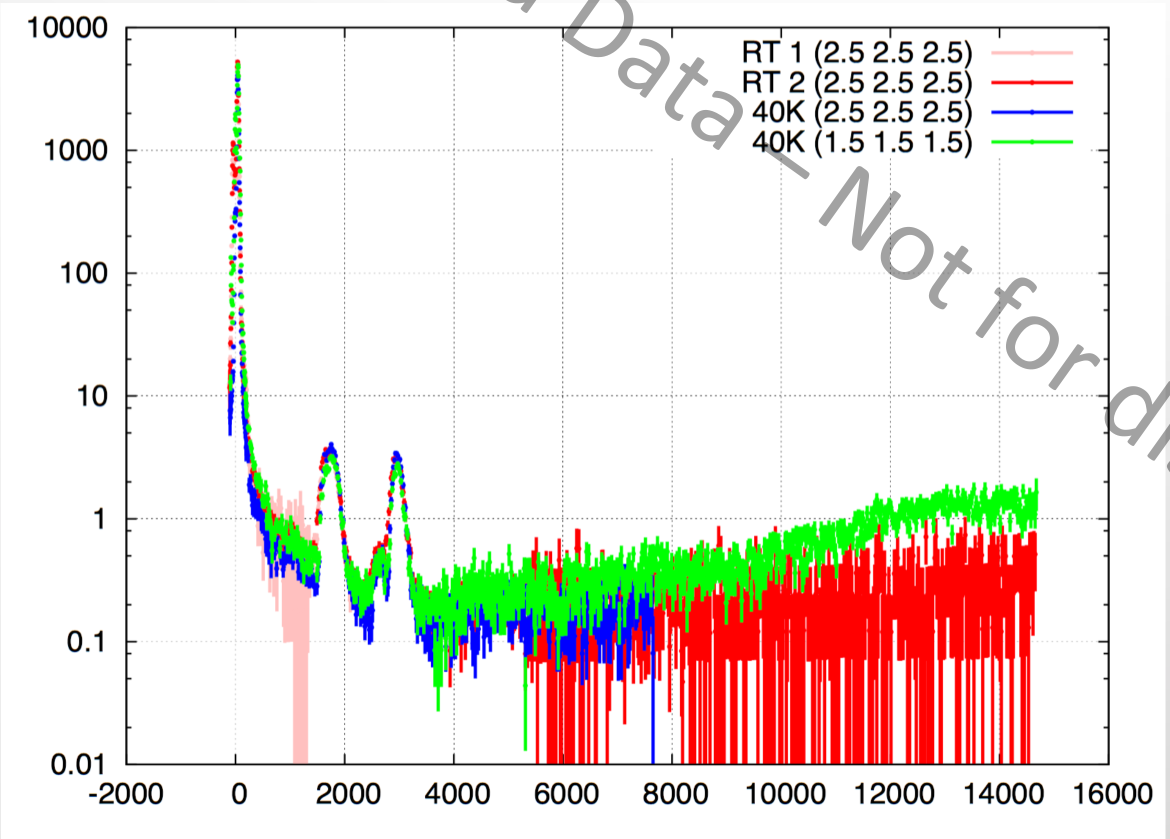


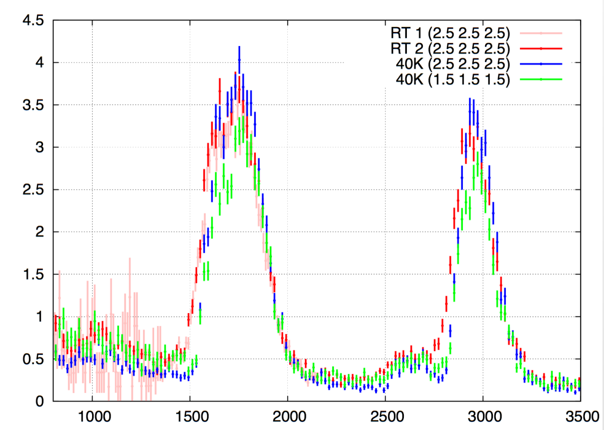
SIXS
Complex orbital excitation
Combining spin/hole motion
Schlappa, et al, Nat 2012

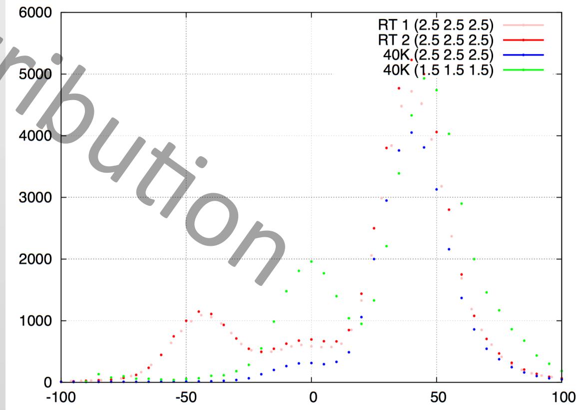
NiO As An Example

(Fairly strong signal)

27 meV resolution, Ishikawa, Baron et al, April 2016







Momentum Transfers

Large Q (>20 nm^{-1}):
- Detailed atomic structure
- Beyond Dipole Approximation (NRIXS)

Low to mid Q (<1 to >10 nm^{-1}):
- Macro/micro crossover (esp. disordered materials)

Low Q limit (as low as reasonable):
- Used to approximate inaccessible macroscopic quantities
  - Sound velocity, viscosity

Q is MOSTLY exactly what you want for periodic systems

Avoiding Q

Specific Potential Exceptions where “Q” is maybe less useful:

1. Extreme anharmonic effects / phonon localization (Solitons)
2. Scattering near defects (e.g. dilute resonant impurities)
3. Disordered Materials

Basically: when phonons are not a good basis set

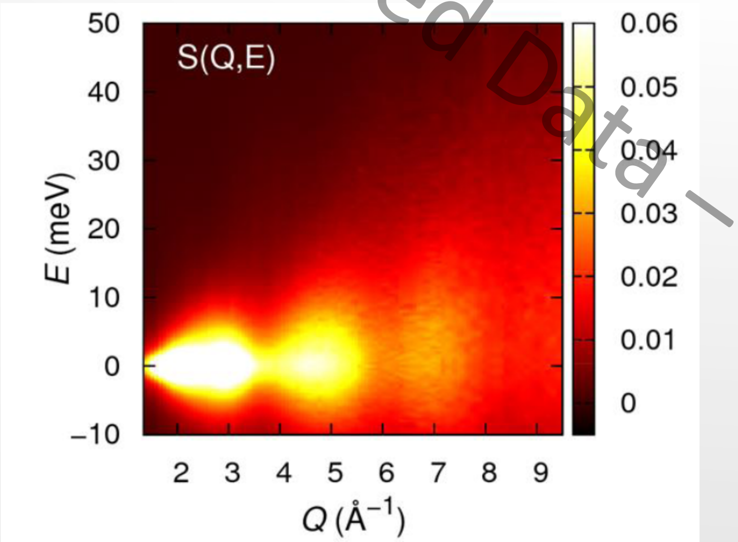
Option:

Fourier transform S(Q,w) into (r,t) space (or (r,w) space)

e.g. van Hove Correlation G(r,t) (or, similar, impulse response)

Fourier Transforming Water

Iwashita et al., in preparation.



G(r,t) removed

Nominal information content is the same but it can change ones viewpoint and allow one to see new things

Requires measurements over large Q/E range (larger is better).

Access to Transverse Dynamics

(atomic dynamics of disordered materials)

Fundamental limitation: S(**Q**,w) directly measures only longitudinal dynamics

$$S(\mathbf{Q},\omega) = \frac{1}{2\pi\hbar}\int dt\, e^{-i\omega t} \int d\mathbf{r} \int d\mathbf{r}' \;\; e^{i\mathbf{Q}\bullet(\mathbf{r}-\mathbf{r}')} \left\langle \rho(\mathbf{r}',t)\rho(\mathbf{r},t=0)\right\rangle$$

(For crystals can get transverse dynamics near a Bragg point)

This is a serious limitation for basic physics:

Liquids and glasses: have significant contribution of transverse dynamics

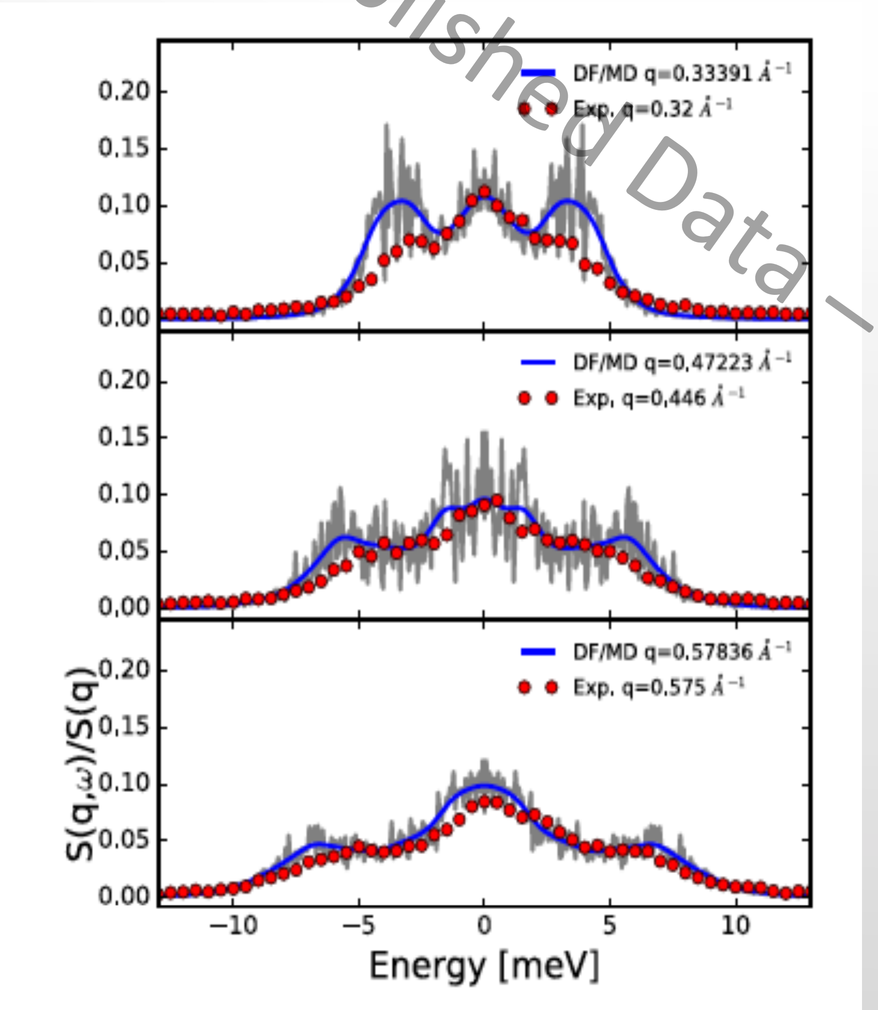
Boson peak = (probably) transverse dynamics

Can see (weak) signatures of transverse dynamics in S(Q,w) but they tend to be blurred and hard to isolate. Some way to isolate these experimentally would probably allow large advance in understanding disordered (technologically relevant??) materials.

Thoughts:
HR-RIXS?
IXS with Correlation?
Confined fluids

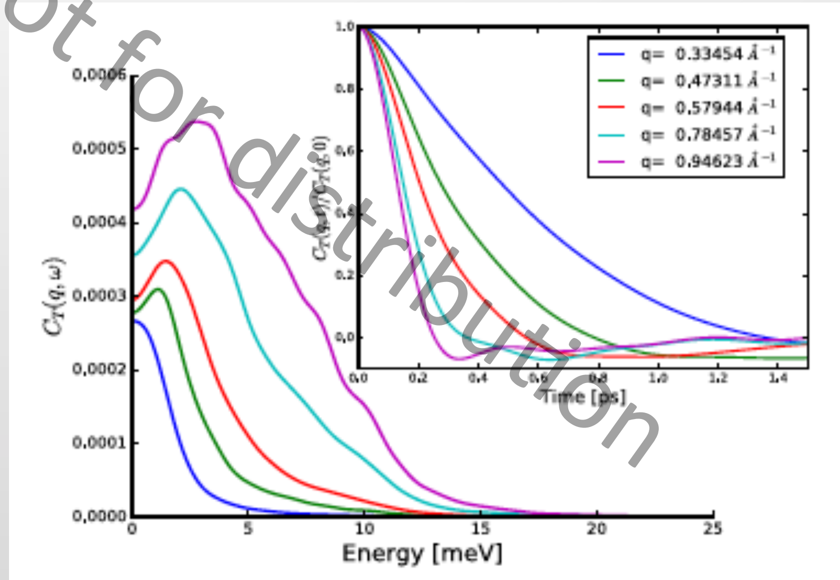
Just One Example:

Transverse Dynamics in Liquid Bi



Data: Inui et al, PRB 2015

Calcs: Ropo, Akolo, Jones (submitted)

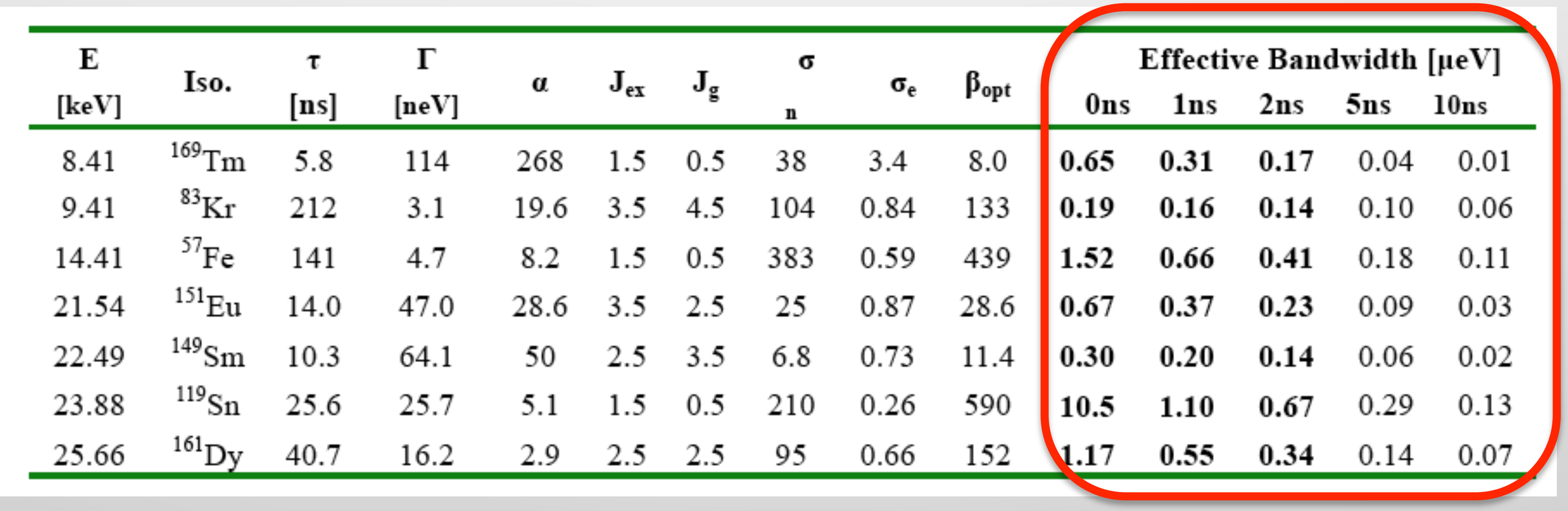


Ultra-High Resolution IXS

Potentially interesting for quasi-elastic scattering (viscosity) and phonon linewidths

Monochromators have been shown, with work to go to ~0.1 meV and one hopes for smaller. Analyzers an issue.
(Can consider a post-sample-collimation setup – Shvyd'ko)

Consider nuclear analyzer…. (Chumakov, et al, PRL 1996)

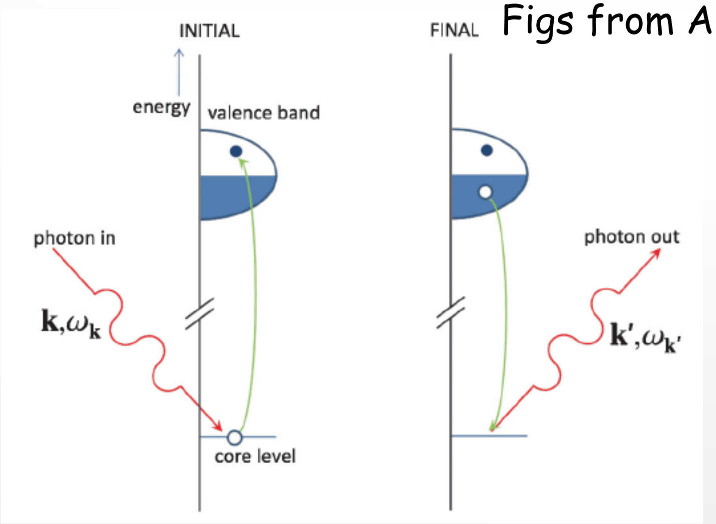


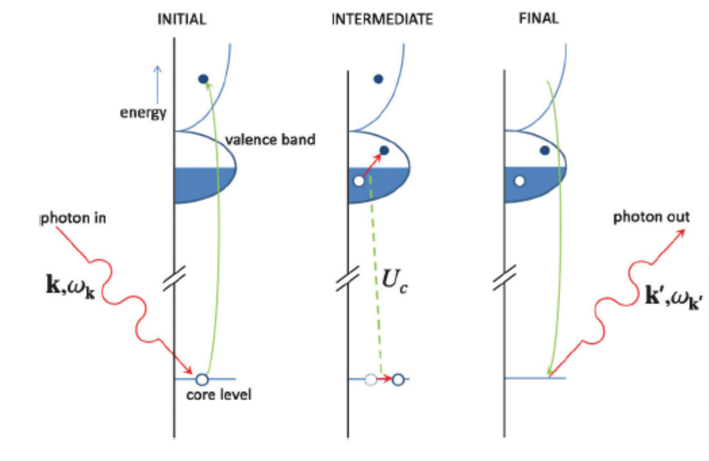
E [keV]	Iso.	τ [ns]	Γ [neV]	α	J_{ex}	J_g	σ_n	σ_e	β_{opt}	Effective Bandwidth [μeV] 0ns	1ns	2ns	5ns	10ns
8.41	^{169}Tm	5.8	114	268	1.5	0.5	38	3.4	8.0	**0.65**	**0.31**	**0.17**	0.04	0.01
9.41	^{83}Kr	212	3.1	19.6	3.5	4.5	104	0.84	133	**0.19**	**0.16**	**0.14**	0.10	0.06
14.41	^{57}Fe	141	4.7	8.2	1.5	0.5	383	0.59	439	**1.52**	**0.66**	**0.41**	0.18	0.11
21.54	^{151}Eu	14.0	47.0	28.6	3.5	2.5	25	0.87	28.6	**0.67**	**0.37**	**0.23**	0.09	0.03
22.49	^{149}Sm	10.3	64.1	50	2.5	3.5	6.8	0.73	11.4	**0.30**	**0.20**	**0.14**	0.06	0.02
23.88	^{119}Sn	25.6	25.7	5.1	1.5	0.5	210	0.26	590	**10.5**	**1.10**	**0.67**	0.29	0.13
25.66	^{161}Dy	40.7	16.2	2.9	2.5	2.5	95	0.66	152	**1.17**	**0.55**	**0.34**	0.14	0.07

Lossy on 0.1 meV scale, Better on ~10 ueV scale…

RIXS

Figs from Ament et al RMP 2011





Direct: L-edge gives d-electrons

5 keV: Lathanum L

Indirect: K-edge for d-electrons

Vanadium K

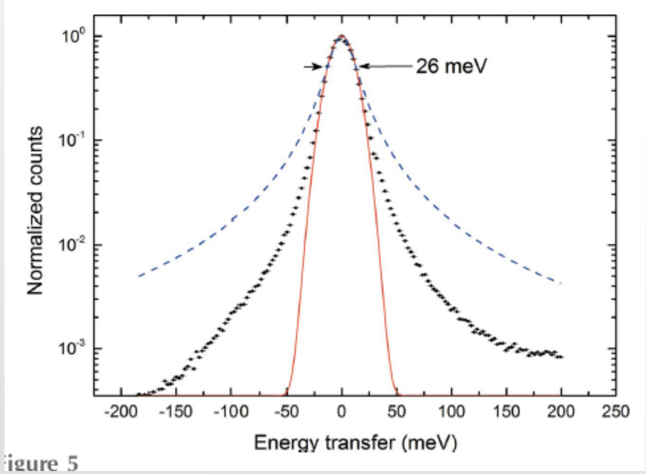
For d-electron physics : L-edge much preferred.
Spin, orbital, charge order all accessible.

Relevance: magnons, orbitons, mixed excitations -> calcs needed but getting better

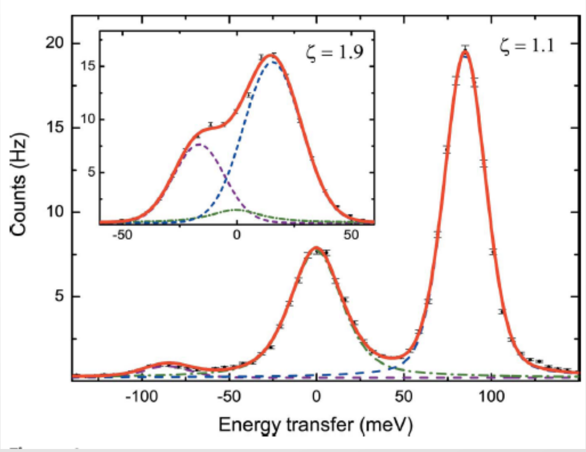
Just for resonant enhancement, either OK (K edge may be larger)

High Resolution RIXS

In present geometries, basically analyzer limited in hard x-ray region as need to match analyzer lattice constant to resonance energy. Materials include quartz and sapphire (analyzers: Yavas et al, jsr2015, nim 2007).



Quartz anal, 2m radius
(100mm diam)



Expect improvement to ~5 meV is straightforward
To meV, or sub-meV, is not so clear, but probably…

Interest:
- coupling to magnetism, density waves,
- possibly alternative view of epc (Devereaux and others)
- resonance specificity

Some Specific Issues/Goals

for this workshop

Goal (?):

Come up with high-profile experiments addressing (broadly? societally?) relevant science that will help get the machine funded.

Targeted studies probably required due to beam time and instrumentation (manpower and money) limits

Especially true for IXS:
multiple big spectrometers probably not an (immediate) option…

Note: pump-probe not addressed here…

Optics Questions & Challenges

(AB Opinions)

Method for meV-resolution IXS:

Spherical Analyzers? Post-Sample Collimation (YS&+spectrograph?).

sample clear aperture (for sample environment -> larger is better)
required beam size (larger is easier, maybe better for rad damage)
allowable beam projection/ sample thickness at finite Q
resolution function -> FWHM? (PSC tail can be better)
parallelization (Q, E-spectrograph (ys))
absolute energy: higher is better

What about low-resolution expts:

NRIXS: Desire 5-10 meV res with sharp tail

Emission? X-Ray Raman?

Sufficiently flexible spectrometer design(s) without sacrificing capability

Development of high resolution RIXS analyzers

5 meV probably OK (work required), 1 meV maybe.

Development of ultra-high resolution non-resonant spectrometer

Mono? PSC optics? Nuclear Analyzer?

Expect incident beam optics to be LN2 cooled
Never forget the fight against radiation damage…

Experimental Suggestions

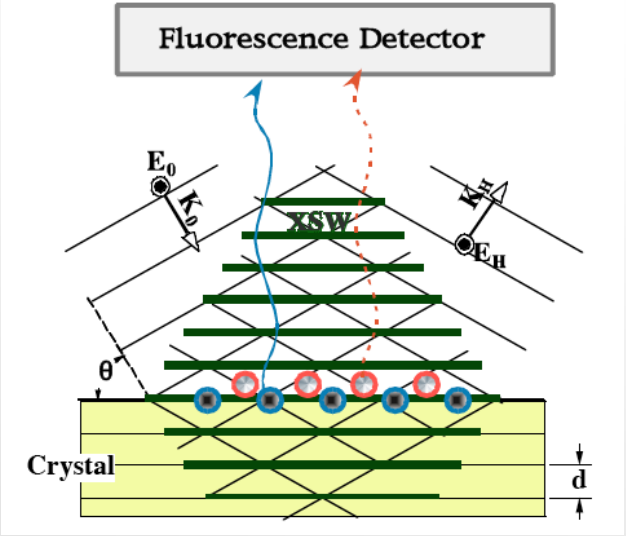
Build on present research programs… handle with care!

1. Using standing waves to investigated phonons
2. Using a meV spectrometer to probe sub-ueV linewidths
3. Reminder of High Pressure challenges and dedication

Coherent 2-Beam Excitation

(Standing Waves)

From Bedzyk, Enc. Of Physcs



Include Bragg Excitation (or specular).
Look at surface (deposited layer) or at bulk.

Change angle – change standing wave maxima

(Formal phonon polarization viewpoint:
Kohl, pss (1985))

$$\frac{d^2 I}{d\Omega\, dE} = \frac{I_0}{2}\frac{k_f}{k}\frac{N}{2s}\sum_{\boldsymbol{q}\sigma} |f_\sigma(\boldsymbol{K}) + e^{-i\chi} f_\sigma(\boldsymbol{K}')|^2\, G(\boldsymbol{q},\sigma) = \\ = I_0 \frac{k_f}{k}\frac{N}{4s}\sum_{\boldsymbol{q}\sigma} \{|f_\sigma(\boldsymbol{K})|^2 + |f_\sigma(\boldsymbol{K}')|^2 + 2\,\mathrm{Re}\,[e^{i\chi} f_\sigma(\boldsymbol{K})\, f_\sigma^*(\boldsymbol{K}')]\}\, G(\boldsymbol{q},\sigma)\,, \qquad (7)$$

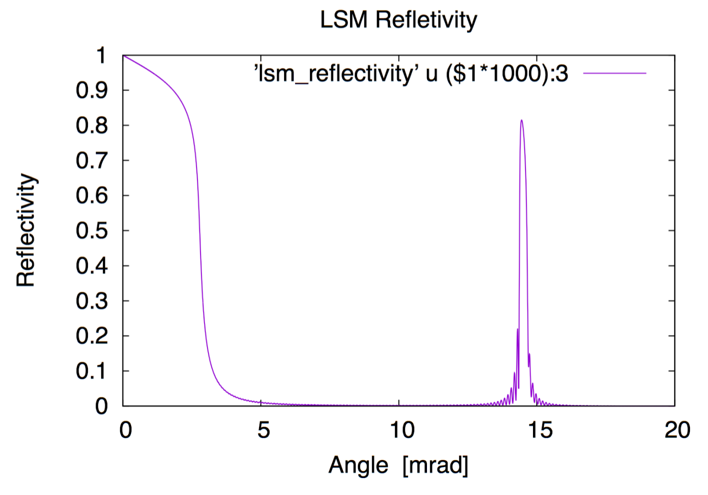
Driving Science: Interface/surface behavior can lead to remarkable properties
Single Layer in Specular GIXS, Multilayers structure

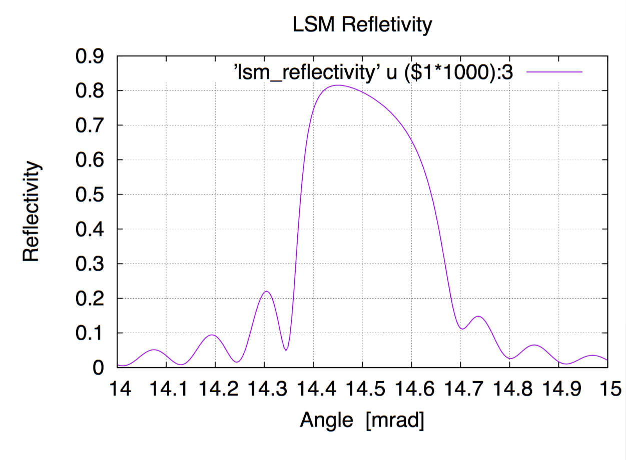
e.g. insulators that superconduct ($LaAlO_2$/STO) Reyren, Science (2007)
High-Tc (100K!) of FeSe on STO – Ge Nat Mtl. (2015)
Understanding how water surfs on graphene

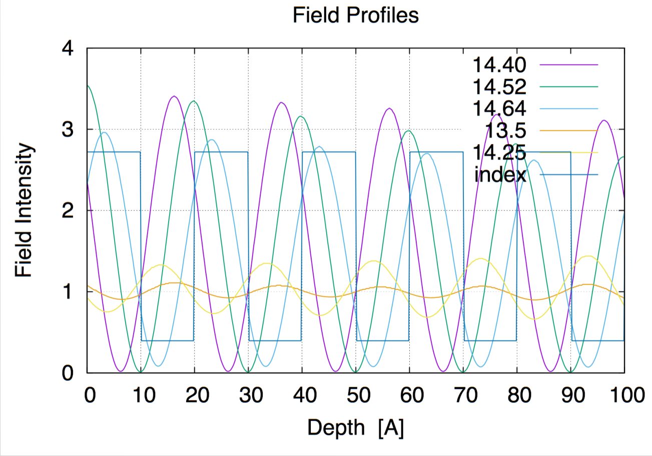
Need High (near unity) reflectivity
Brilliance Hungry Expt (small beam & well collimated)

Multilayer

Less Demanding Example – Now In Progress

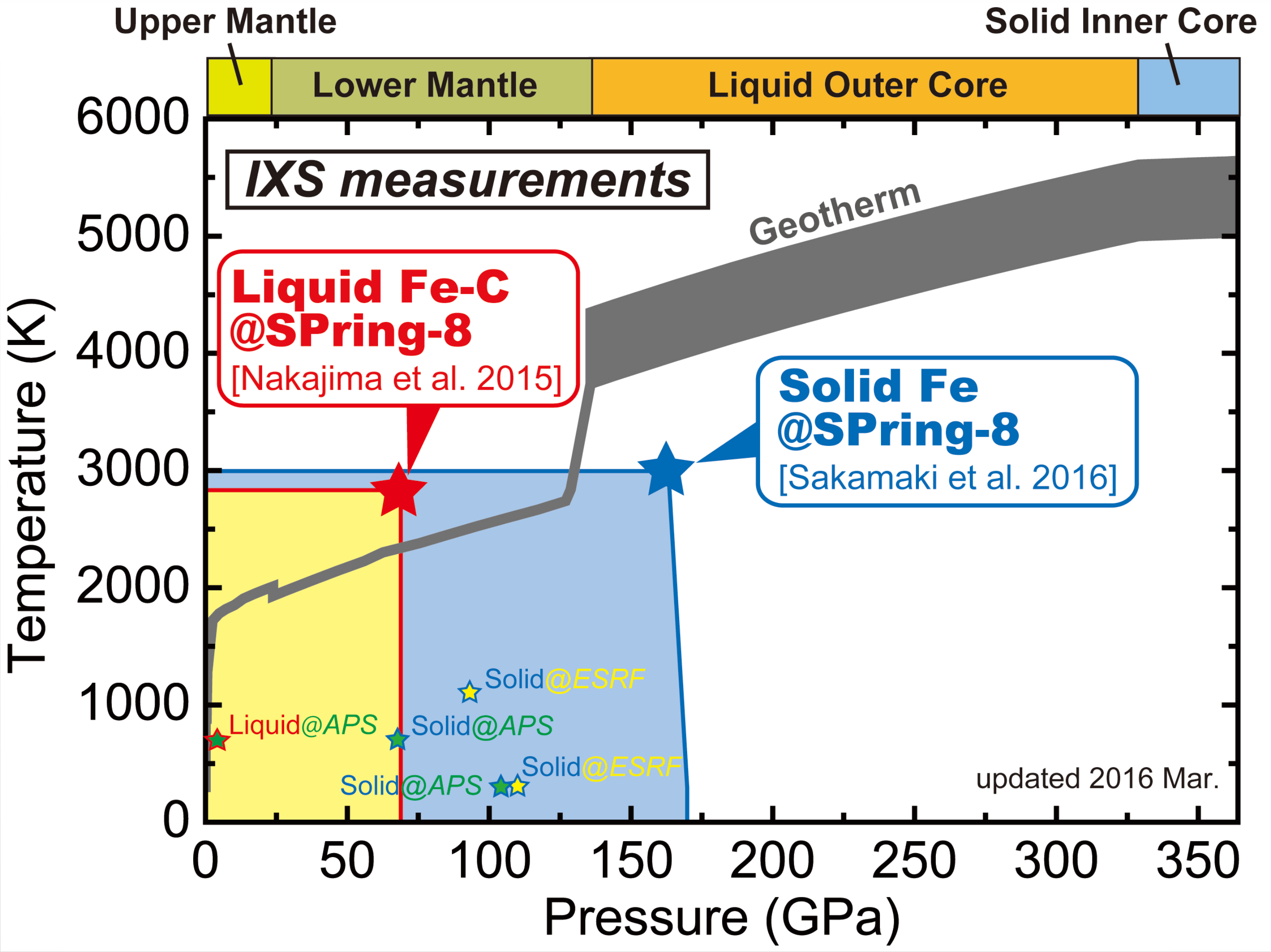






Select Position on RC
Allow focus onto phonons
in Si or in W

(More) Extreme Conditions



Faster Experiments help a lot when in marginally stable conditions

Sound Velocity. Viscosity.

Using a meV Spectrometer to probe (sub) neV linewidths

Detailed Balance

Fundamental Symmetry of a Sample in Thermal Equilibrium

$$\frac{S(\mathbf{Q},\ \omega)}{S(\mathbf{Q},-\omega)} = e^{\hbar\omega/k_B T}$$

Strictly:

$$\frac{S(\mathbf{Q},\ \omega)}{S(-\mathbf{Q},-\omega)} = e^{\hbar\omega/k_B T}$$

But what if you violate the conditions of thermal equilibrium?
i.e.: if you induce a thermal gradient

Boltzmann Transport Theory: $n_s - \overline{n_s} = -\tau_s \frac{\partial \overline{n_s}}{\partial T}\ v\nabla T$

Small (5%) effect, but direct probe of phonon lifetime for longer lifetimes