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Inelastic X-ray Scattering Experiments on Liquid Indium in the Diamond-Anvil-Cell

Ahmet Alataş

Inelastic X-ray & Nuclear Scattering Group

Advanced Photon Source

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Synchrotron High-Pressure Mineral Physics and Materials Science

U.S. Department of Energy

UChicago Argonne LLC

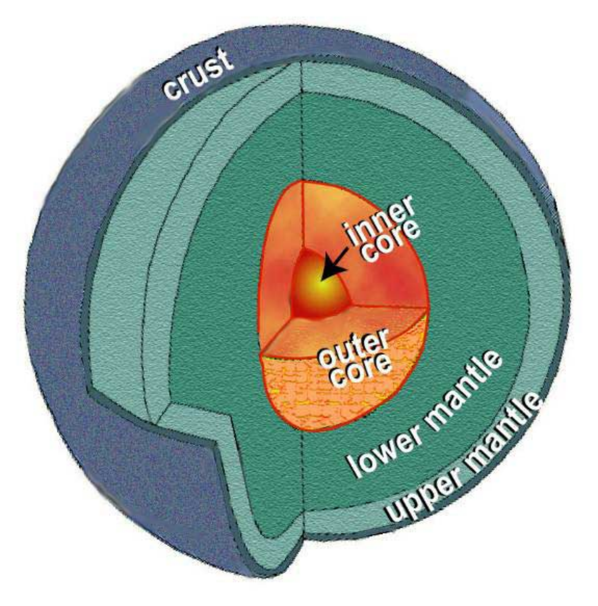
Office of Science U.S. DEPARTMENT OF ENERGY

Outline

- Motivation
- IXS spectrometer
- Liquids
- Application
  - Liquids under ambient pressure
  - Liquid indium under pressure

Motivation – Understanding Interior of Planets

Earth

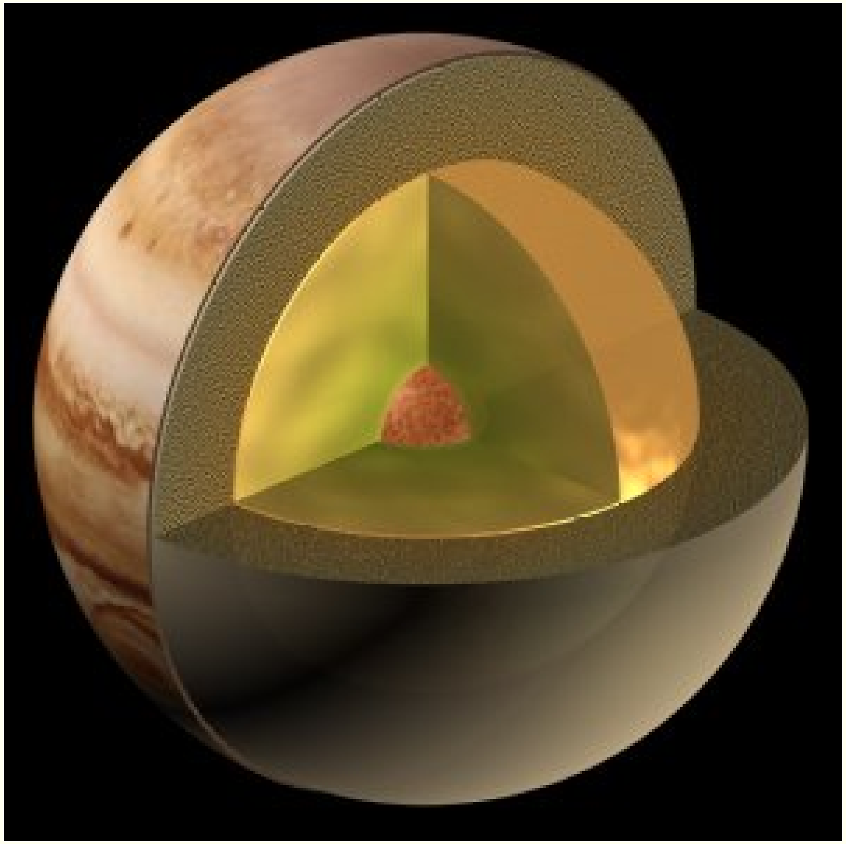


Density?

Sound velocity?

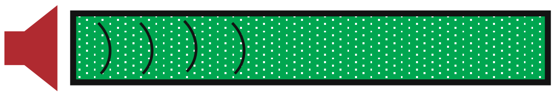
Viscosity?

Jupiter

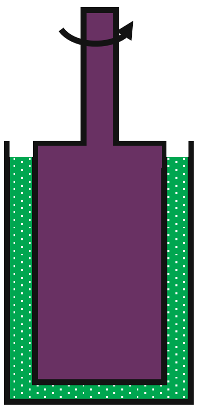


Motivation – Sound velocity & Viscosity

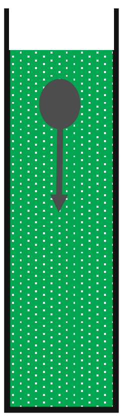
Macroscopic



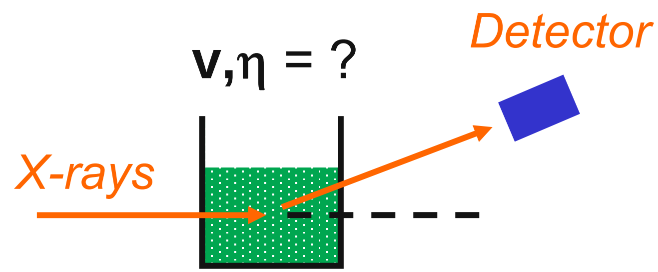
$v \sim s / t$



$F \sim \eta_s v$



Microscopic: APS



Various IXS studies on single & poly crystals:

H.K Mao, *etal*, Science 292, 914 (2001)

G. Fiquet *etal*, Phys. Earth Planetary Sci. Lett., 225, 243 (2004)

D. Antonangeli, *etal*, Phys. Earth Planetary Sci. Lett., 225, 243 (2004)

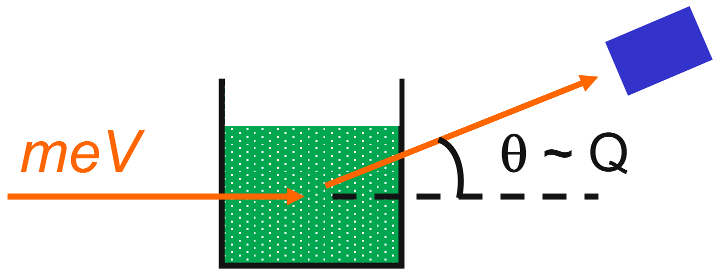
D. Antonangeli, *etal*, Phys. Rev. Lett., 03, 215505 (2004)

Inelastic X-ray Scattering

Detector: δE ~ 1 meV

X-rays:

E 20 ~ keV, δE ~ 1 meV



Intensity (Q, ω) ~ S (Q, ω) $\overset{\text{FT}}{\leftrightarrow}$ Density fluctuations (r, t)

ΔE (meV)

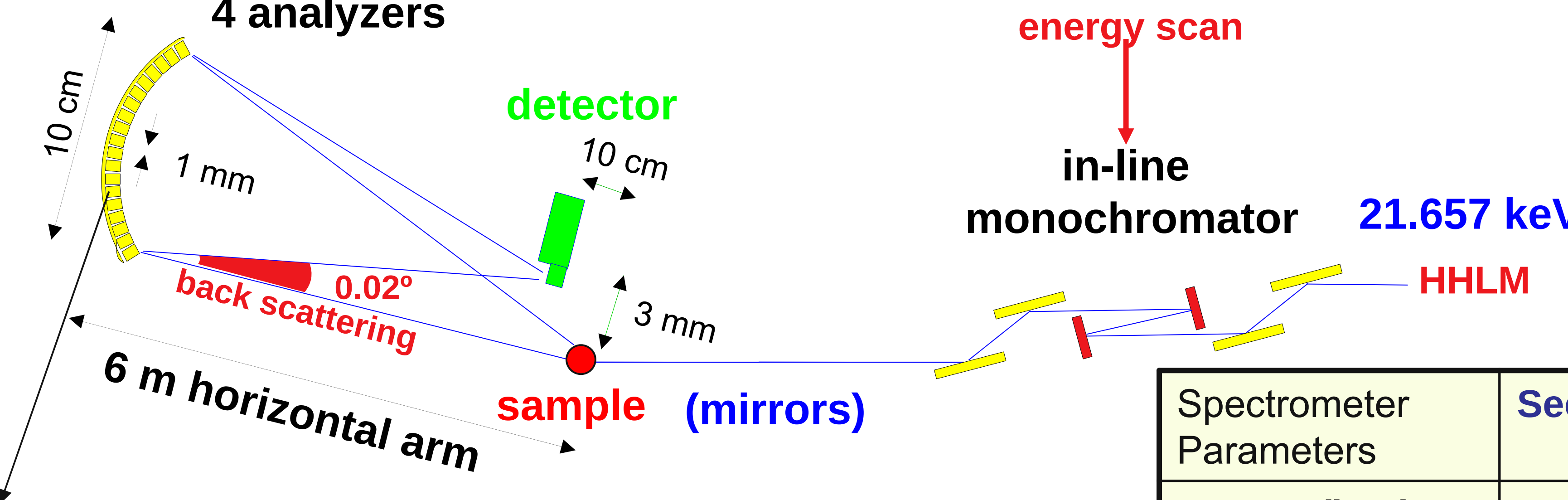
→ sound waves (phonons)

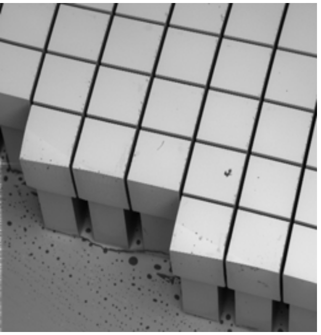
v ~ meV / Q

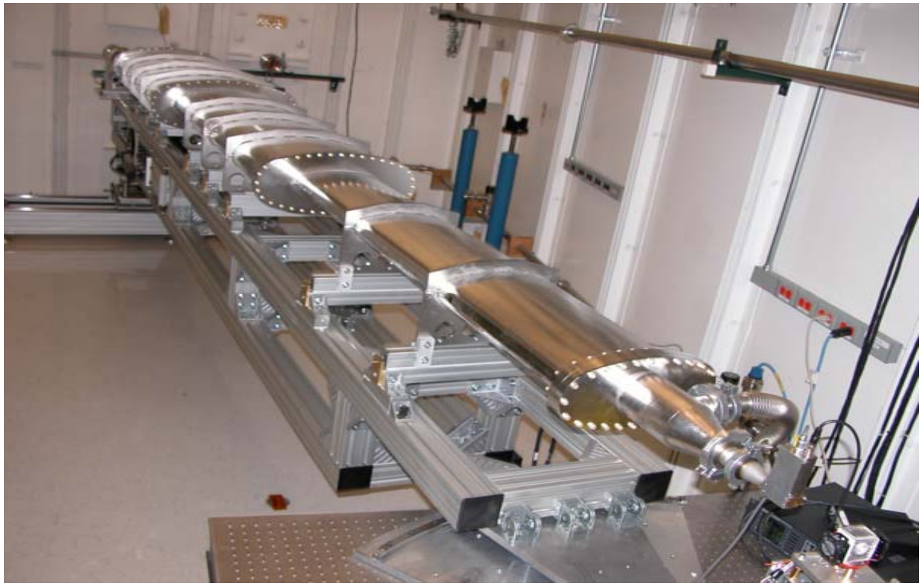
η ~ damping, linewidth

S (Q, ω): Dynamical Structure Factor

IXS Spectrometer at 3 ID-C





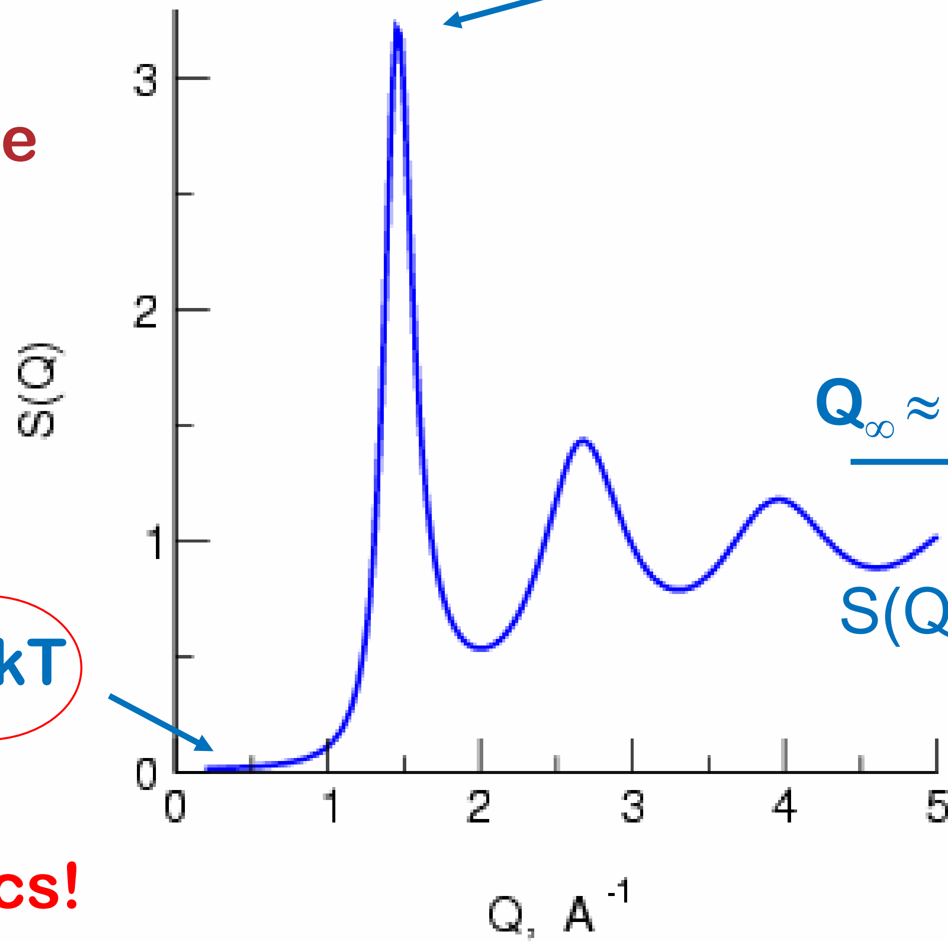


Spectrometer Parameters	Sector 3
Energy (keV)	**21.6**
Reflection	**18 6 0**
# of analyzers	**4**
ΔE_{total} (meV)	**2.2-2.4**
Q-range (nm^{-1})	**32**
ΔQ (nm^{-1})	**1.8 (0.7 used)**
Flux (phts/sec)	**4.5×10^9**
Beam size (μm^2)	**150x200**

Liquid static structure factor S(Q)

$\mathbf{Q_{max}} \approx 2\pi/\sigma_{\mathbf{HS}}$

Static structure factor



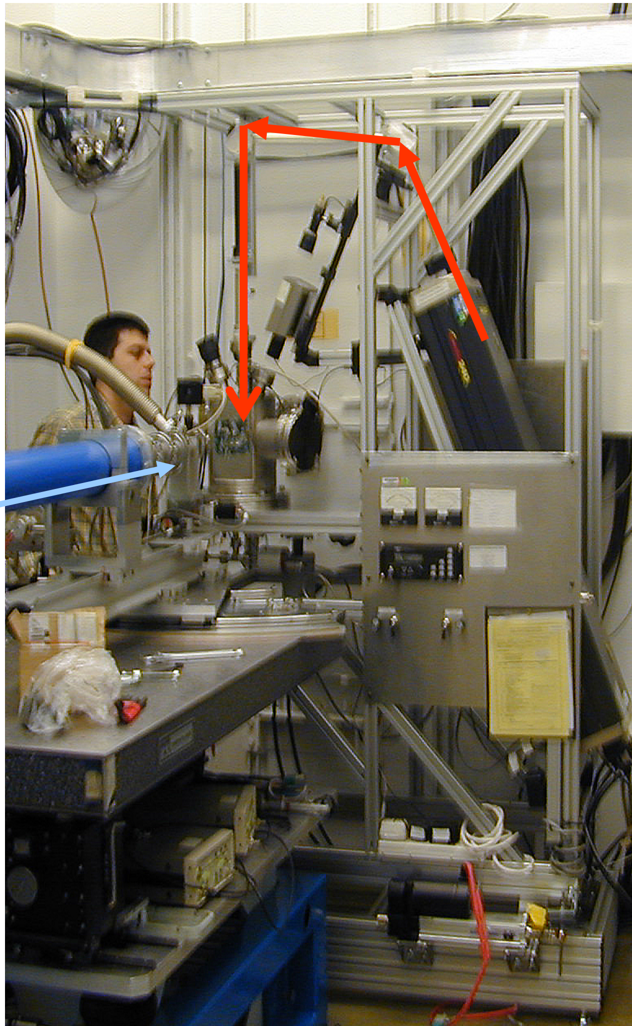
$\mathbf{Q_{\infty}} \approx 2\pi/\delta\sigma_{\mathbf{HS}}$

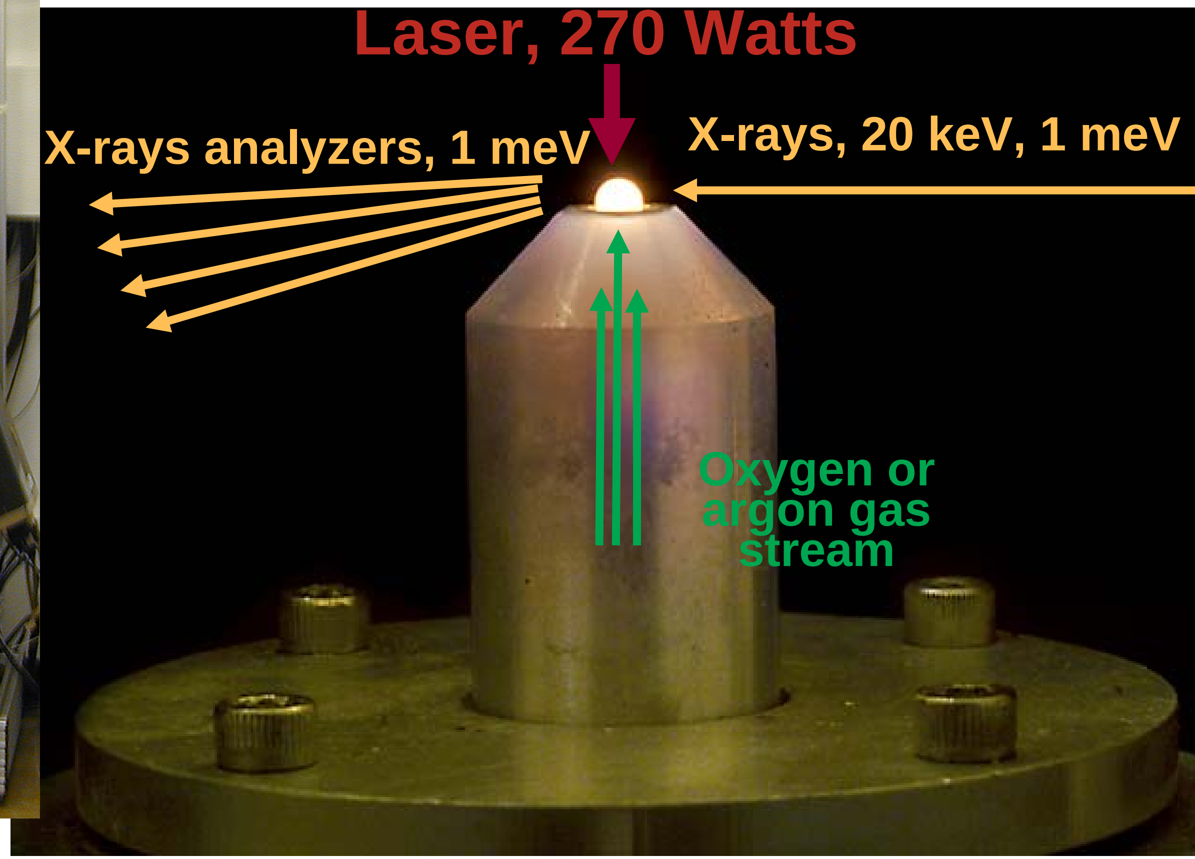
$S(Q\rightarrow\infty) = 1$

$\mathbf{S(Q=0) =} \chi\rho\ \mathbf{kT}$

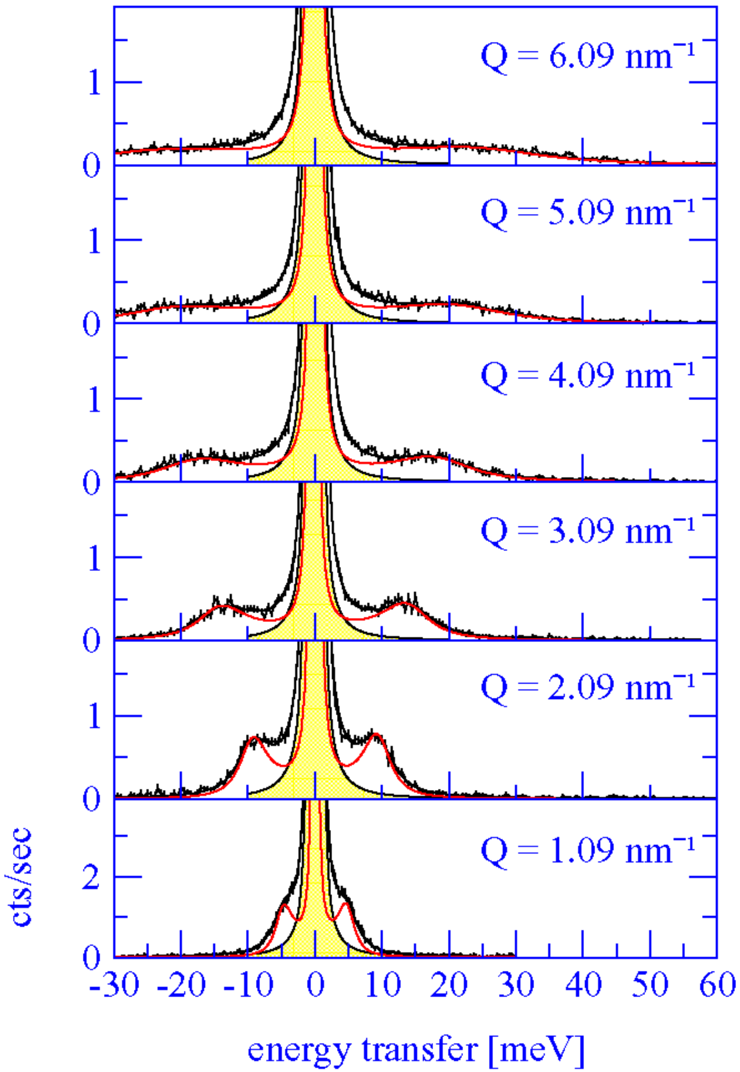
Dynamics!

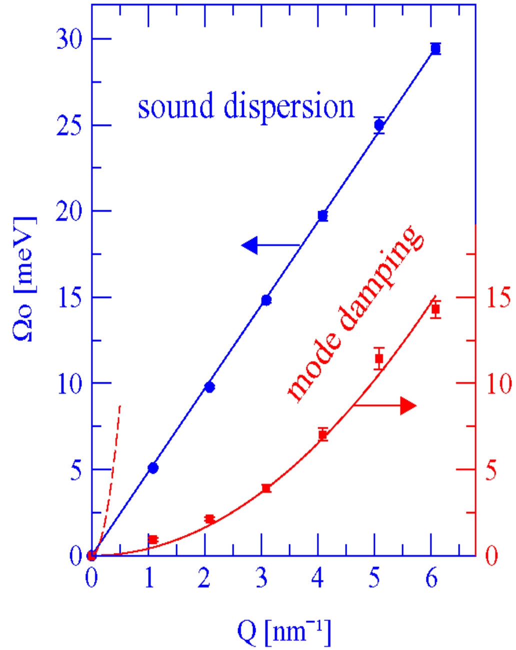
Liquids under ambient pressure – Levitation method

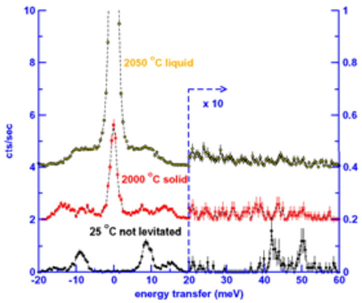




Liquid Al_2O_3 @ 2050°C







c = 7350 m/s
solid:10500 m/s

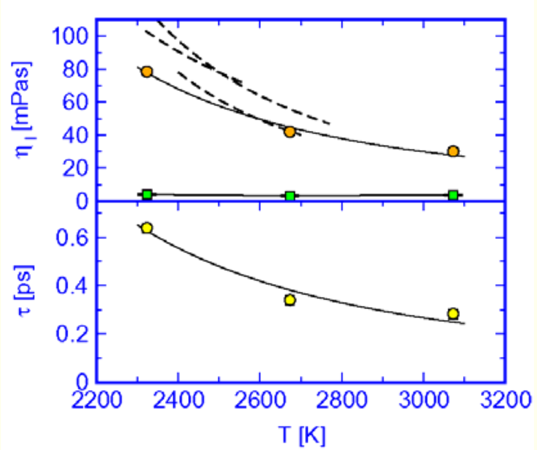
Damping is too low by factor 10 for hydrodynamic interpretation!

However:perfect viscoelastic oscillator

Hydrodynamics to Viscoelastics

Frequency dependent viscosity

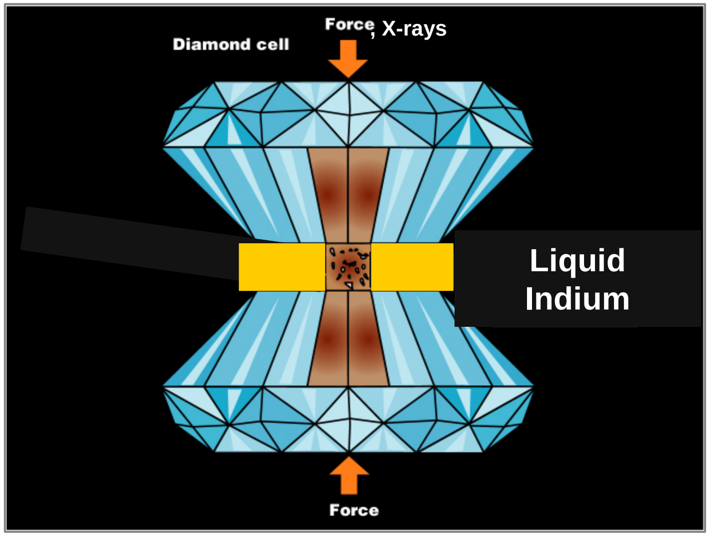
$$\eta(\omega)=\frac{\eta_0}{1+i\omega\tau}+\eta_\infty$$



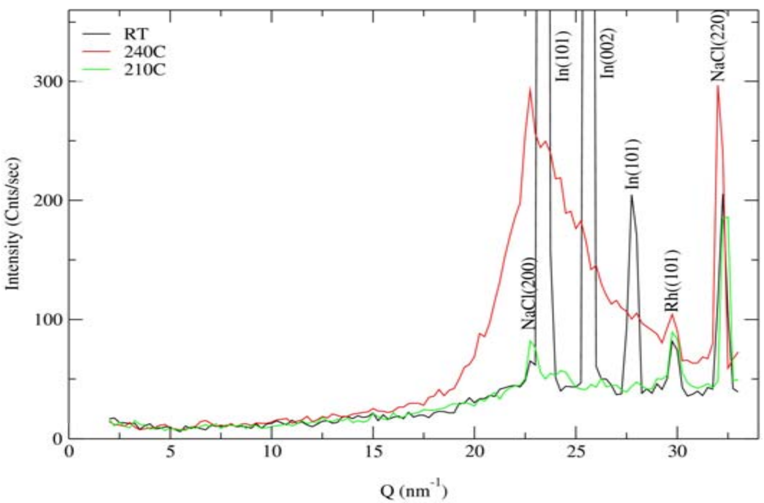
Alumina : H. Sinn et al.,Science, 299: 2047, 2003
Silicon : A. Alatas etal, J. Phys. Chem. Solids, 66, 2230 (2005)
Titanium: A.H. Said etal., PRB, 74, 172202 (2006)

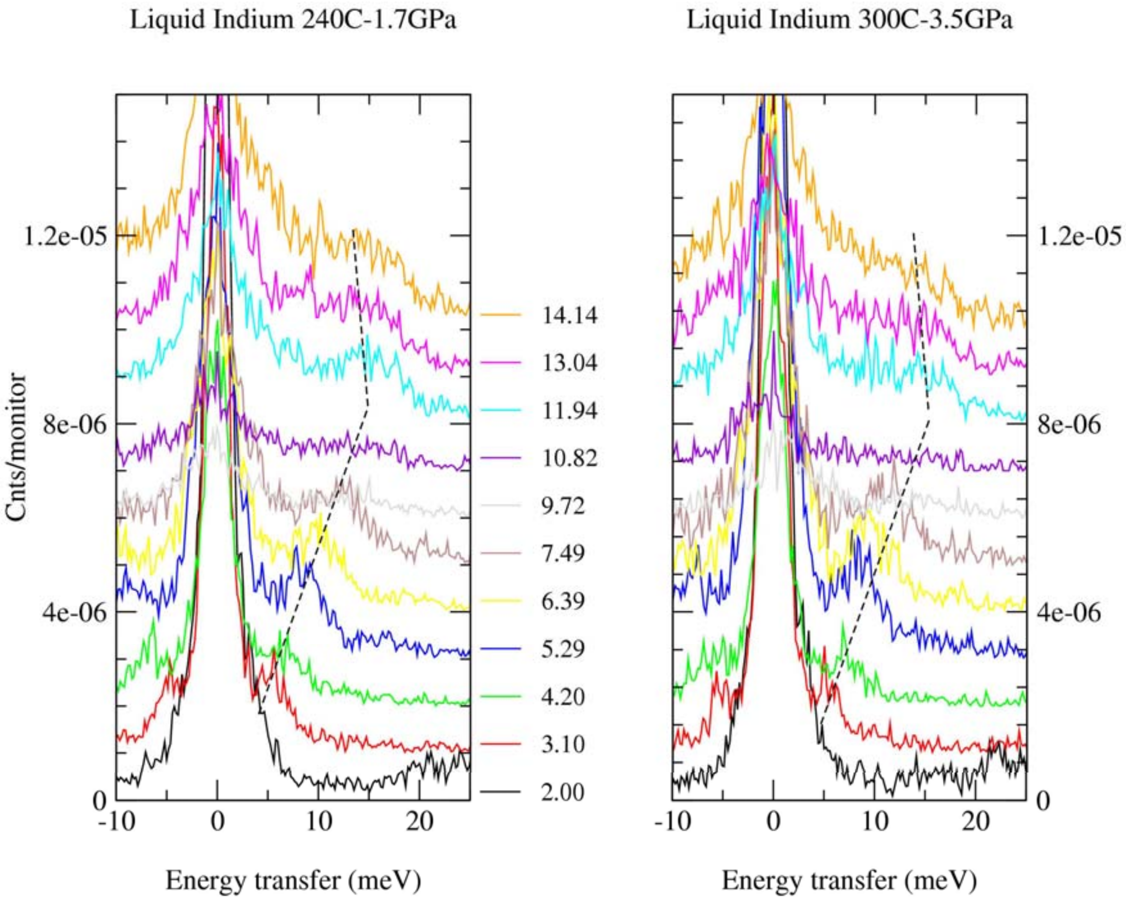
Liquid Indium under Pressure

Cell externally heated

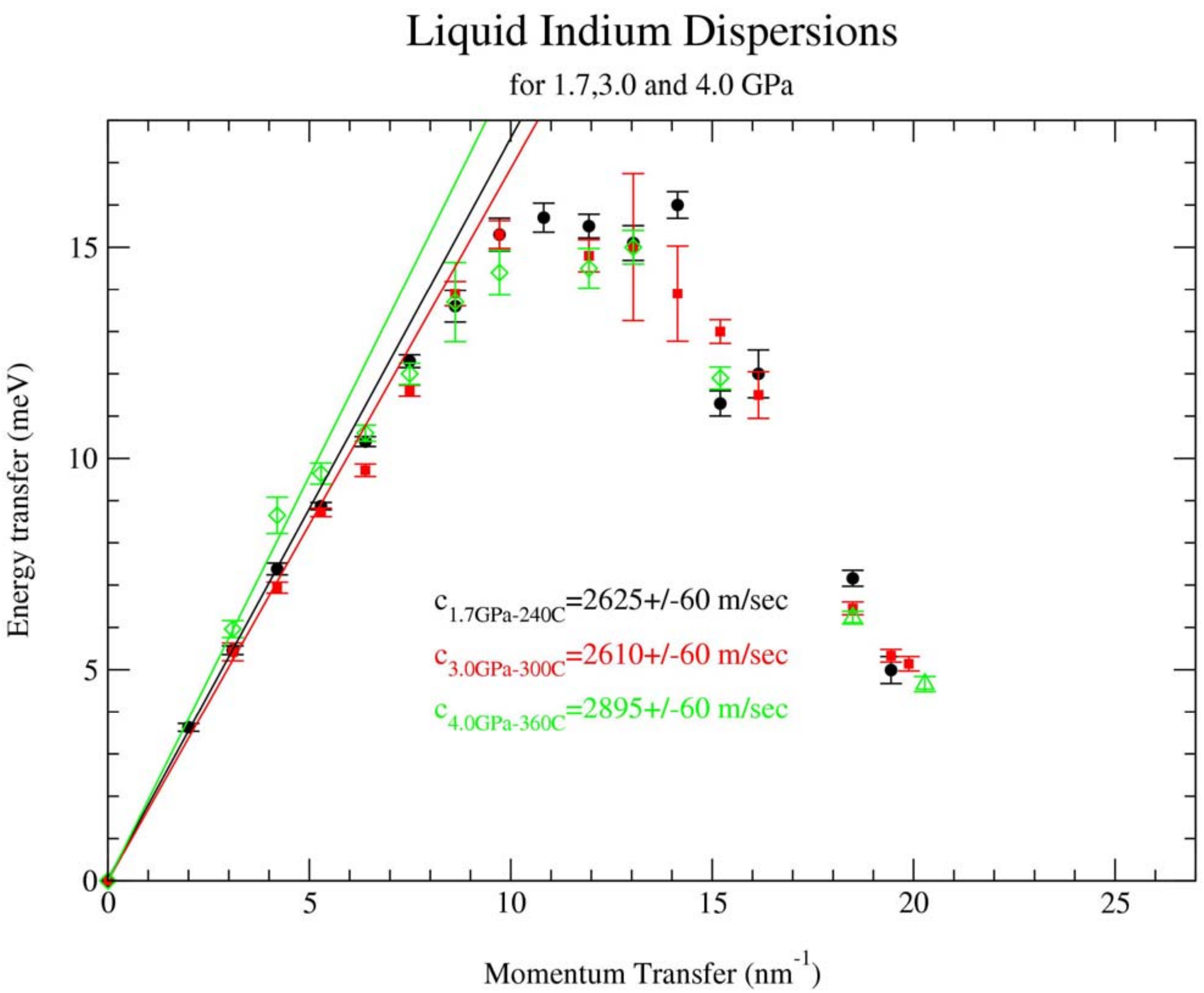


S(Q) of Indium at 2 GPa





Sound Velocity of Liquid Indium



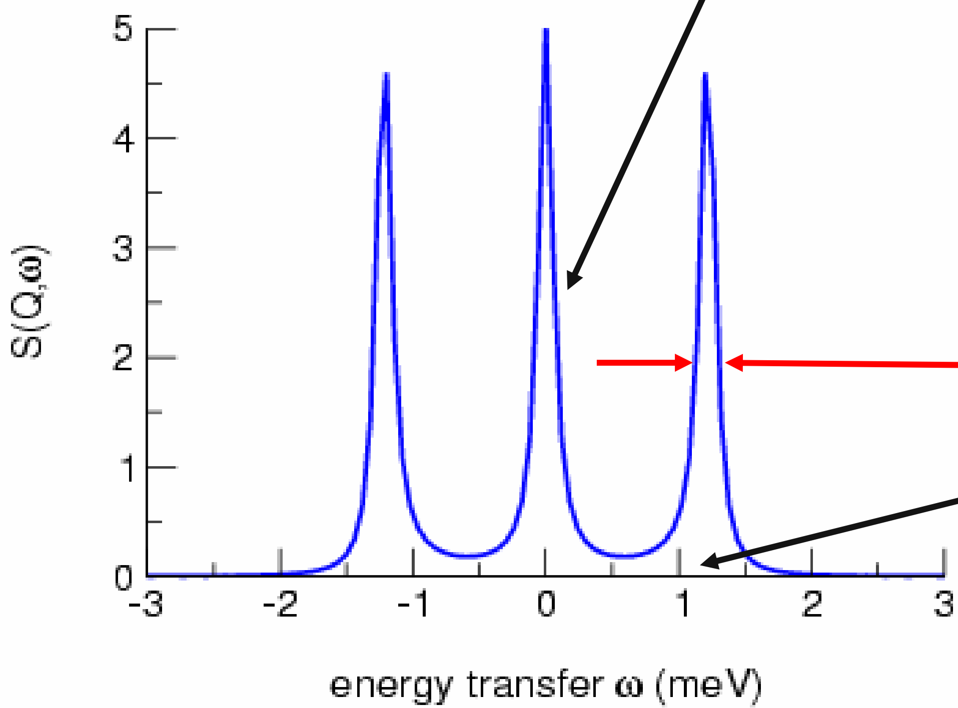
Viscosity from IXS

Liquids in the Hydrodynamic region - S(Q~0) : 3 Lorentzian Peaks

$$\frac{S(Q,\omega)}{S(Q)} \approx A_0 \frac{\Gamma_0}{\omega^2+\Gamma_0^2} + A_s \frac{\Gamma_s}{(\omega-\omega_s)^2+\Gamma_s^2} + A_s \frac{\Gamma_s}{(\omega+\omega_s)^2+\Gamma_s^2}$$

Rayleigh-peak

$$\Gamma_0 = \frac{\kappa}{\rho m c_p} Q^2$$



Brillouin doublet:

$$\Gamma_S = \frac{1}{2\rho m}\left[\frac{4}{3}\eta_S + \eta_B + \boxed{\frac{A_0}{2A_S}}\frac{\kappa}{c_P}\right] Q^2$$

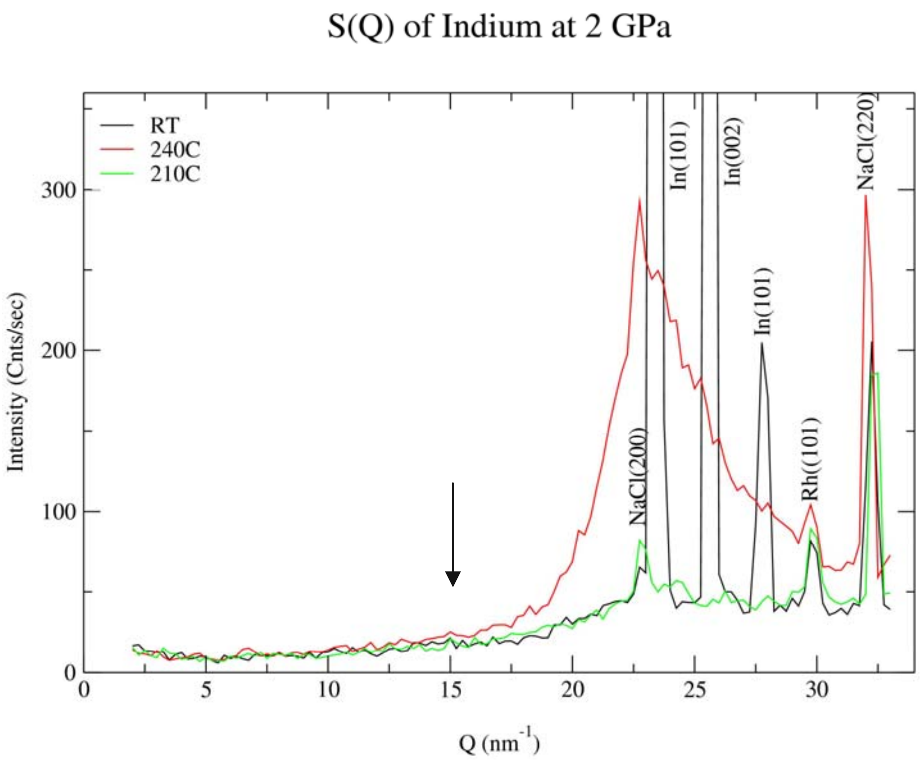
Landau-Placzeck ratio

$$\omega_S = \pm c_S Q$$

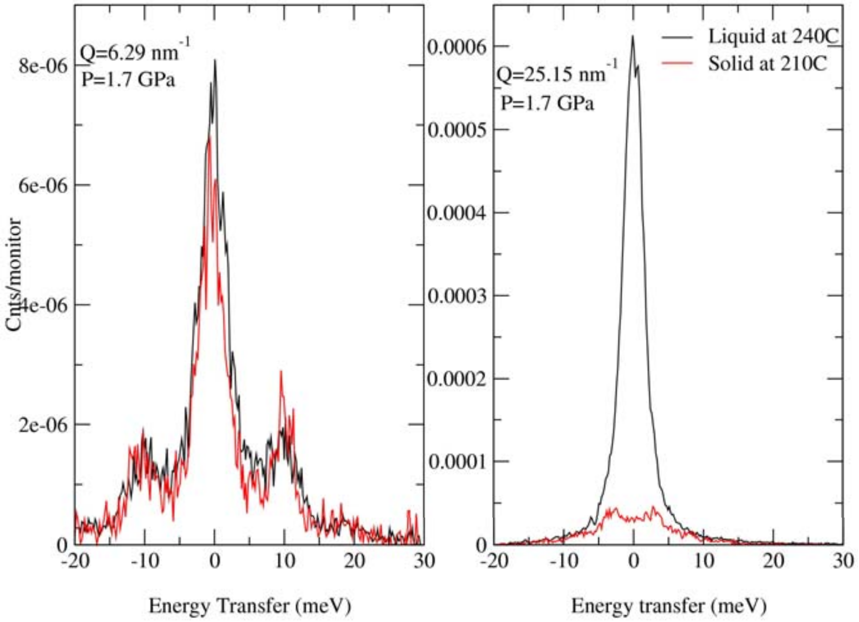
Light scattering

Central Peak

Implication of background up to 15 nm^{-1} from S(Q)



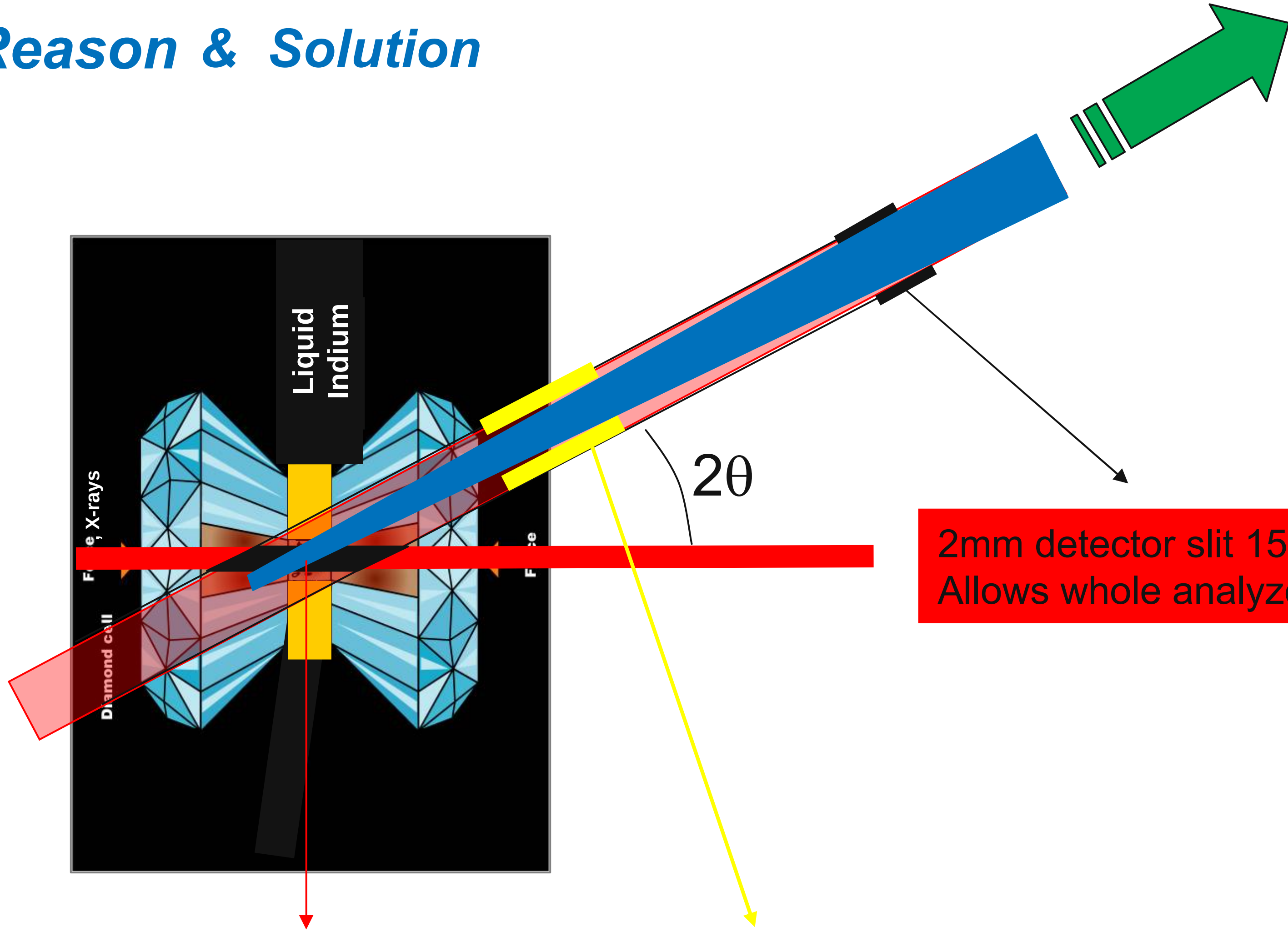
Elastic peak from hot solid sample shows background at small Q



Reason: scattering from diamonds

Reason & Solution

Analyzer
~ 6m away



2mm detector slit 15 cm from cell
Allows whole analyzer to be illuminated

Scattering
volume

Collimator between cell and detector slit

Acknowledgments

IXN Group at APS

- E.E Alp
- B. M. Leu
- A. H. Said
- H. Sinn*
- W. Sturhahn
- J.Zhao

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- G. Shen

Consortium for Advanced Radiation Sources, University of Chicago

- V. B. Prakapenka

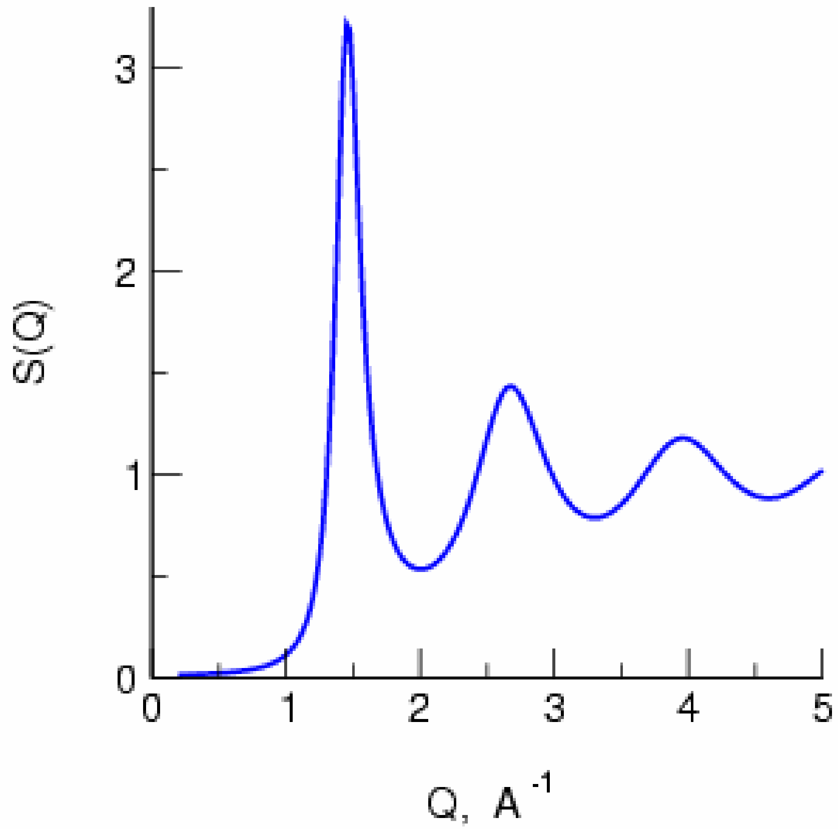
Summary

- IXS experiment successfully applied to liquid in the DAC.
- Reason for elastic background identified.
- Use of collimator will reduce this background.
- New opportunity for studying liquids under pressure.

And

- Smaller beam size available at Sector 30 (5 x 40 μm^{-1}).

Can we get viscosity and viscoelasticity from only S(Q) ??



Sound Damping: Mode-Coupling Approach

S(Q) → **Self consistent Mode Coupling Approximation** ← ρ

Self consistent Mode Coupling Approximation → F(Q, t)

F(Q, t) ↓ Fourier Transform

S(Q, ω)

↓ fit with generalized hydrodynamics

η(Q), τ(Q), η_∞(Q)

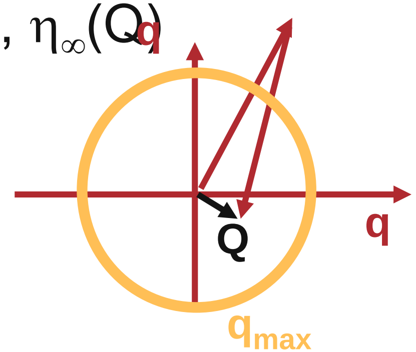
No fit parameters !!

generalized Langevin equation:

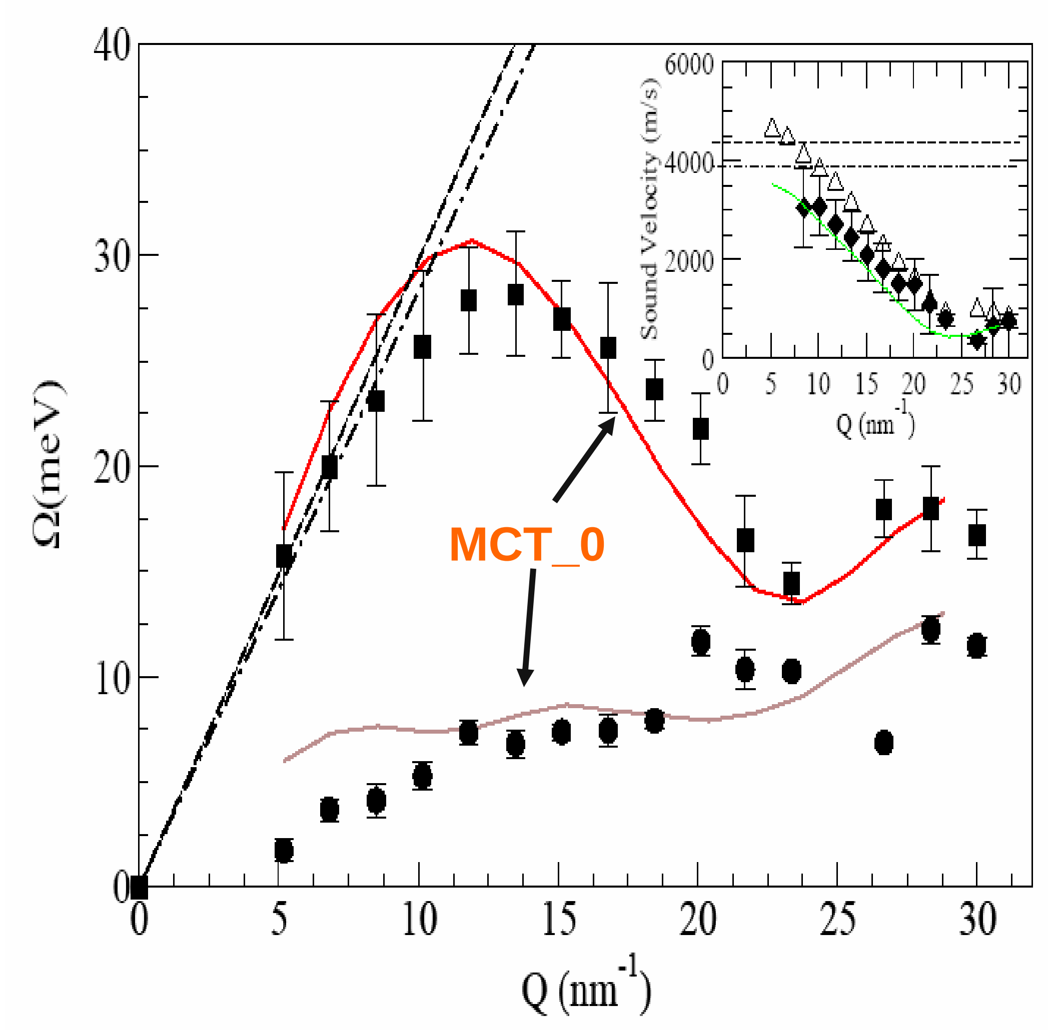
$$\ddot{F}(Q,t)+\int_0^t dt'\ M(Q,t')\,\dot{F}(Q,t-t')+\Omega^2 F(Q,t)=0$$

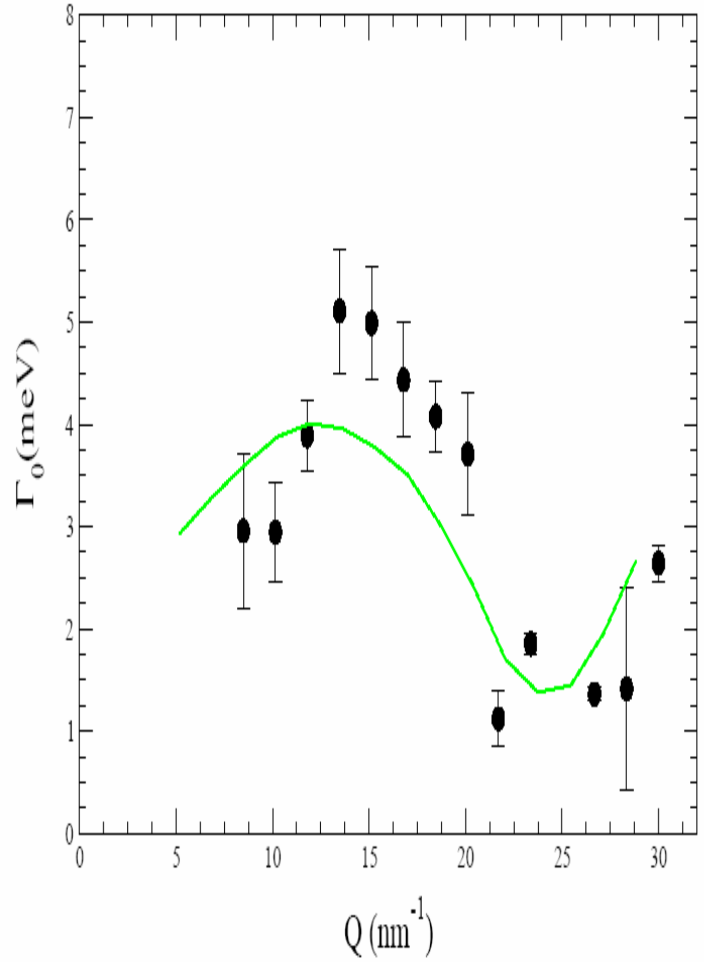
$$\Omega^2=\frac{Q^2\,k_B T}{S(Q)}$$

MCT is self-consistent scheme for S(Q,ω)
very successful for relaxation at glass transition
Computing time approx. 2 hours on PC
(→ W. Schirmacher, ATI)



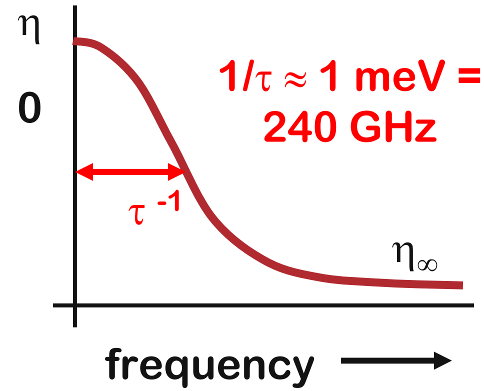
Liquid Titanium at 1750 º C





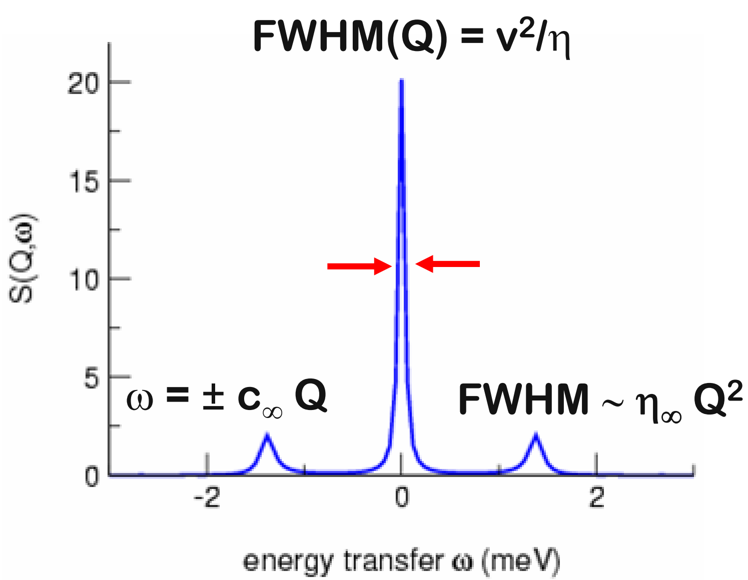
Said et al. PRB, 74, 172202 (2006)

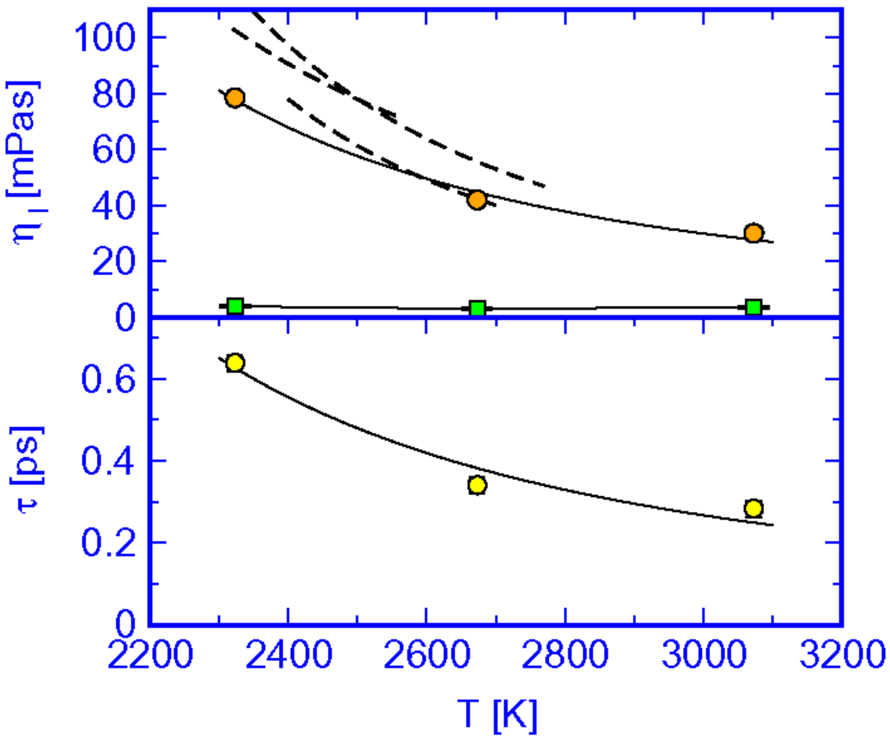
Viscoelasticity in Liquid Alumina



Frequency dependent viscosity

$$\eta(\omega) = \frac{\eta_0}{1 + i\omega\tau} + \eta_\infty$$





H. Sinn et al.: Science, 299: 2047, 2003