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Recent Progress in Non-Resonant Inelastic X-ray Scattering

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Materials Dynamics Group
RIKEN SPring-8 Center

Presented at the 11th International Conference on
Inelastic X-Ray Scattering (IXS 2019)
26 June 2019

Outline

Main Target: What has been happening the last few years (in-house)

Transformation from
making the BL & Spectrometers run -> specific experimental goals

Introduction: Orientation & Context

Progress in High Pressure DAC Work:
5 um Beam, Soller Screen for Background Reduction
Applications in a talk by E. Ohtani

Progress with Liquids:
Practical sub-meV setup for sub nm^{-1}
Crossover from hydrodynamic to fast sound in liquid iron

Progress using thermal currents to manipulate phonons
How to break detailed balance in an interesting way
Investigate microsecond-scale phonon lifetimes in silicon

Collaborators

High Pressure: H. Fukui, Y. Nakajima, D. Ishikawa, et al.
Ohtani-Lab (Ikuta), Hirose-Lab (ELSI)

Liquids: D. Ishikawa (T-Gradient), M. Inui (High T), *et al*

Thermal line-width: T. Fukuda & D. Ishikawa

D. Ishikawa
MDG & JASRI

H. Fukui
MDG & U. Hyogo

Y. Nakajima
MDG & Kumamoto U.

T. Fukuda
MDG & JAEA

E. Ohtani
Tohoku U.

K. Hirose
ELSI & Todai

M. Inui
Hiroshima U.

meV IXS: Orientation/Background

meV-resolved non-resonant IXS to investigate atomic dynamics.
Phonons in Crystals, excitations in liquids, etc.
Measure the Dynamic Structure Factor, $S(\mathbf{Q},\omega)$

Vienna Summer School: Dorner, Fujii, Hastings, Moncton, Siddons et al (1980)
(Priv. Comm. & Notes from D. Moncton)

BL Proposal: Dorner & Peisl (1983)

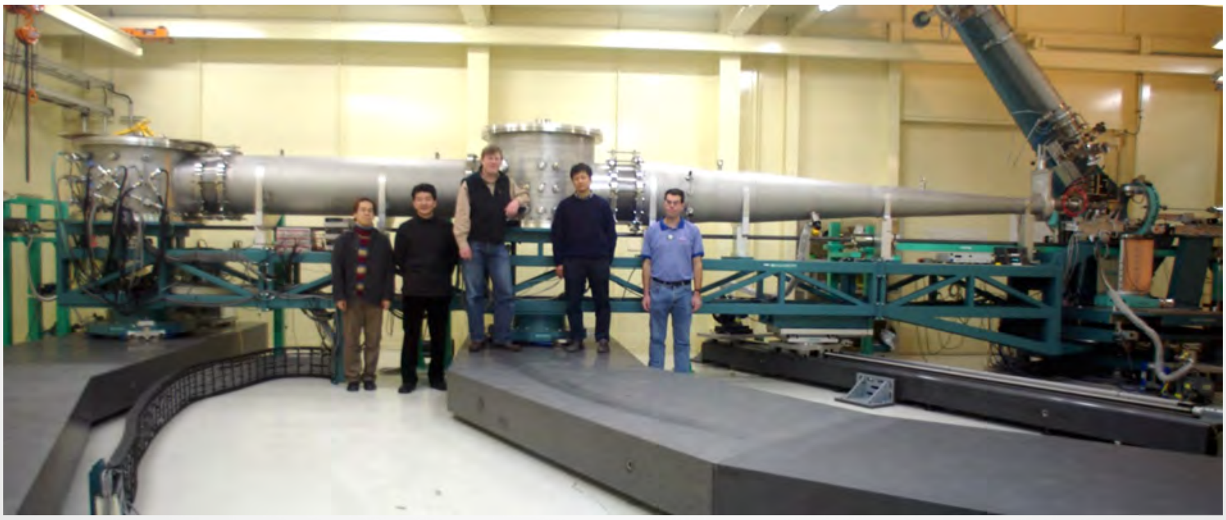
First Phonons (Be) at DESY: Burkel, Peisl, Dorner (1987)

3rd Generation beamline (ID16) at ERSF : Sette, Ruocco, Krisch *et al* (1995)

Presently
- 1 Beamline (ID28) at ESRF (Shutdown for Upgrade)
- 2 Beamlines (Sector 3 and Sector 30) at APS
- 2 Beamlines (BL35 and BL43LXU) at SPring-8
- (1 Beamline at NSLS-II (Different setup, PSC))

Review: Springer Handbook & arXiv 1504.01098

The First meV Instrument at SPring-8
(BL35XU)



Operational from ~2002

Baron, Tanaka, *et al*, J. Phys. Chem. Solids (2000)

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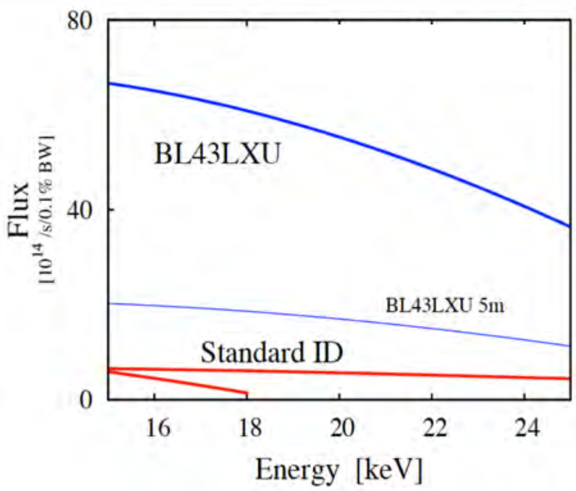
Higher Flux for IXS

-> Propose a new beamline (BL43LXU)
using a long (30m) straight section



In-Vacuum ID technology well matched to IXS (15-25keV) at 8 GeV
Short (19mm) period/small (6mm) gap ID: Fundamental
-> Win/Win: more flux & less heat load per unit flux

Note: After BL43 Discussions,
request (granted) to upgrade BL35 ID

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Support from the Scientific Community

BL43LXU Collaborators

RIKEN-JASRI Collaboration

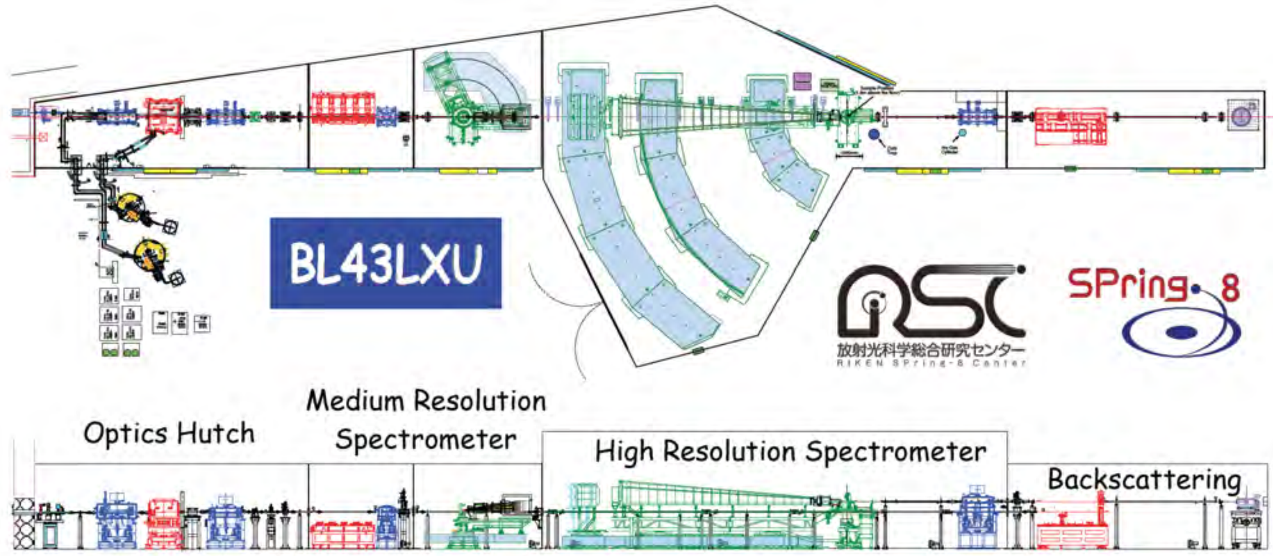
Initial Discussions & Design (Beginning in 2004):

Electron Optics: Kouchi SOUTOME, Hitoshi TANAKA
Insertion Devices: Takashi TANAKA, Hideo KITAMURA
Mono & Cooling: Tetsuro MOCHIZUKI
Front End: Sunao TAKAHASHI
Hutches and Shielding: Kunikazu TAKESHITA
Transport Channel & Optics: Haruhiko OHASHI, Shunji GOTO
Spectrometer (2008-): Daisuke ISHIKAWA

More Complete List of Contributors Includes:

M. Abe, H. Aoyagi, H. Arita, N. Azumi, D. Ellis, K. Fukami, H. Fukui, Y. Furukawa, S. Goto, Y. Harada, D. Ishikawa, Y. Ishizawa, H. Kimura, H. Kitamura, H. Konishi, T. Matsushita, Y. Matsumoto, T. Mochizuki, N. Murai, H. Ohashi, T. Ohata, H. Ohkuma, M. Oishi, M. Oura, S. Sasaki, J. Schimizu, Y. Senba, M. Shoji, K. Sorimachi, K. Soutome, S. Takahashi, M. Takata, K. Takeshita, T. Takeuchi, H. Tanaka, T. Tanaka, S. Tsutsui, H. Uchiyama, T. Wagai, J. Yahiro, M. Yamamoto, H. Yamazaki

Director/Facilitator: T. ISHIKAWA



Growing Pains

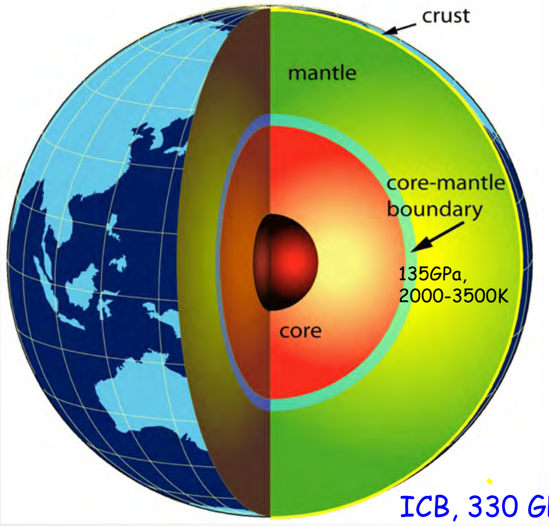
3 x 5m/6mm gap IDs, 2 Spectrometers (IXS/EE)

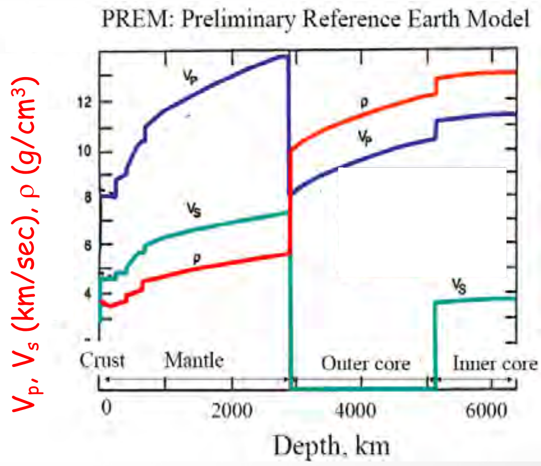
Full power operation from April of 2015

Steering the e- beam through 3 IDs (more careful feedback)
Heating/Melting of ID covers (intra-ID absorber -> Few% losses)
Beam spatial structure due to mirrors (improved polishing -> OK)
Technology Transfer for HR analyzers (New company taught)
Difficult fabrication of MR analyzers (New process developed)
Stability of M1 (AB Correct Toyama's design)

Note: Problems involving ring systems (e-beam, IDs) take ~year time scales to diagnose and fix. Others mostly faster.

IXS & Geoscience





ICB, 330 GPa,
>~5000K

Earth's Center, 365 GPa
>~5000K

Velocity (and ρ) well know from seismic measurements.
Needed: Lab measurements relating T, Density & Composition to P

A catalogue of sound velocities **in extreme conditions**...

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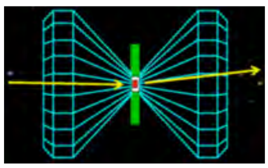
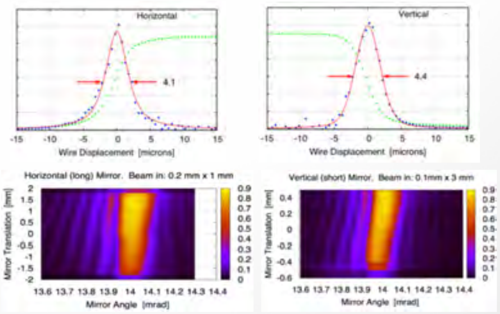
IXS with Diamond Anvil Cells (DACs)

High Pressure: Squeeze the sample between two diamonds



Diamonds: 2 x 1.5 mm Thick (typical)
Sample: ~ 20-200 um diam. x ~2-20 um thick
Scattering from C, gasket & pressure medium
Higher P -> smaller sample, harder expt.

Elliptical KB (Multilayer) for 17.8 keV
Accepts: 1 x 3mm^2
Focus Size: 5 x 5 um^2 (FWHM)
Throughput ~ 60%



Baron, Ishikawa, Fukui & Nakajima, Y. (2019).
AIP Conf. Proc. **2054**, 20002. DOI: 10.1063/1.5084562.

High T: (Double Sided) Laser Heating

Dedicated Systems
Tohoku/RIKEN
Tokyo Tech

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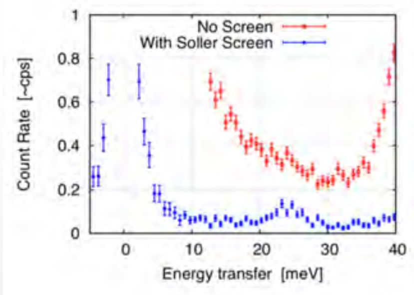
"Soller Screen"

HP DAC work limited by background from diamonds (or Be gasket)
ie: parasitic scattering from within a few mm of the sample

Usual Solution: A small slit near the sample to limit the view to only the sample
(One Detector)

For IXS: Would like a Soller slit but spacing is prohibitively small (~50 um spacing needed)

"Soller Screen": Two Flat W plates at 5 mm and 10mm from the sample with laser cut slots
Accepts beam to every other (every second) analyzer (24→12) and drastically reduces the background



Baron, Ishikawa, Fukui & Nakajima, Y. (2019).
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HP DAC Work

Program looking at Elasticity/C_{ij} in single crystals via Christofel's eqn
(Fukui, Yoneda & Co-Workers)

Program looking at liquids (HP/HT) including iron & related materials
(Nakajima, Kuwayama, Hirose, et al)

Program looking at powder samples (iron & others, mostly HP, some HT)
(Sakamaki, Fukui, Ikuta, Ohtani, et al)

"Using an x-ray scattering beamline
to change the composition of the Earth's core"
Talk by E. Ohtani (Thursday PM)

Program Looking at Hydrogen
H_3S (Fukui, Nakajima, et al)
Hydrogen Solid Vibron (Liu, Ding, Mao, et al)
Liquid Hydrogen Vibron (AB, DI, Liu, Mao, et al) MR & HR

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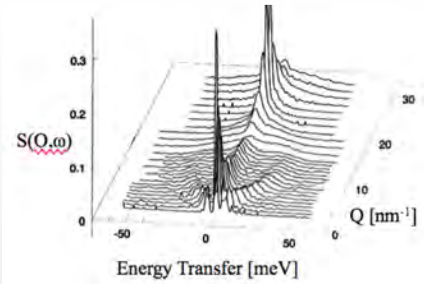
Liquids

Essentially impossible to separate structure from dynamics

Long-standing area of interest as IXS is, mostly, a unique probe at Q->0.
(IXS: E >>> Energy-Transfer, INS: Energy ≈ Energy-Transfer -> limits INS E range)

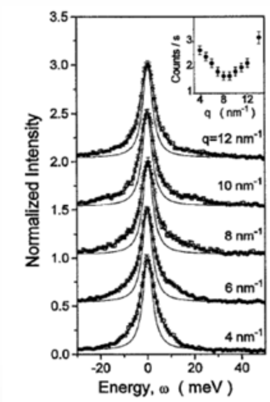


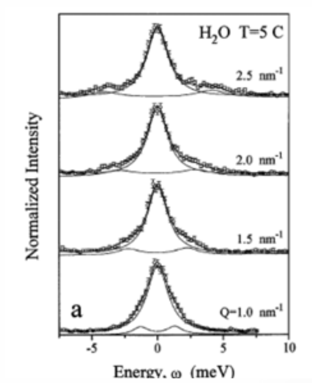
Liquid Mg, Kawakita, et al 2004

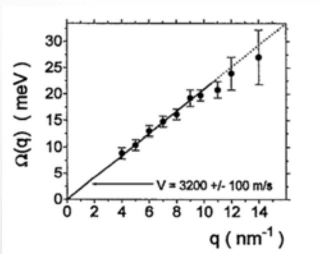
Points of Interest:
- Acoustic Dispersion/Fast Sound
- Acoustic line-width
- Quasi-elastic line-width
- Transverse dynamics
- Landau-Placzek?

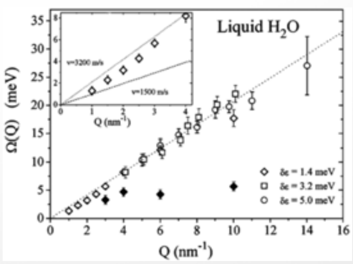
Usual Focus: Crossover from continuum to atomistic behavior
-> Lower Q and higher resolution is critical

Sound Crossover in Water







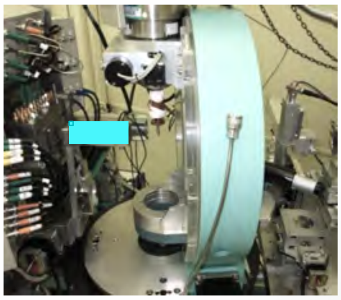


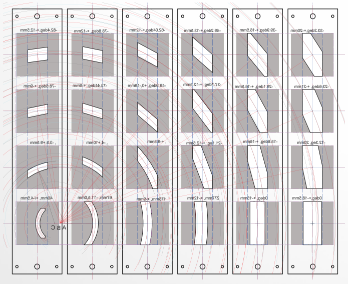
Sette et al, prl 1995

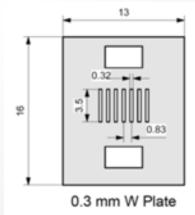
Sette et al, prl 1996

For Metals: Scopigno, Ruocco, Sette, RMP 2008

Analyzer Masks & Soller Slit







Slits at fixed |**Q**|
Improves Rates for fixed
Q Resolution dQ/Q~ 10%

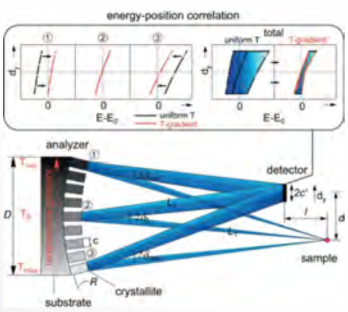
Limits acceptance to near the sample
Reduces background from
Windows & Sample Environment

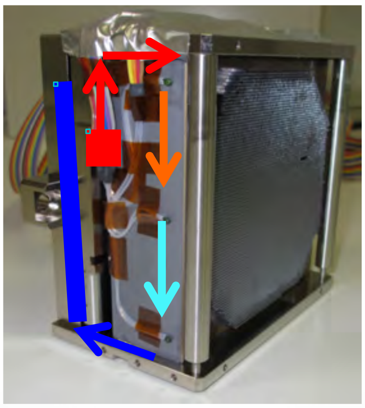
Baron, Ishikawa, Fukui & Nakajima, Y. (2019).
Conf. Proc. **2054**, 20002. DOI: 10.1063/1.5084562.

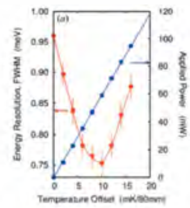
Temperature Gradient Analyzers

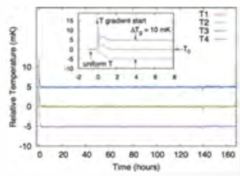
to Improve Resolution

BL43 Analyzers: 9.8 m Radius,
90x94 mm^2 **on a Rectangular Substrate**

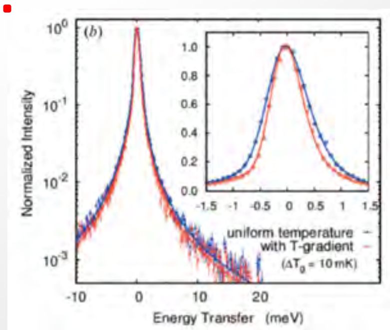








Control to ~0.0003 K



Usual operation is slightly
off of the ideal Roland Circle
-> Helps if you vary "d"
using a T-Gradient

Make the analyzer substrate
part of a thermal circuit

Si (13 13 13):
~0.95 -> 0.75 meV at 25.7 keV

Ishikawa & Baron, JSR 2010
Ishikawa, Ellis, Uchiyama & Baron, JSR 2015

Practical: Reduce BW onto BX to 0.2 eV (x3)

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Liquid Iron

T_m=1538 C

Geological Relevance!

More Fundamental Physics

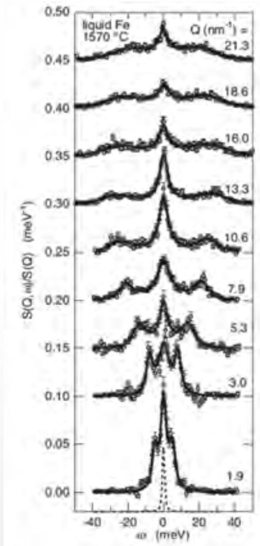
Liquid TMs have a relatively high specific heat ratio.

$$\gamma \equiv \frac{C_p}{C_v} \sim 1.8$$

Liquid Fe, Ni, Co...

Relation to Landau-Placzek Ratio (hydrodynamics):

$$\frac{I_R}{2I_B} = \frac{Quasieleastic}{Phonon} = \gamma - 1$$



Hosokawa , et al PRB 2008

Started as a straight-forward experiment...

...became a more complicated one

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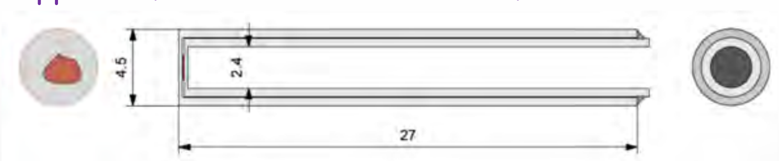
Liquids at High Temperature

Basically two options for ~day-long expts:

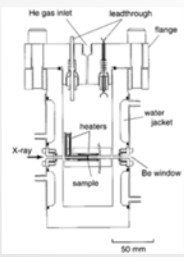
Levitation -> typically mm scale samples -> light materials only

Sample Cell -> requires sealed (single crystal) cell for high T

Extensive experience (Hiroshima U.) with cells milled out of single crystal sapphire (Tamura, Inui & Hosokawa, RSI 1999)



And in principle the right environment ("Marburg Chamber" – Hosokawa & Pilgrim, RSI2001)



But the chamber failed

Heaters (W or Mo) died too quickly at ~1600C

One run ~30 hours, but never again. (Weeks of work by MI)

Looked (AB) like a contamination / sealing issue

-> New chamber using carbon composite heater

8 days at ~1600C, Multiple cycles OK

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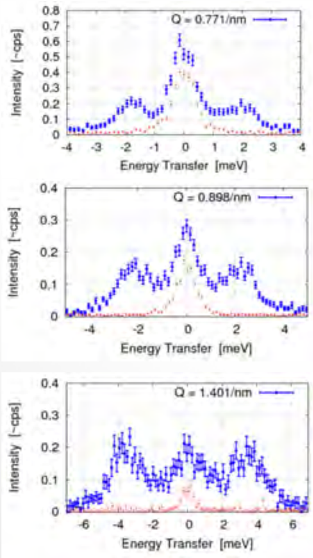
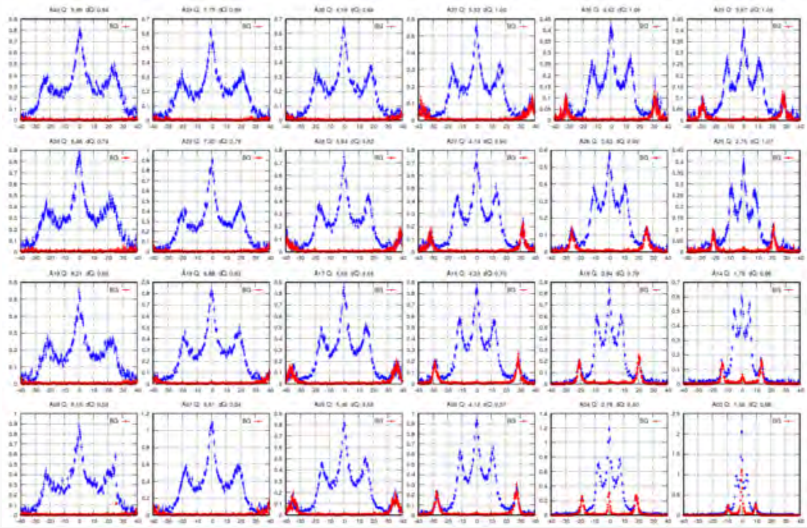
Liquid Iron IXS Data

T ~ 1560 C





Si (13 13 13), 0.8 meV at 25.702 keV
1 or 2 analyzers only (time limited)

Si (11 11 11), 1.3 meV at 21.747 keV
24 Analyzer Array

Blue = IXS Data (no bkgd subtraction)
Red = Background (scaled by transmission)

Baron, Inui, Ishikawa, Kajihara, Nakajima, Matsuda, et al., In progress

Iron Fit Results (dho)

Unpublished results removed

Detailed Balance

Symmetry property of scattering from a sample in thermal equilibrium

$$S(\mathbf{Q},-\omega) = e^{-\hbar\omega/k_BT}\; S(\mathbf{Q},\omega)$$

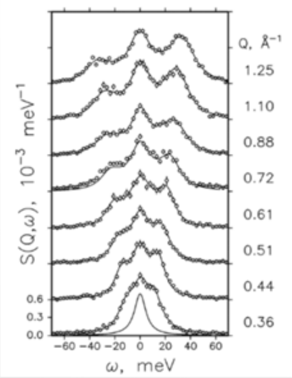
Slightly Simplified

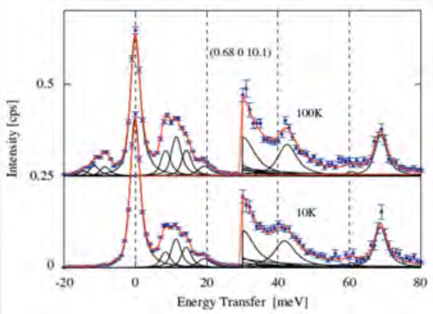
The anti-stokes (E,ω<0) side is a lower-intensity copy of the stokes (E,ω>0) side with the intensity scale factor set by the energy transfer and the temperature

Really just a statement about the population of bosons in thermal equilibrium…

Used: built in to most IXS data fitting

Liquid Li, 215C
Sinn et al, 1997





YBCO
Baron et al, 2008

See, e.g., Squires, 1978, Lovesey, 1984

Thermal Conductivity

Insulators

A Boltzmann transport equation relates the local temperature gradient to the deviation of phonon populations from their equilibrium value

∇T related to $n_s - \bar{n}_s$

Generally complex (all phonon scattering mechanisms need to be included, etc…)

BUT: often simplified by a "life-time approximation" LTA by assuming each phonon mode has a lifetime and if a thermal gradient is removed it will exponentially relax to its equilibrium population with that lifetime…

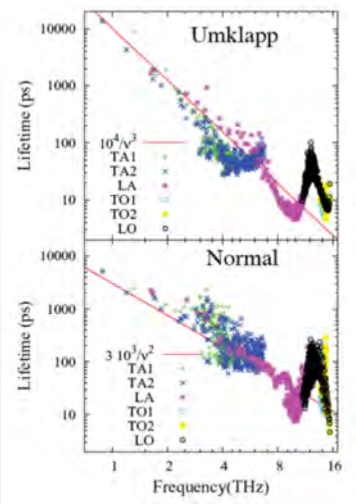
$$n_s - \bar{n}_s = -\tau_s \frac{\partial \bar{n}_s}{\partial T} \mathbf{v}_s \bullet \nabla T$$

$$\mathbf{v}_s = \text{phonon group velocity}$$

Breusch, 1987

Phonon Thermal Lifetimes

Anharmonic lifetimes are important for understanding thermal conductivity but it is superficially absurd to use a meV spectrometer to probe thermal lifetimes (excepting extreme cases)



Esfarjani et al PRB 2011
277K

Thermal Lifetime (τ): ns – μs
Line-width (Γ): μeV – neV

$$\Gamma\tau = \hbar = 0.66 \ \text{meV-ps}$$

Work at the intersection of:

Detailed Balance
&
Thermal Conductivity

One-Phonon Cross Section

Harmonic Approximation

$$S_s(\mathbf{Q},\omega) = |F_s(\mathbf{Q})|^2 \ \delta_{\mathbf{Q}-\mathbf{q},\tau} \left[n_s\,\delta(\omega+\omega_s) + (n_s+1)\,\delta(\omega-\omega_s) \right]$$

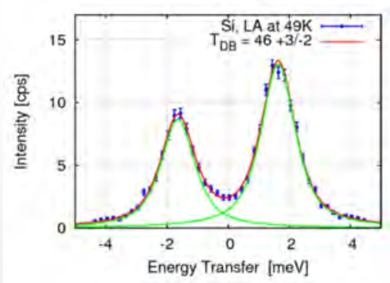
$$s = \mathbf{q}, j \ = \text{Phonon Mode}$$

$$n_s = \langle a_s^+ a_s \rangle \qquad n_s + 1 = \langle a_s a_s^+ \rangle$$

$$|F_s(\mathbf{Q})|^2 = \frac{1}{\omega_s} \left| \sum_d \frac{f_d(\mathbf{Q})}{\sqrt{2M_d}}\, e^{-W_d}\, \mathbf{Q}\bullet\mathbf{e}_{sd}\, e^{i\mathbf{Q}\bullet\mathbf{r}_d} \right|^2$$

Thermal Equilibrium: $n_s = \bar{n}_s \equiv \dfrac{1}{e^{\hbar\omega_s/k_BT} - 1}$ $\qquad \dfrac{S(\mathbf{Q},\omega_s)}{S(\mathbf{Q},-\omega_s)} = \dfrac{\bar{n}_s+1}{\bar{n}_s} = e^{\hbar\omega_s/k_BT}$

$$T_{DB} = \frac{\hbar\omega_s}{k_B \ln(I_+/I_-)}$$



Ingredients

1. Use IXS to measure phonon populations via the asymmetry

$$T_{DB} = \frac{\hbar\omega_s}{k_B \ln\left(I_+ / I_-\right)} \qquad \frac{n_s + 1}{n_s} = e^{\hbar\omega_s / k_B T_{DB}}$$

2. A relation between experimentally determined quantities and the phonon thermal lifetime

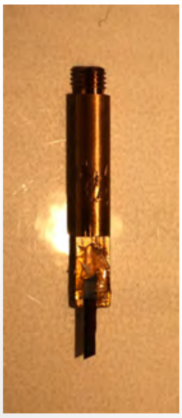
$$n_s - \overline{n}_s = -\tau_s \frac{\partial \overline{n}_s}{\partial T} \mathbf{v}_s \bullet \nabla T$$

Note the dot product -> Effect depends on the relative direction of the phonon group velocity and the thermal gradient

Acoustic Modes
High Symmetry Directions $\mathbf{q} \parallel \mathbf{v}_s$

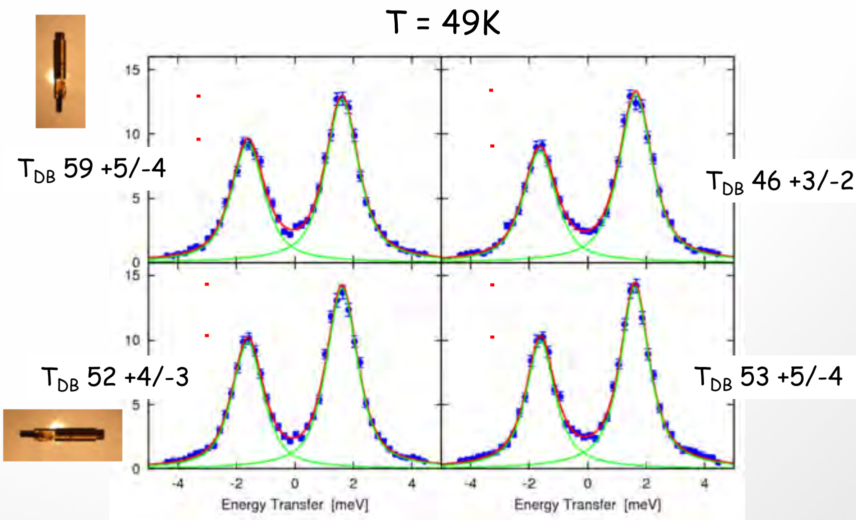
Data - Control

Silicon Wafer
0.28 x 1.7 x 8 mm^3



Mounted inside a Be Cap
With Exchange Gas
With Thermal Shielding

i.e. Standard Operating Procedure

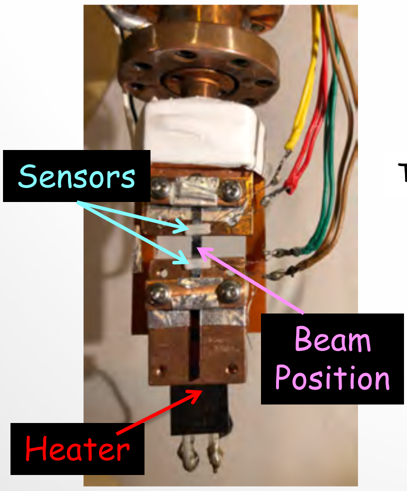


Measure at **Q** = (4 -4 0) or (4 4 0) +- **q**
q parallel and perpendicular to axis of eventual gradient

e.g. **q**=(0.018 -0.018 0) Δ**q**=(0.004 0.006 0.005) rlu
Slits: 6x6 mm^2 9m from sample

Data With A Gradient

Cryostat
cold head

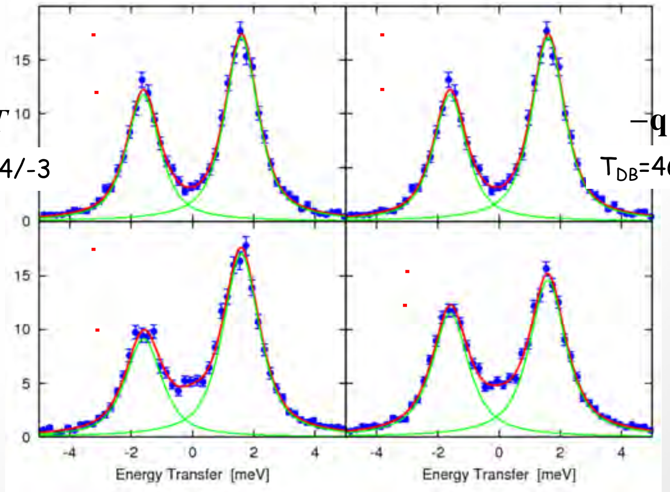


Mounted with a heater to create a thermal gradient

T_{cen}=48K, 1.5 W, 1.2 K/mm

$\mathbf{q} \perp \nabla T$ T_{DB}=49 +4/-3

$-\mathbf{q} \perp \nabla T$ T_{DB}=46 +5/-3



$\mathbf{q} \parallel \nabla T$ T_{DB}=29 +2/-1

$-\mathbf{q} \parallel \nabla T$ T_{DB}=78+11/-9

$$\bar{\tau} = 2.4\ (0.5)\ \mu s$$

Thermal Linewidths

Reasonable violation of detailed balance. The symmetry is broken by breaking the condition (thermal equilibrium) where it applies.

Probe of lifetimes on the us scale (comparable to a linewidth < 1 neV) by coupling to the phonon via a thermal gradient

No scattering method can do this directly (measure linewidth)….
INS has good resolution but dispersing modes only to ueV level

Messages to Take Home

BL43LXU is beginning to do experiments that are difficult elsewhere
using both an excellent beamline and additional specific instrumentation

Robust program going to extreme conditions (P/T) in DACs.

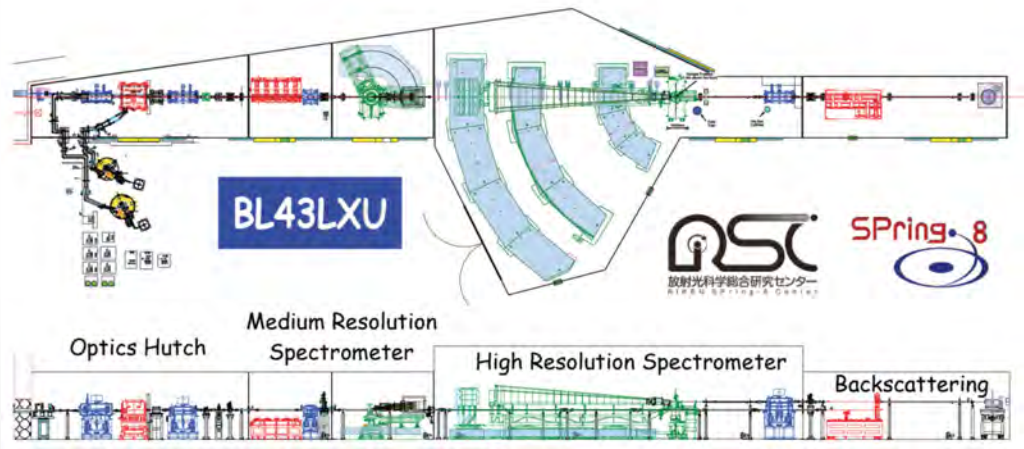
Ongoing work to make practical measurements with high resolution
at low momentum transfers for higher Z liquid investigations.
Push toward a 2000C sample environment (1600 OK, 1990 done, but not yet stable)
Extend fundamental understanding of liquids

Violation of detailed balance in a reasonable fashion
& a new method of probing thermal linewidths.
Possible Relevance to Spintronics, Thermal diodes, etc

Note: this talk only touched on a few aspects of a larger experimental program(s)

- Magneto-elastic coupling & 7T Magnet
- Superconductors & Ferroelectrics
- Single Crystal Elasticity (w/DAC)
- Vibron in liquid and solid hydrogen. (Phonons in H_3S)
- Electronic excitations w/25 meV resolution (NRIXS – Sr_2CuO_3, etc)
- Continuous parameter analysis of liquid spectra
- Symmetry based model for intensity fitting of phonon spectra

AQRB
June 2019



Thank You!

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June 2019