Spooky forest in the hills above Kobe

Hydrodynamic Interaction Between Quasi-elastic and Acoustic Modes

by Inelastic X-Ray Scattering

Alfred Q. R. Baron

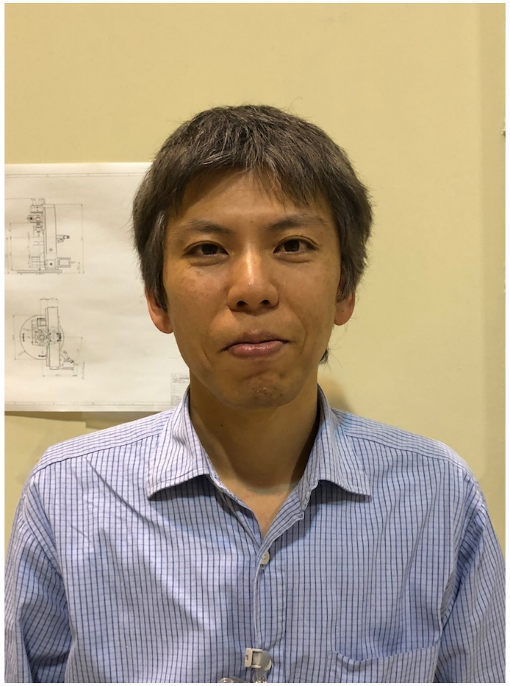
Materials Dynamics Group

RIKEN SPring-8 Center

12th International conference on Inelastic X-Ray Scattering

IXS2022, Oxford, United Kingdom, 25 August, 2022

Collaboration



Daisuke ISHIKAWA

SPring-8: MDG & JASRI

Instrumentation: Optics & T Control
& Liquid Dynamics

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Motivation

Broad: Extend IXS measurements into an interesting and (mostly) previously inacessible region of (Q,E) space

Specific: Investigate the proper response function for liquid dynamics

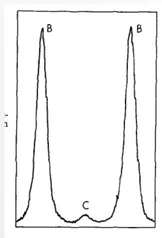
Maybe shed light on long-standing controversy about water

Basic Science Question:

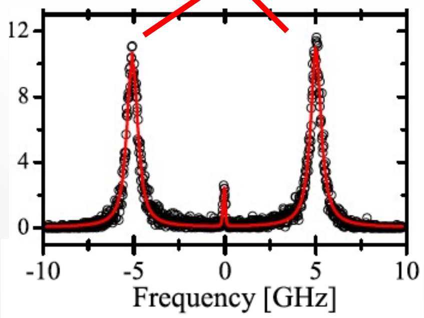
How do liquid dynamics change as you move from the long-wavelength (continuum or hydrodynamic) regime into the mesoscale (nm-scale) atomistic regime?

The example of water:

Acoustic Mode
Stokes & Anti

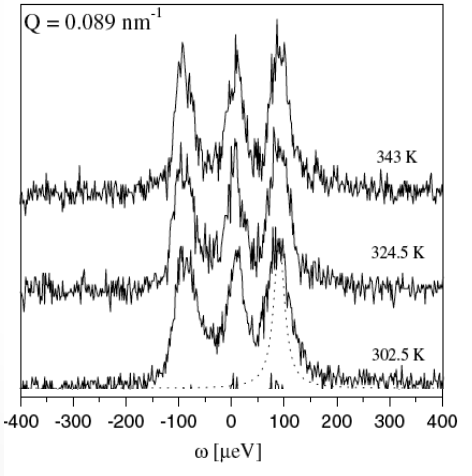


O'Conner, 1967



Zykova, 2017

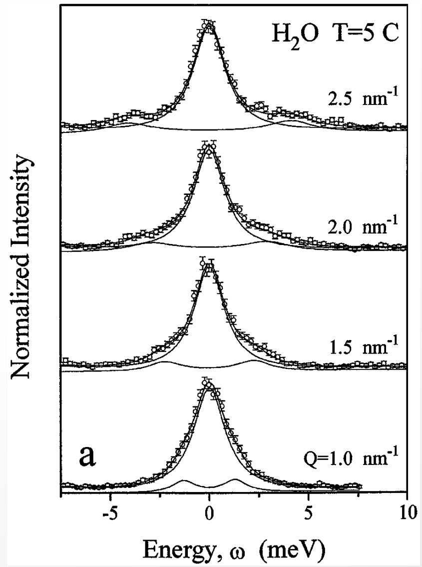
Long wavelength: Q~0.02 nm $^{-1}$
Brillouin Light Scattering
Linearized Navier-Stokes gives
"Rayleigh-Brillouin Triplet"



Masciovecchio, 2004

Shorter: Q<0.09 nm $^{-1}$
Inelastic UV spectroscopy
IUVS

?



Sette, 1996

Shorter and *only short*!
Q>1 nm $^{-1}$
IXS, 1.4 meV

Broad Motivation:
Fill the hole in the Q space where response changes….

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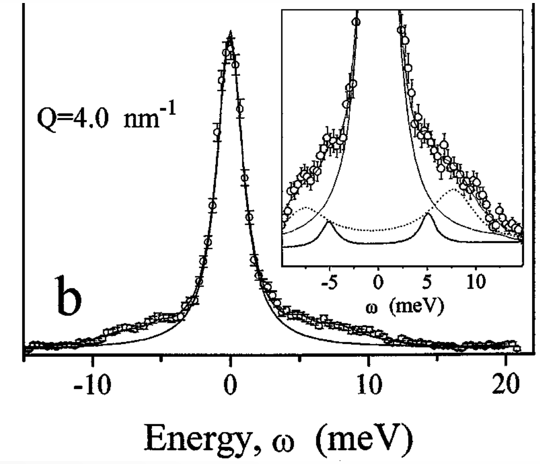
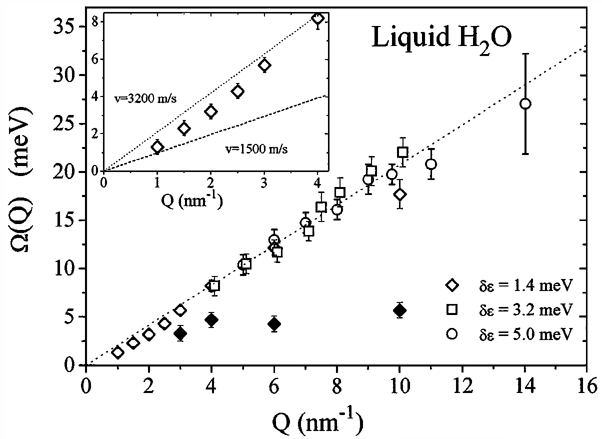
IXS 1996

VOLUME 77, NUMBER 1 PHYSICAL REVIEW LETTERS 1 JULY 1996

Transition from *Normal* to *Fast* Sound in Liquid Water

F. Sette,[1] G. Ruocco,[2] M. Krisch,[1] C. Masciovecchio,[1] R. Verbeni,[1] and U. Bergmann[1]
[1]*European Synchrotron Radiation Facility, B.P. 220, F-38043 Grenoble, Cedex France*
[2]*Universitá di L'Aquila and Istituto Nazionale di Fisica della Materia, I-67100, L'Aquila, Italy*
(Received 1 April 1996)





$$\pi \frac{S(Q,\omega)}{S(Q)} = I_0 \frac{z_0}{\omega^2 + z_0^2} + I_1 \frac{2 z_1 \Omega^2}{\left(\omega^2 - \Omega^2\right)^2 + 4 z_1^2 \omega^2}$$

Basic Function: L+DHO but then 2nd DHO
L+DHO from previous INS (Teixiera et al)

Observed full transition to fast sound (not main topic here but actually a bit surprising)
Presence of an extra mode that appears for Q> 2.5 nm^{-1}
Suggest transverse mode based on analogy with solid water (ice)

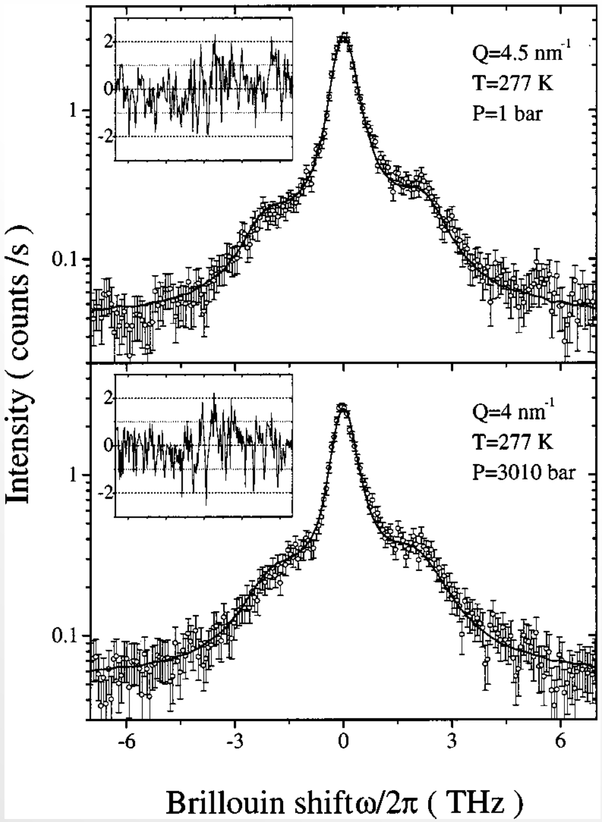
AB Confirmed in first measurement in BL35 (~2000)

IXS 1999

PHYSICAL REVIEW E VOLUME 60, NUMBER 5 NOVEMBER 1999

**Viscoelastic behavior of water in the terahertz-frequency range:
An inelastic x-ray scattering study**

G. Monaco,[1] A. Cunsolo,[2] G. Ruocco,[3] and F. Sette[2]
[1]*Istituto Nazionale di Fisica della Materia, c/o ESRF, Boîte Postale 220, F-38043 Grenoble Cedex, France*
[2]*European Synchrotron Radiation Facility, Boîte Postale 220 F-38043 Grenoble Cedex, France*
[3]*Universitá di L'Aquila and Istituto Nazionale di Fisica della Materia, I-67100, L'Aquila, Italy*
(Received 12 April 1999)



Change the analysis to a memory function (time domain) approach

No direct sound speed in the theory

But also no additional mode:
Just one relaxation & acoustic mode is enough

INS 2000

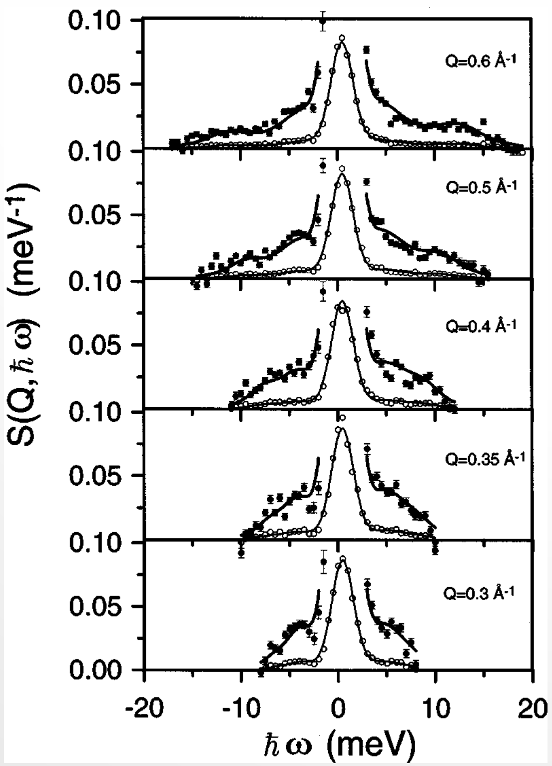
PHYSICAL REVIEW E VOLUME 62, NUMBER 3 SEPTEMBER 2000

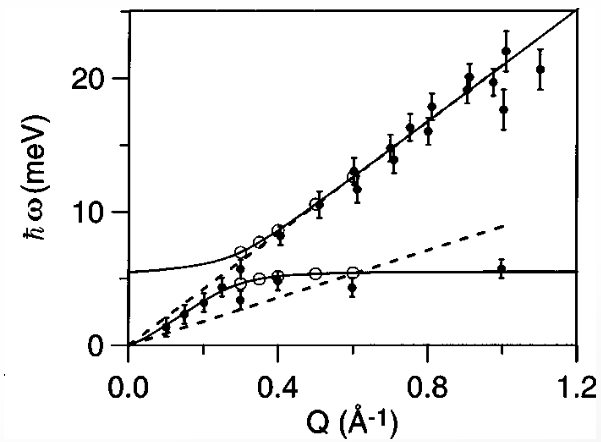
High-resolution neutron scattering measurement of the dynamic structure factor of heavy water

C. Petrillo* and F. Sacchetti
Istituto Nazionale per la Fisica della Materia, Unitá di Perugia, Perugia, Italy
and Dipartimento di Fisica, Università di Perugia, Via A. Pascoli, I-06123 Perugia, Italy

B. Dorner
Institut Laue Langevin, Boîte Postale 156, Grenoble, France

J.-B. Suck
Institute of Physics, Material Research and Liquids, TU Chemnitz, D-09107 Chemnitz, Germany
(Received 20 December 1999)





L+DHO model
See "extra" mode
Suggest anticrossing.

Multiple Pictures

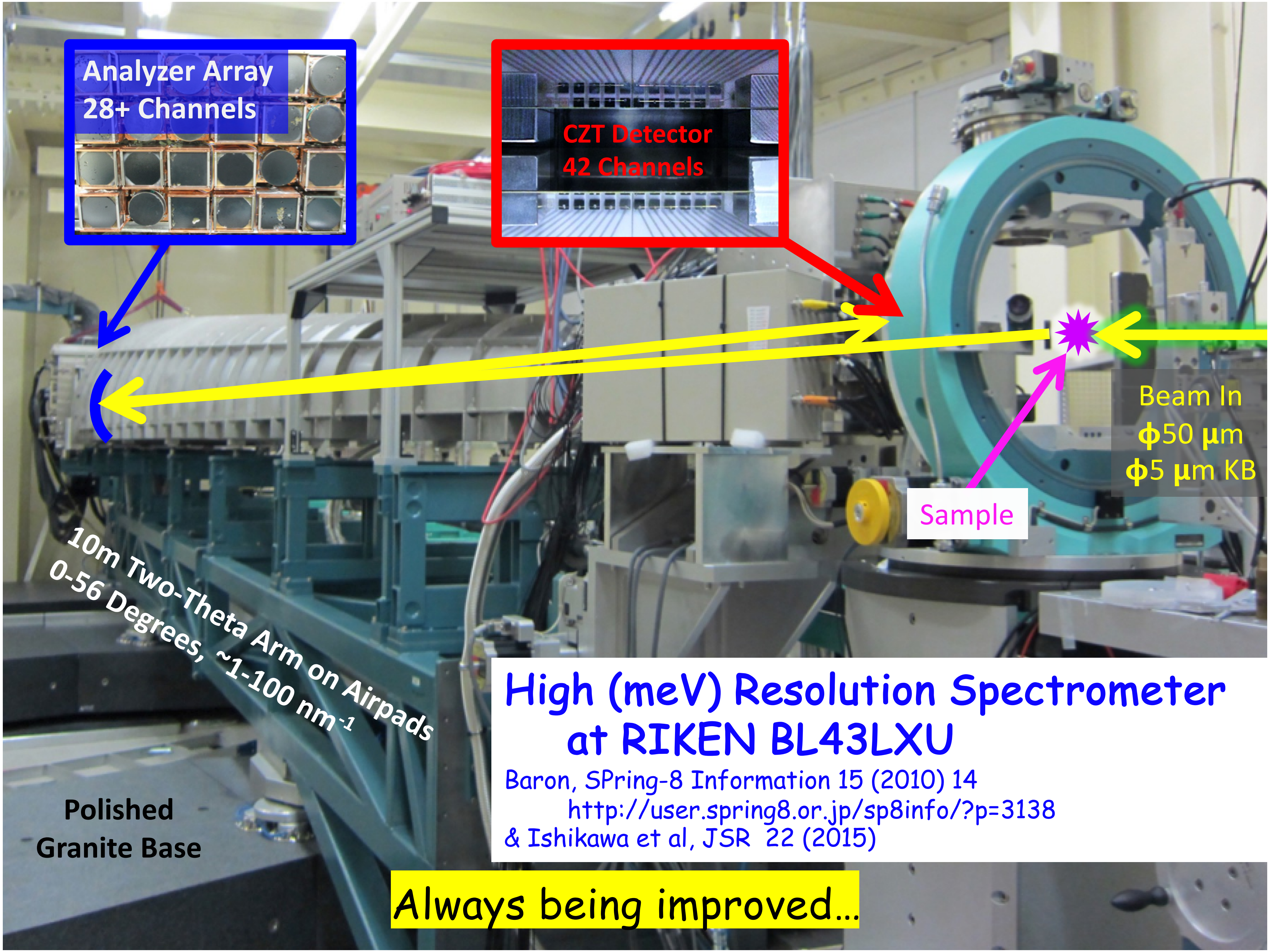
Extra mode due to appearance of transverse dyanmics (IXS)

Extra mode & anticrossing (INS)

No extra mode in viscoelastic (memory function) approach.

Continues: publications in 2008, 2012, & more recent
suggesting one or another or mutliple models

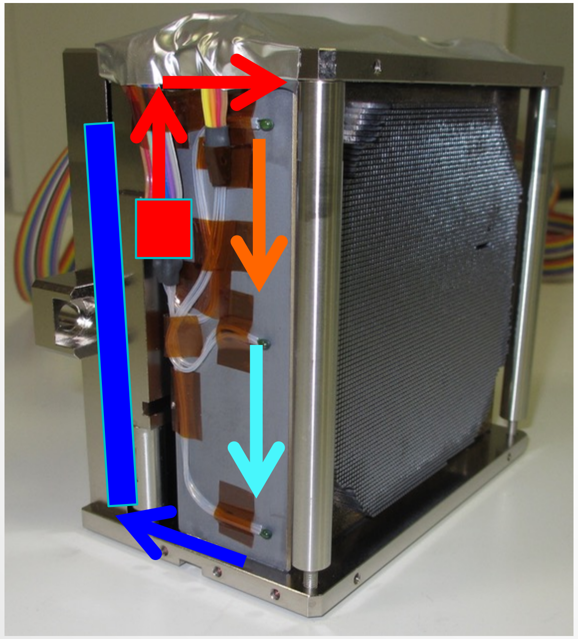
Note: important for some theoretical approaches
Onset of transverse dynamics taken as indication
of onset of critical change dynamical response


Analyzer Array
28+ Channels
CZT Detector
42 Channels
Beam In
ϕ50 μm
ϕ5 μm KB
Sample
10m Two-Theta Arm on Airpads
0-56 Degrees, ~1-100 nm^{-1}
Polished
Granite Base
High (meV) Resolution Spectrometer
at RIKEN BL43LXU
Baron, SPring-8 Information 15 (2010) 14
http://user.spring8.or.jp/sp8info/?p=3138
& Ishikawa et al, JSR 22 (2015)
Always being improved…

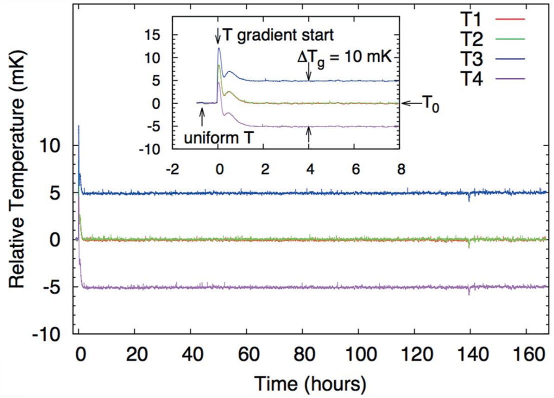
General Push to Low Q & High Resolution

Temperature Gradient Analyzers to Improve Resolution
Grdient compensates for changing Bragg angle

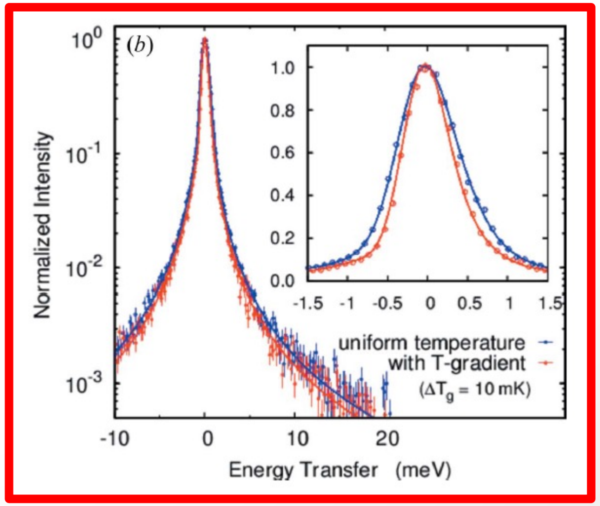
BL43 Analyzers: 9.8 m Radius,
90x94 mm^2 **on a Rectangular Substrate**



Make the analyzer substrate
part of a thermal circuit



Control to ~0.0003 K



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

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

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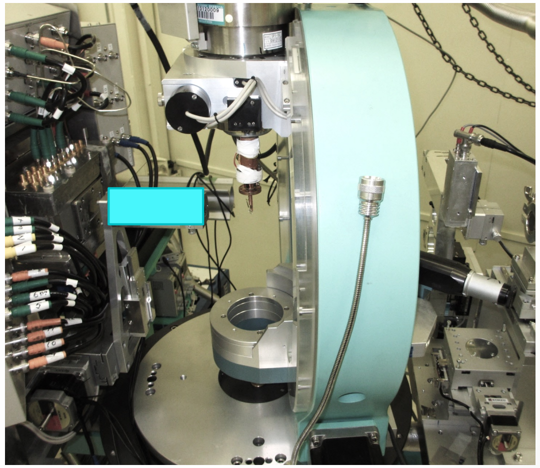
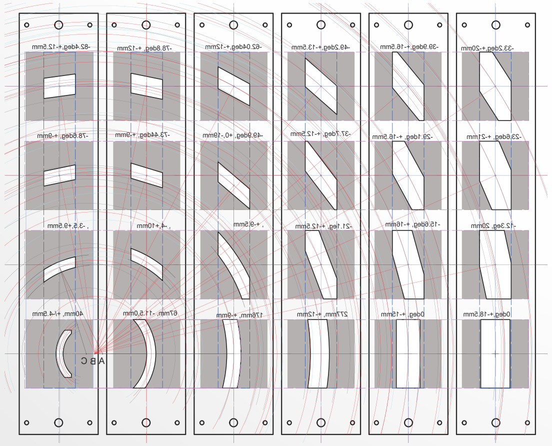
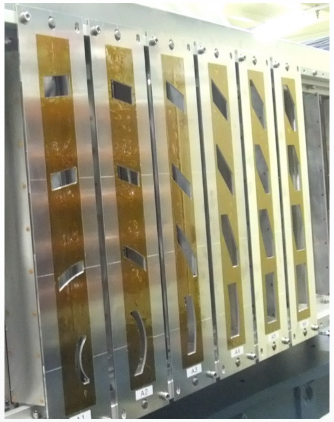
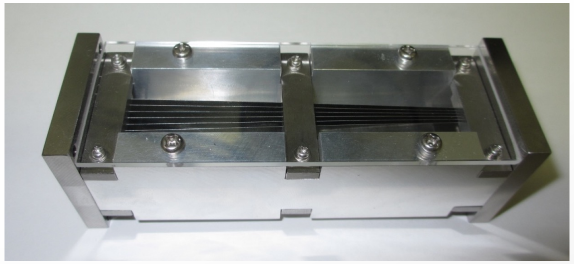
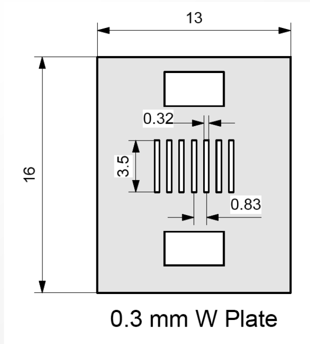
Analyzer Masks & Soller Slit









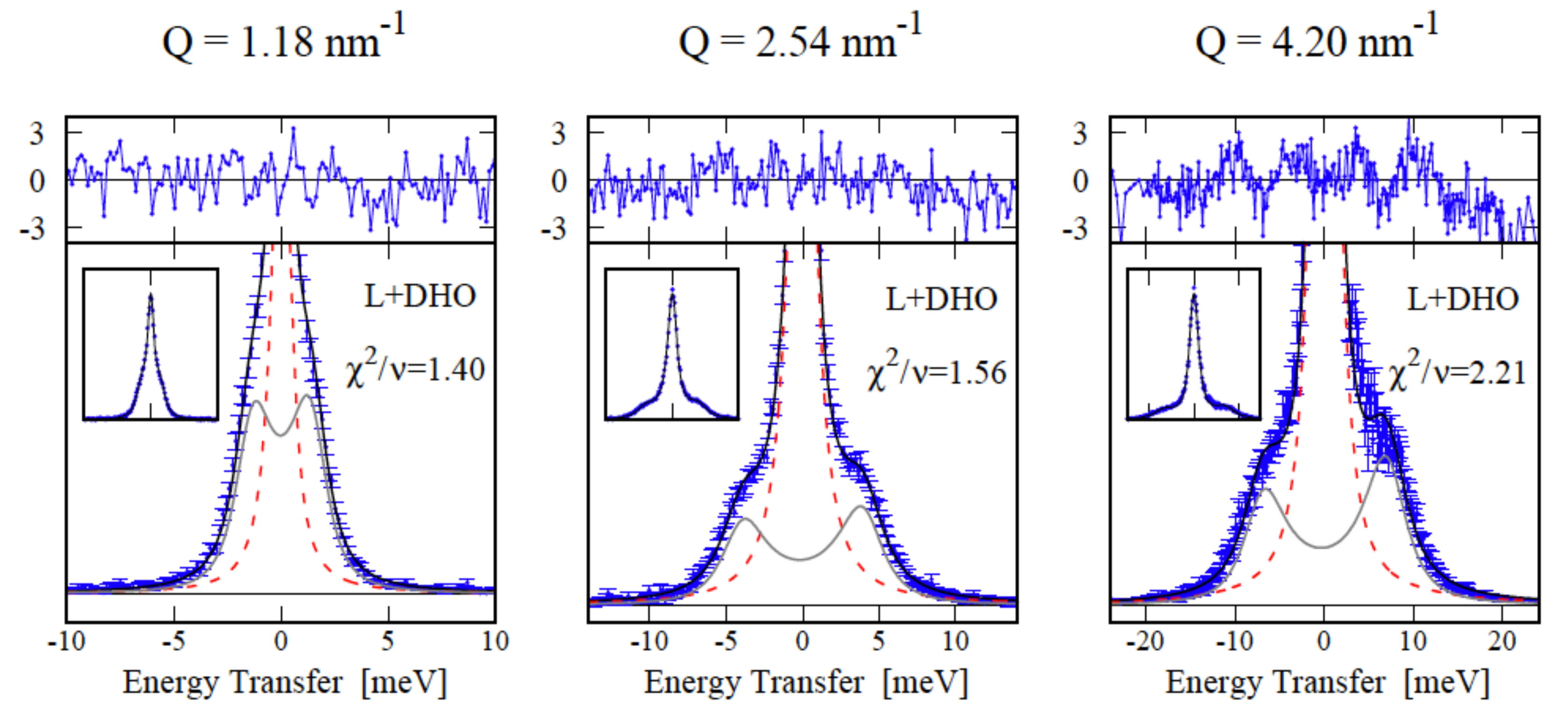


Slits at fixed |Q|
Improves Rates for fixed
Q Resolution: dQ/Q~ 8-15%

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

Ambient Water, IXS Data

ΔE=0.8-1.3 meV, $\Delta Q/Q$~10%



$$\pi \frac{S(Q,\omega)}{S(Q)} = I_0 \frac{z_0}{\omega^2 + z_0^2} + I_1 \frac{2 z_1 \Omega^2}{\left(\omega^2 - \Omega^2\right)^2 + 4 z_1^2 \omega^2}$$

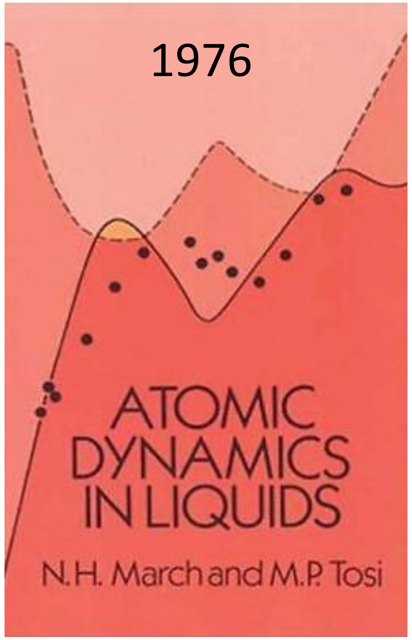
L+DHO Model

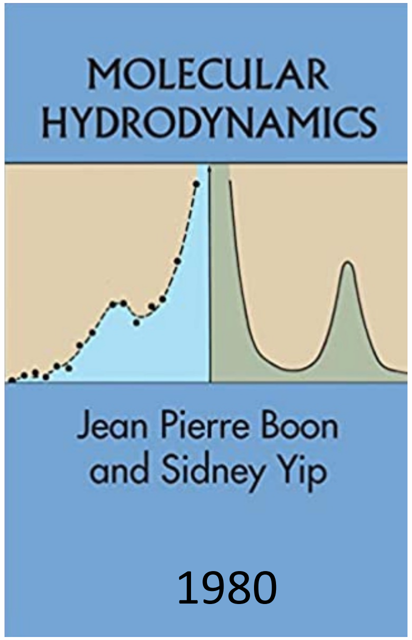
-> clearly something is missing

$P(\chi^2)$ ~ 10^{-2} to 10^{-7}

Ishikawa & Baron, JPSJ 2021

What is the correct lineshape?





Solve the Navier Stokes Equation for Small Dispalcements....
Different forms but can be reduced to same equation

$$S(Q,\omega) = \frac{S(Q)}{\pi}\left[I_0 \frac{z_0}{\omega^2 + z_0^2} + I_1 \frac{2 z_1 \Omega^2}{\left(\omega^2 - \Omega^2\right)^2 + 4 z_1^2 \omega^2} + I_0 z_0 \frac{\Omega^2 - \omega^2}{\left(\omega^2 - \Omega^2\right)^2 + 4 z_1^2 \omega^2} \right]$$

z_0 = Quasielastic Width

z_1 = Acoustic Mode Width

Ω = Sound Mode Energy

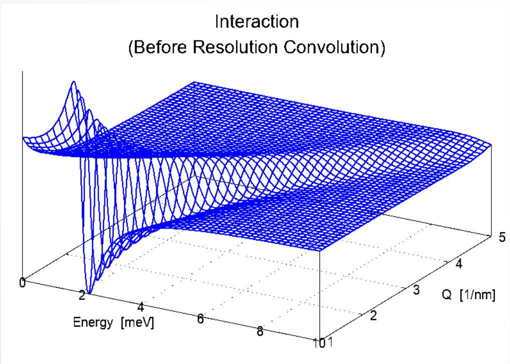
What is the extra term?

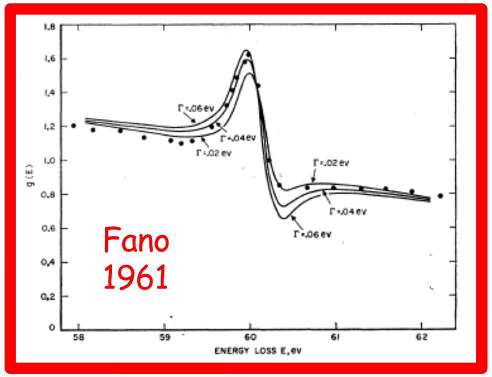
Texts & previous work: “asymmetry parameter” “b” but no explanation.

AB&DI: Interaction from overlap of acoustic mode and tail of the quasielastic

$$- I_0 z_0 \frac{\Omega^2 - \omega^2}{\left(\omega^2 - \Omega^2\right)^2 + 4 z_1^2 \omega^2}$$

Parameters from *both* acoustic and quasielastic mode





Same form as used for rotation-translational coupling (Michel & Naudts, JCP 1978)

Similar (in shape) to a Fano profile (resonance on a continuum)

In this case the continuum is the tail of the quasi-elastic intensity

Interaction Contribution

Note: Impact on spectra is reduced by Q integration!
(bipolar, crossing point disperses)

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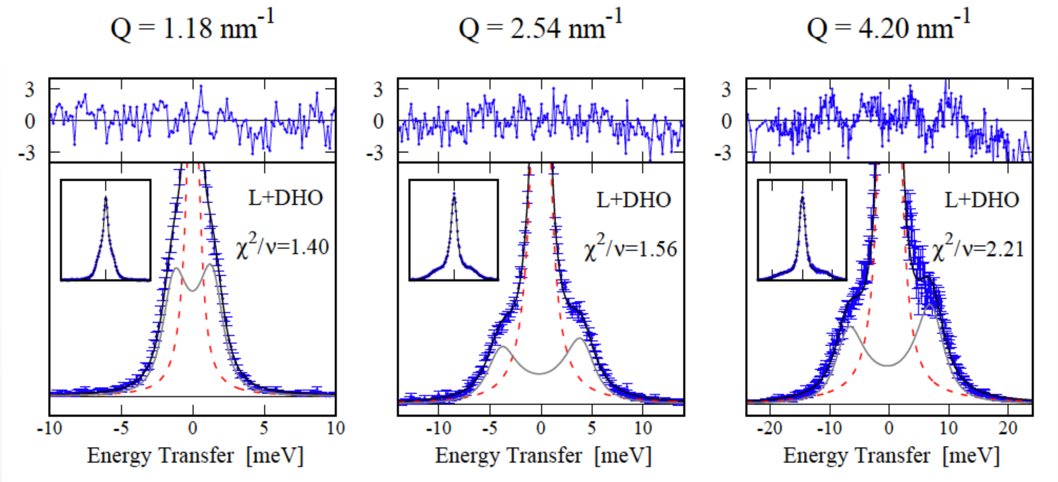
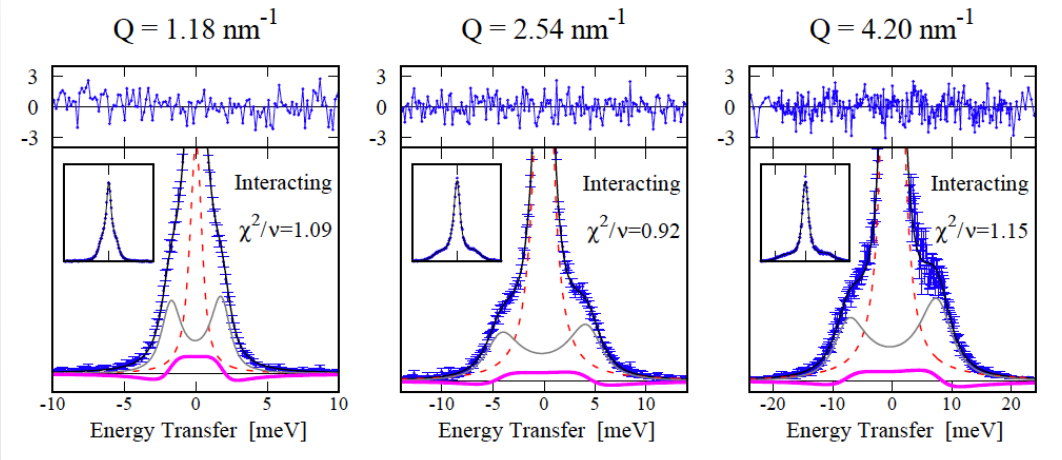
High Resolution IXS Data

ΔE=0.8-1.3 meV, ΔQ/Q~10%



L+DHO Model

$$\pi \frac{S(Q,\omega)}{S(Q)} = I_0 \frac{z_0}{\omega^2 + z_0^2} + I_1 \frac{2 z_1 \Omega^2}{\left(\omega^2 - \Omega^2\right)^2 + 4 z_1^2 \omega^2}$$

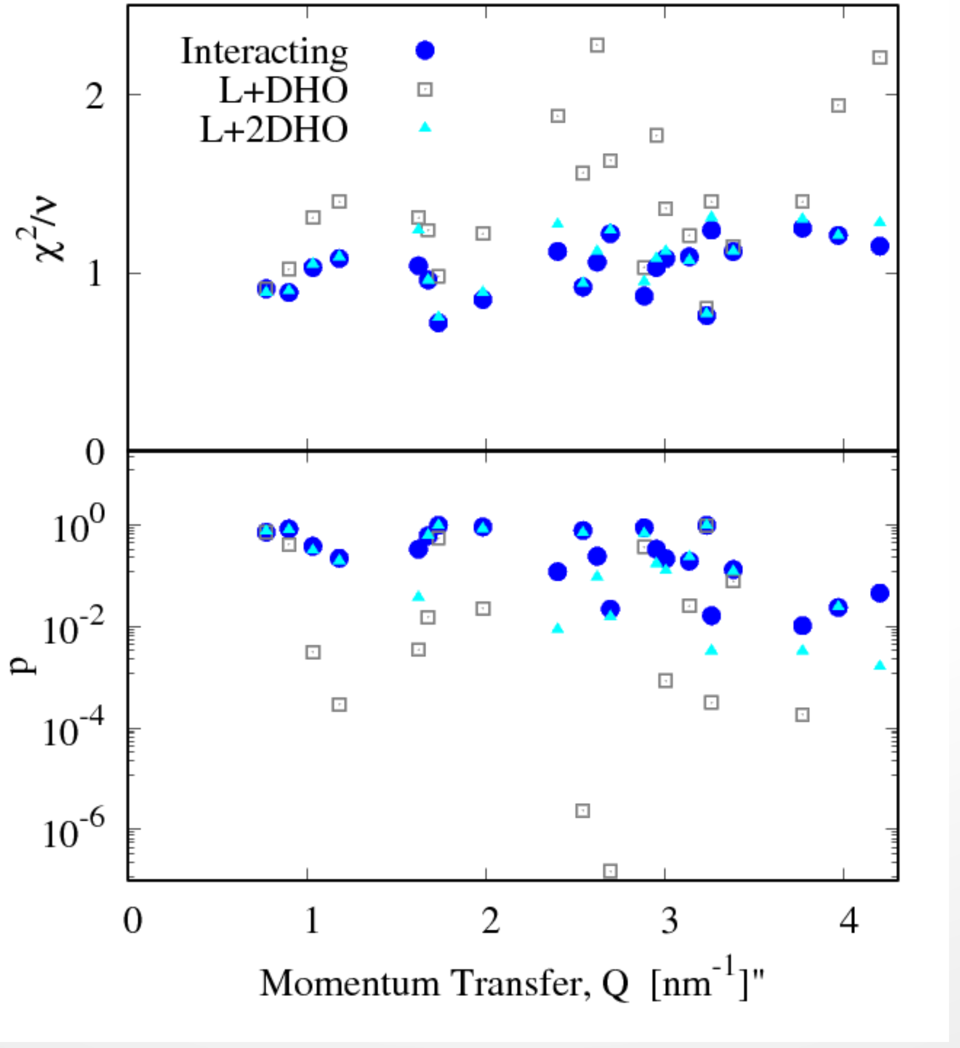


Hydrodynamic Theory

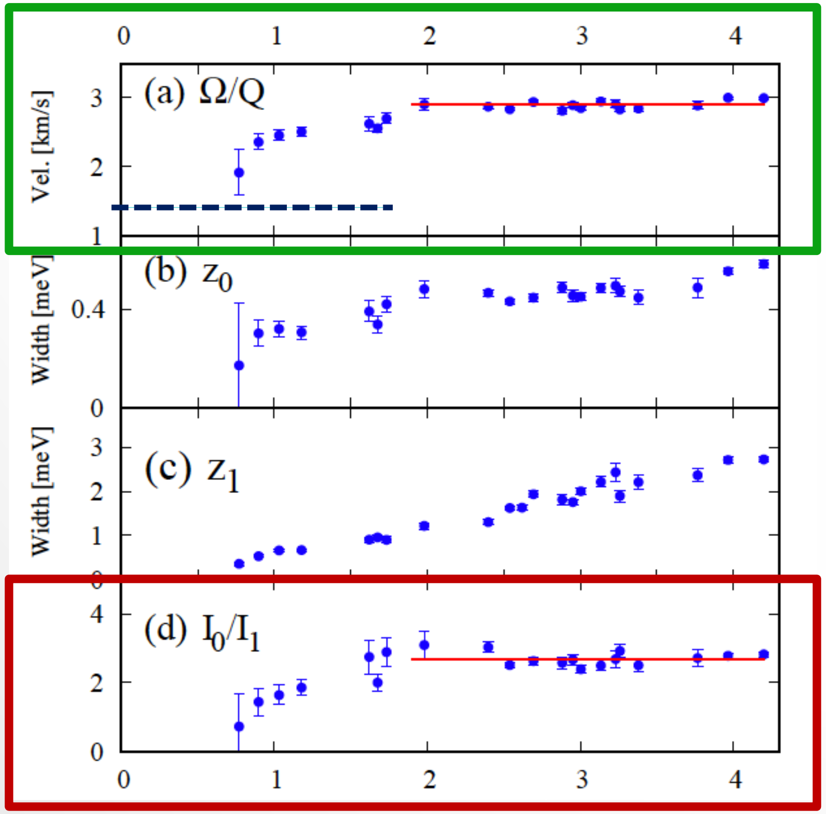
$$S(Q,\omega) = \frac{S(Q)}{\pi}\left[I_0 \frac{z_0}{\omega^2 + z_0^2} + I_1 \frac{2 z_1 \Omega^2}{\left(\omega^2 - \Omega^2\right)^2 + 4 z_1^2 \omega^2} + I_0 z_0 \frac{\Omega^2 - \omega^2}{\left(\omega^2 - \Omega^2\right)^2 + 4 z_1^2 \omega^2} \right]$$

Ishikawa & Baron, JPSJ 2021

Goodness of Fit



Other Parameters



Q

Fitting Results
Ishikawa & Baron, JPSJ 2021

Fast sound clearly observed
Saturated by 2 nm^{-1}

Intensity of the viscous relaxation
(quasi-elastic peak)
increases at higher Q

-> Modified Landau Placzek relation

At high Q: $\frac{I_0}{I_1} = \gamma - 1 \rightarrow \frac{I_0}{I_1} = \gamma\left(\frac{c_\infty}{c_0}\right)^2 - 1$

Estimate: 2.9 Measure 2.7 (not bad)

Concluding Comments

Explanation of a long-standing controversy

No extra propagating mode below 4 nm^{-1} and probably not below 7 nm^{-1}

Agrees with memory function analysis (Monaco, et al – see also Bafile et al PRE 2006)

A good basis from which to begin to explore deviations from simple theory.

Journal of the Physical Society of Japan **90**, 083602 (2021) **Letters**

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Interaction of Acoustic and Quasi-Elastic Modes in Liquid Water on Nanometer Length Scales

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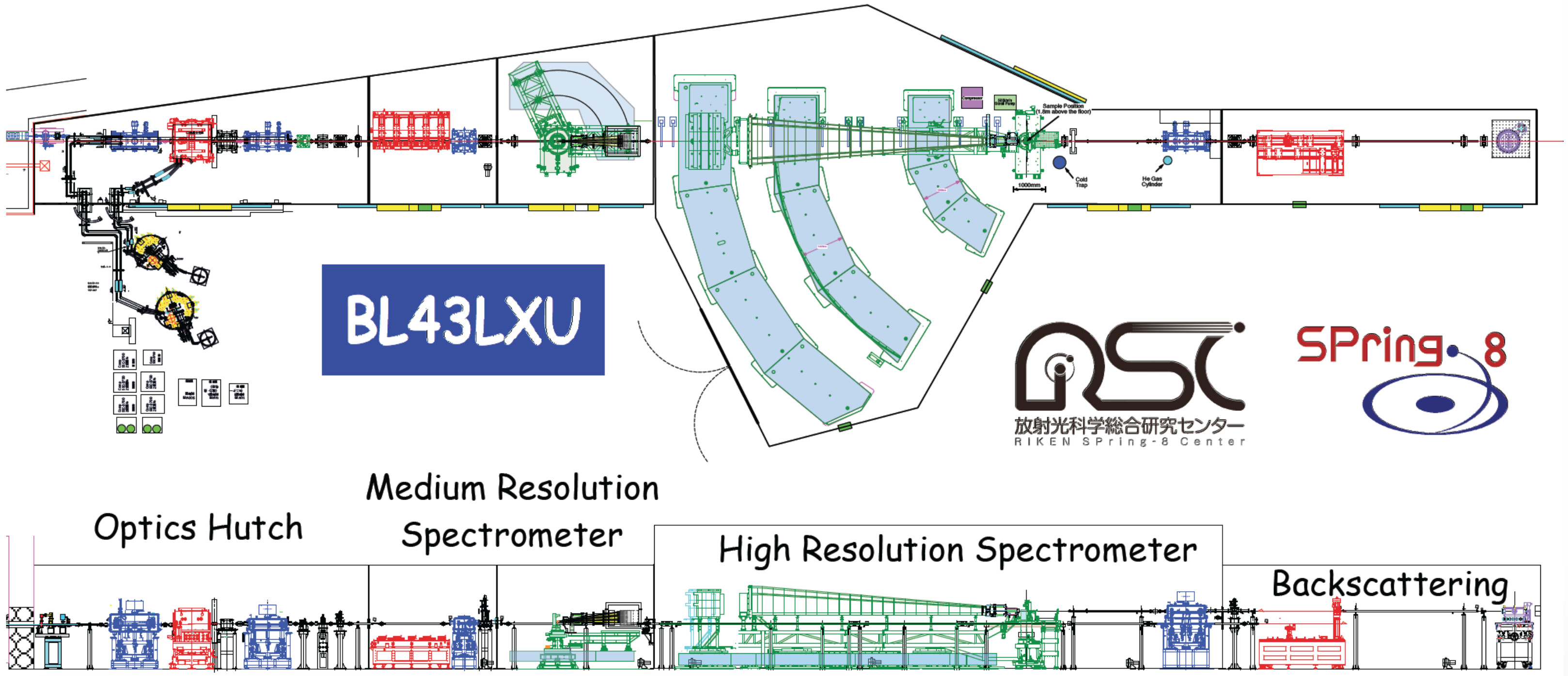

Thank You!

Note: Post-Doc Positions Available
(PhD Students within Japan)

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Sunset in Quetico Provincial Park (Canada)