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

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Outline

- Motivation
- IXS spectrometer
- Liquids
- Application
  - Liquids under ambient pressure
  - Liquid indium under pressure
Motivation – Understanding Interior of Planets

Earth

Jupiter

Density?
Sound velocity?
Viscosity?
Motivation – Sound velocity & Viscosity

Macroscopic

\[ v \sim \frac{s}{t} \]

Microscopic: APS

\[ F \sim \eta_s v \]

Various IXS studies on single & poly crystals:

Inelastic X-ray Scattering

X-rays: $E \sim 20 \text{ keV}, \delta E \sim 1 \text{ meV}$

Detector: $\delta E \sim 1 \text{ meV}$

$\theta \sim Q$

Intensity $(Q, \omega) \sim S(Q, \omega) \leftrightarrow$ Density fluctuations $(r, t)$

$\Delta E (\text{meV}) \rightarrow$ sound waves (phonons)

$v \sim \text{meV} / Q$

$\eta \sim$ damping, linewidth

$S(Q, \omega)$: Dynamical Structure Factor
IXS Spectrometer at 3 ID-C

4 analyzers

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) | 4.5x10^9
Beam size (μm^2) | 150x200
Liquid static structure factor $S(Q)$

Static structure factor

$S(Q=0) = \chi\rho \ kT$

Dynamics!

$Q_{\text{max}} \approx \frac{2\pi}{\sigma_{\text{HS}}}$

$Q_{\infty} \approx \frac{2\pi}{\delta\sigma_{\text{HS}}}$

$S(Q \to \infty) = 1$
Liquids under ambient pressure – Levitation method

Laser, 270 Watts

X-rays analyzers, 1 meV

X-rays, 20 keV, 1 meV

Oxygen or argon gas stream
Liquid Al$_2$O$_3$ @ 2050°C

Damping is too low by factor 10 for hydrodynamic interpretation!

However: perfect viscoelastic oscillator

Hydrodynamics to Viscoelastics

Frequency dependent viscosity

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

$\text{Alumina : H. Sinn et al., Science, 299: 2047, 2003}$
$\text{Titanium: A.H. Said et al., PRB, 74, 172202 (2006)}$
Liquid Indium under Pressure

Cell externally heated

Liquid Indium

S(Q) of Indium at 2 GPa

Argonne National Laboratory
Sound Velocity of Liquid Indium

Liquid Indium Dispersions
for 1.7, 3.0 and 4.0 GPa

Energy transfer (meV)

Momentum Transfer (nm⁻¹)

$c_{1.7\text{GPa-240C}} = 2625 +/- 60 \text{ m/sec}$
$c_{3.0\text{GPa-300C}} = 2610 +/- 60 \text{ m/sec}$
$c_{4.0\text{GPa-360C}} = 2895 +/- 60 \text{ m/sec}$
Viscosity from IXS
Liquids in the Hydrodynamic region - \( S(Q\sim 0) : 3 \) Lorentzian Peaks

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

\[
\Gamma_0 = \frac{\kappa}{\rho mc_p} Q^2
\]

Rayleigh-peak

Brillouin doublet:

\[
\Gamma_s = \frac{1}{2 \rho m} \left[ 4 \eta_s + \eta_B + \frac{A_0}{2 A_s} \frac{\kappa}{c_p} \right] Q^2
\]

\[
\omega_s = \pm c_s Q
\]

Landau-Placzeck ratio

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

- **Collimator between cell and detector slit**
  - 2mm detector slit 15 cm from cell
  - Allows whole analyzer to be illuminated

- **Scattering volume**
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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)**
- **ρ**

Self consistent Mode Coupling Approximation

- **F(Q, t)**
- **S(Q, ω)**

Fourier Transform

- *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 °C

Said et al. PRB, 74, 172202 (2006)
Viscoelasticity in Liquid Alumina

Frequency dependent viscosity

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

FWHM(Q) = \(\nu^2/\eta\)

\[ \omega = \pm c_\infty Q \]

FWHM \sim \eta_\infty Q^2

\[ 1/\tau \approx 1 \text{ meV} = 240 \text{ GHz} \]