Time-resolved characterization of supercooled liquids using a containerless sample environment and stroboscopic data acquisition

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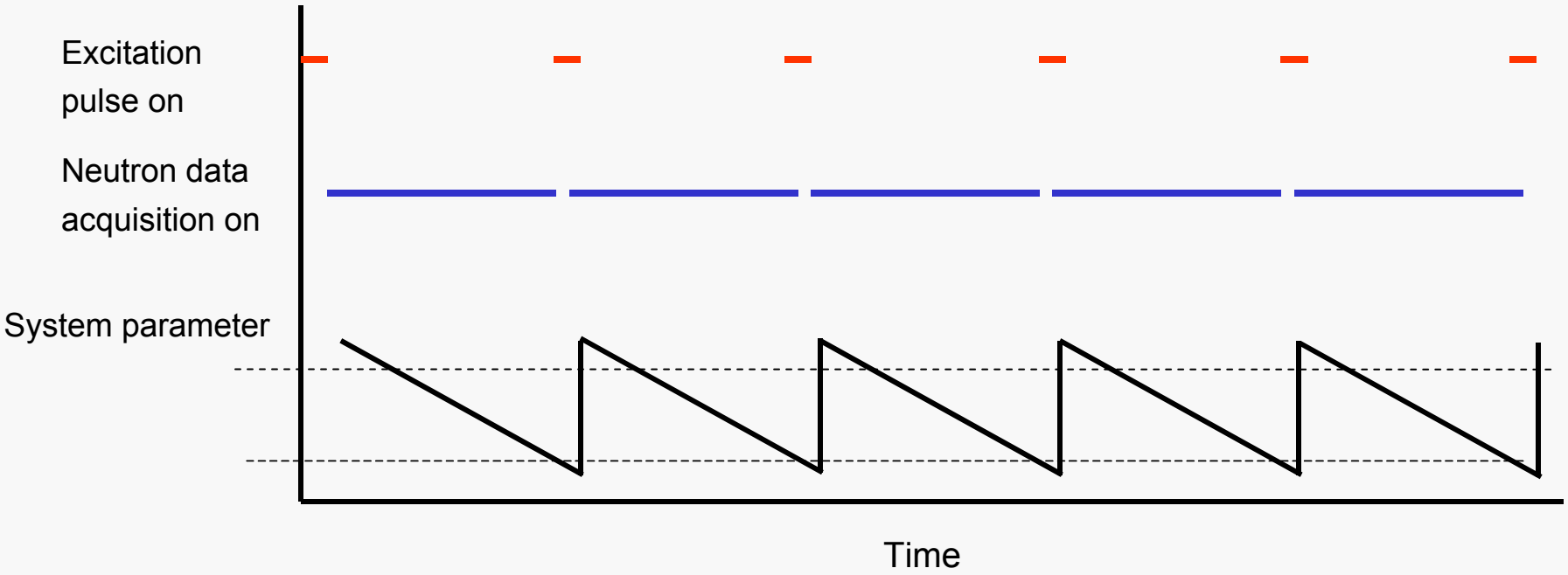
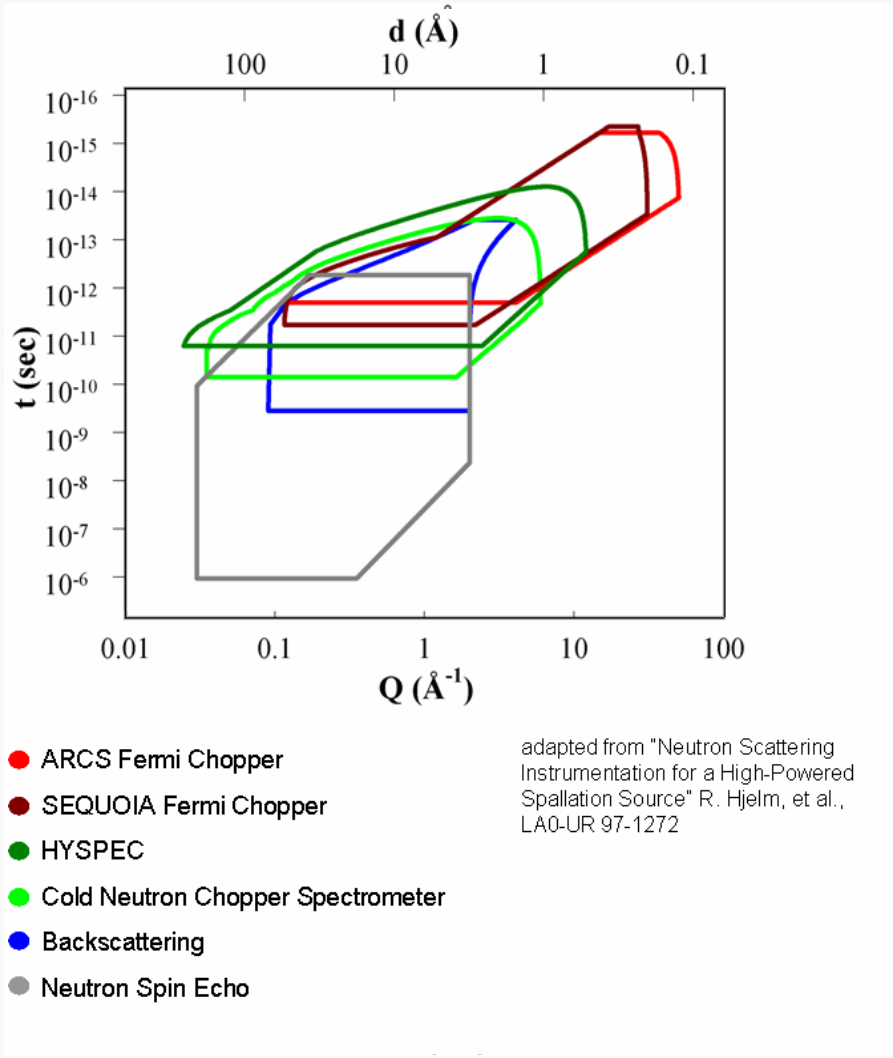
Stroboscopic (pump-probe) measurements

- Process should be reproducible and reversible.
- Pump can be thermal, mechanical, magnetic, electrical, photonic, or combinations.



Stroboscopic experiments add six orders of magnitude to neutron time-scale

- Stroboscopic data-acquisition can in principle be implemented at any neutron diffractometer
- Shortest time scale is in the order of 10^{-5}s, longest time-scale in principle unlimited

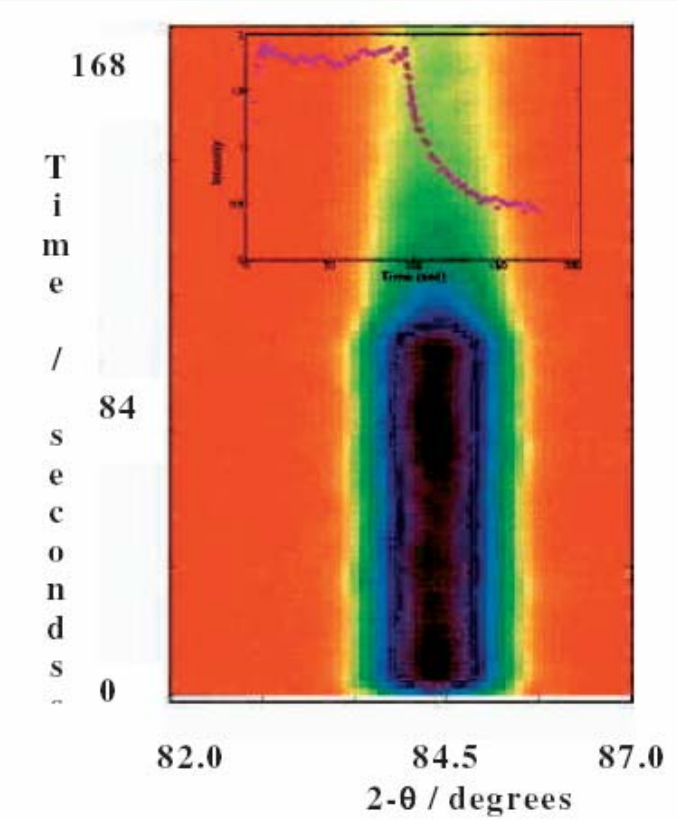


Scientific Opportunities

- Phase transitions and nucleation, approach to the glass transition
- Catalysis - Intermediates in reversible reactions
- Photochemically induced reactions
- Structure changes induce by mechanical stresses, electric or magnetic fields
- Transient high p,T conditions
- Negative pressures (System metastable with respect to cavitation)

Stroboscopic measurements: Recent studies

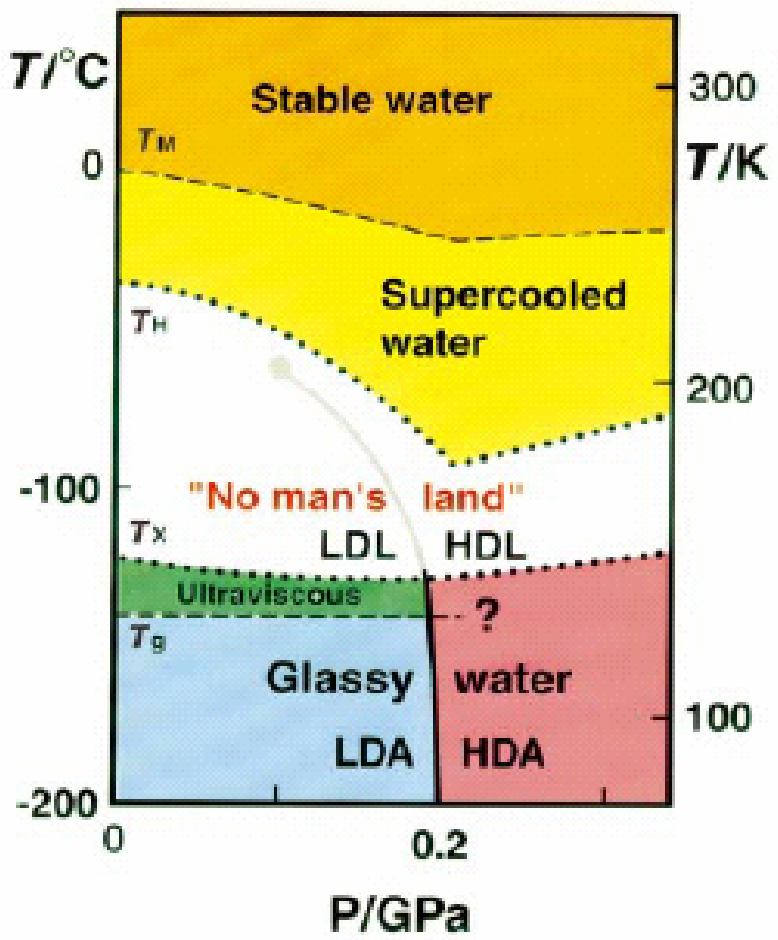
- A handful experiments at ILL and ISIS
- Scientific problems: Phase transitions, ordering induced by flow or electric fields



Diffraction from a dispersion of $Ni(OH)_2$ particles in D_2O showing the change in intensity of the 004 peak in one direction when flow starts. Each time slice is two seconds [16].

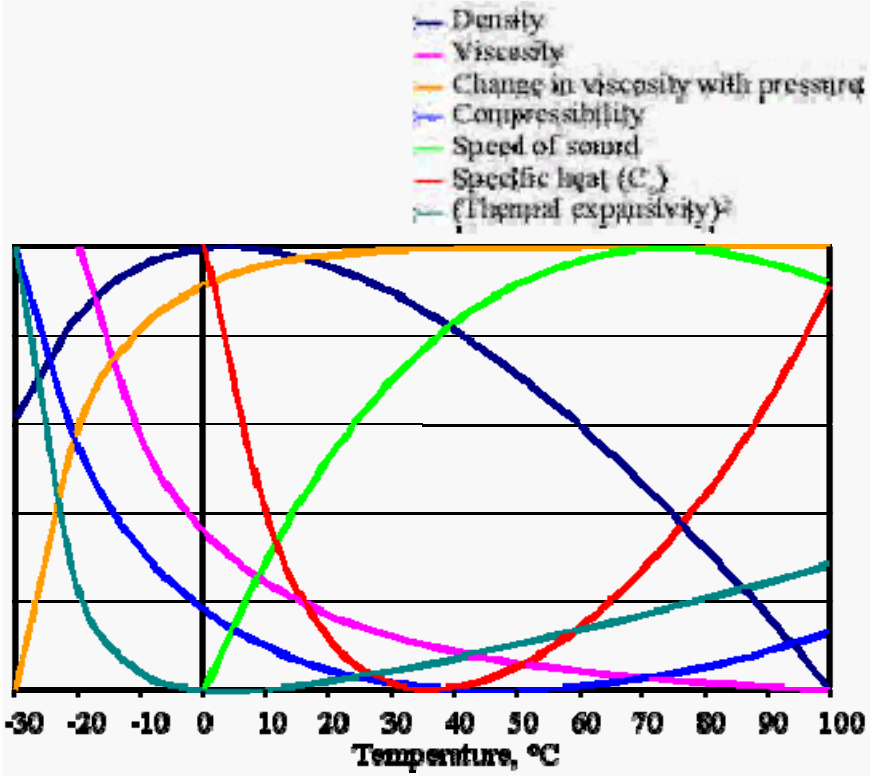
A. B. D. Brown, S. M. Clarke, A. R. Rennie, P Convert, and T. Hansen, Annual ILL Report (1997)

Supercooled water



H. E. Stanley et al. *PCCP*, **2**, 1551 (2000)

The forty-one Anomalies of Water[1]

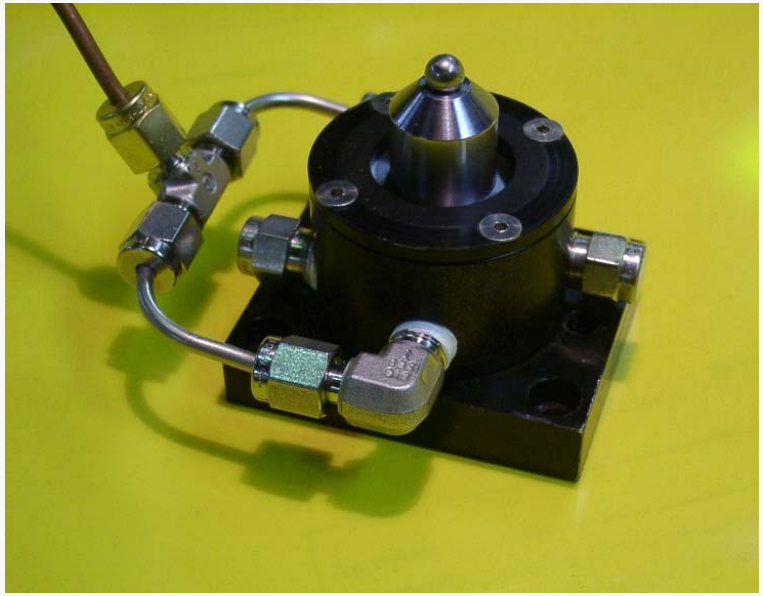


[1] http://www.lsbu.ac.uk/water

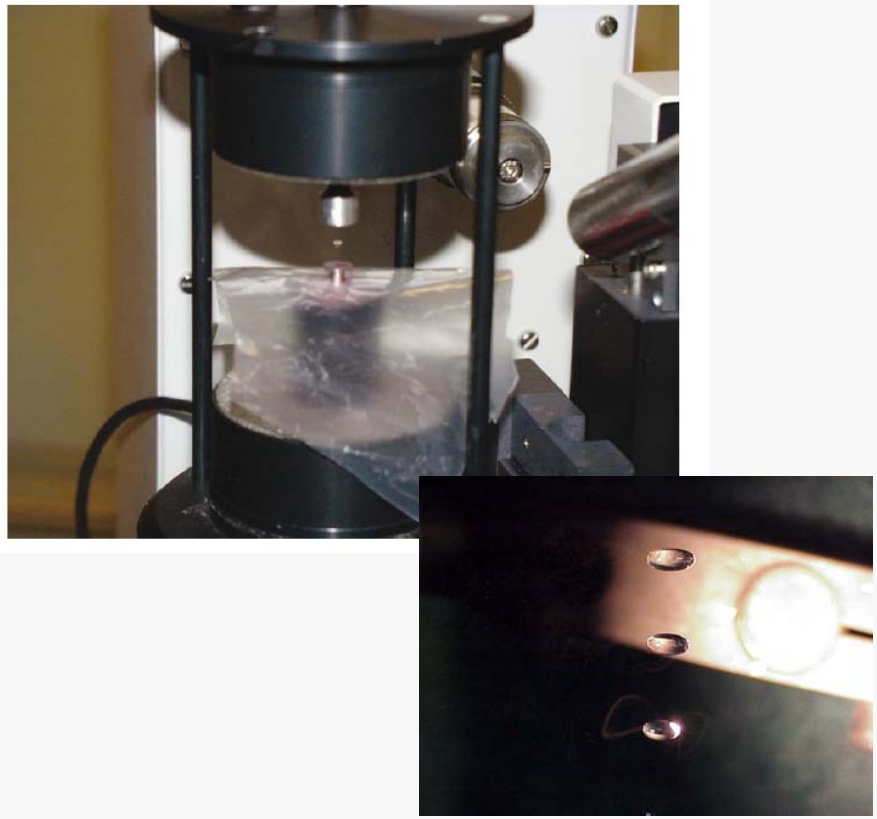
Containerless sample environment

- Sample “floats” for example on a gas stream
- No contact with a container wall → no heterogeneous nucleation sites
- Longer lifetime of supercooled state
- So far, mainly used for high temperature studies (e.g. refractory ceramics)

Tasks: Aerodynamic vs. acoustic levitation

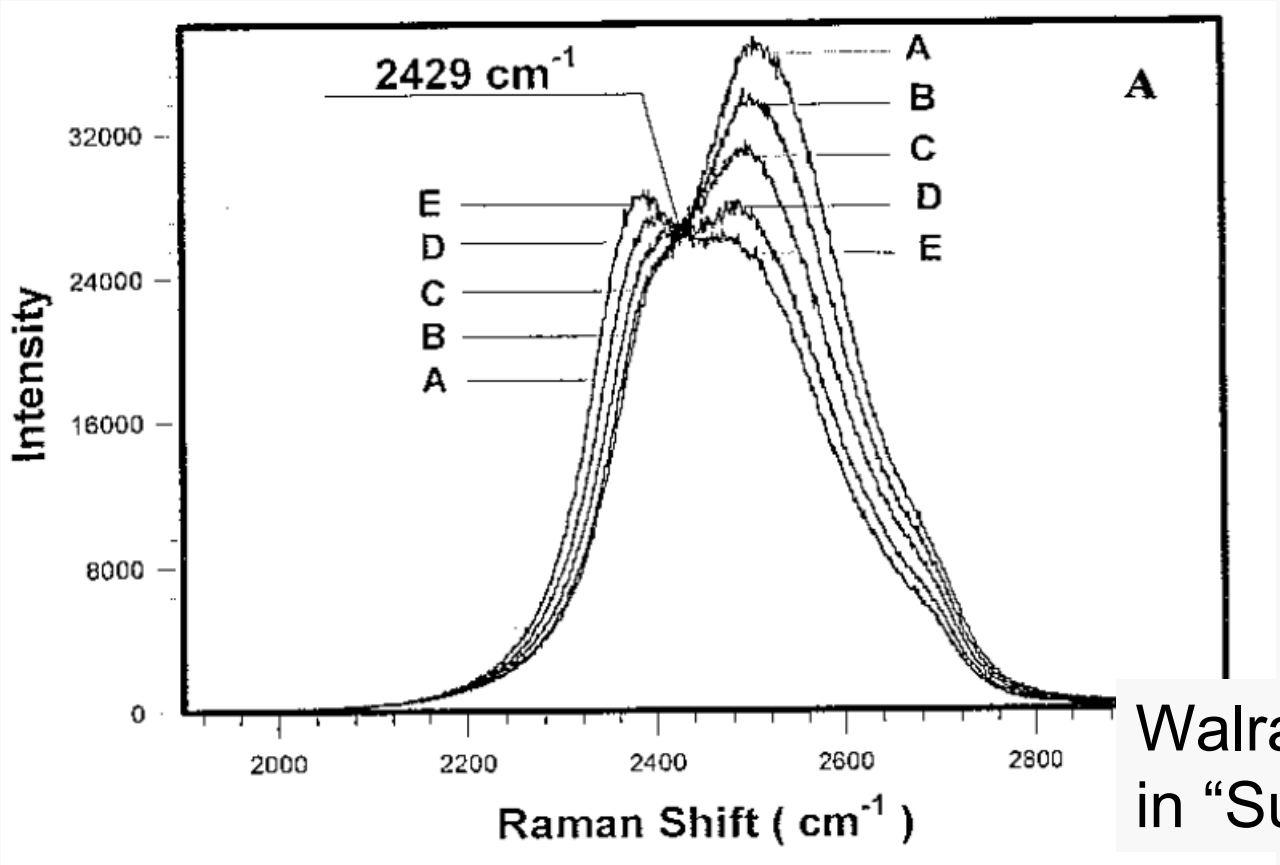


The gas stream levitator nozzle of a levitation system used at the Glass Liquids and Amorphous Diffractometer at the Intense Pulsed Neutron Source at Argonne National Laboratory



Acoustic levitator with floating water droplet [Y. Cerenius, Å. Oskarsson, S. Santeson, L. Nilsson, L. Kloo, J. Appl. Cryst. **36** (2003) 163]

Temperature measurement



Walrafen et al.
in “Supercooled liquids”

Use of spectroscopic properties to determine the temperature?

Static and dynamic measurements

- Due to the absence of heterogeneous nucleation sites levitated supercooled liquids are fairly stable so that at higher temperatures static measurements become possible
- Static supercooling experiments will provide an important test that the results obtained by dynamic supercooling are representative.
  - Perform static experiment
  - Compare with dynamic result
  - Extended temperature range with dynamic experiment