

Workshop on Topics in Soft Condensed Matter
November 29, 2015, MIT, Cambridge, MA

ABSTRACT BOOKLET

Workshop Agenda

November 29, 2015

Room 24-213, Nuclear Science and Engineering Department, MIT

9.00 am *Preparation*

9.30 am *Sow Hsin Chen - Early days*

J. Michael Rowe (University of Maryland and National Institute of Standards and Technology)

10.00 am *Retrospective of a Long and Distinguished Career*

Sidney Yip (Massachusetts Institute of Technology)

10.30 am *Coffe Break*

11.00 am *Experimental Tests of the Liquid-Liquid Phase Transition Hypothesis*

H. Eugene Stanley (Boston University and Boston University School of Medicine)

11.30 am *Atomistic Origin of Viscosity and Shear Thinning*

Takeshi Egami (University of Tennessee and Oak Ridge National Laboratory)

12.00 pm *Lunch (Courtesy of Prof. Ju Li, Nuclear Science and Engineering Department, MIT)*

1.00 pm *The Boson Peak in Confined Water*

Francesco Mallamace (Università di Messina, Italy)

1.30 pm *25 Years of Science with Sow-Hsin*

Piero Baglioni (Università di Firenze, Italy)

2.00 pm *Microscopic Dynamics of Hydration Water in Aqueous Solutions of Lysozyme and Trehalose upon Cooling*

Paola Gallo (Università Roma Tre, Italy)

2.20 pm *Coffe Break*

2.50 pm *How much time does it take to measure slow relaxations in water?*

Eugene Mamontov (Oak Ridge National Laboratory)

3.10 pm *Neutron spectroscopy of ultra-confined water*

Alexander I. Kolesnikov (Oak Ridge National Laboratory)

3.30 pm *Structure and phase transitions in metallic glasses*

Xun-Li Wang (City University of Hong Kong)

7.00 pm **Chinese Banquet** (The organizers thank the Nuclear Science and Engineering Department, MIT for hosting this event)

Joyful Garden (1234 Soldiers Field Road, Brighton, MA 02135)

Sow Hsin Chen – Early days

J. Michael Rowe

University of Maryland and National Institute of Standards and Technology

In celebration of Sow Hsin's long and distinguished career and his many contributions to science and technology, I will present a somewhat whimsical but brief description of his early days as a graduate student at McMaster University as seen by a very new graduate student with no training beyond undergrad. Although we were all formally grad students, he was more of a postdoctoral research assistant and mentor to Eric Svensson and me. I will also, again briefly, discuss our continued interactions over the next 50+ years.

Retrospective of a Long and Distinguished Career

Sidney Yip

Massachusetts Institute of Technology

Some recollections and observations from a colleague who has been with you throughout your journey, from early collaborations in neutron scattering to sharing recent ideas about the glass transition.

Experimental Tests of the Liquid-Liquid Phase Transition Hypothesis

H. Eugene Stanley

Boston University and Boston University School of Medicine

We will introduce some of the 69 documented anomalies of the most complex of liquids, water—focusing on recent progress in understanding these anomalies by combining information provided by recent spectroscopy experiments and simulations on water in bulk, nanoconfined and biological environments. We will interpret evidence from recent experiments designed to test the hypothesis that liquid water has behavior consistent with the hypothesized “liquid polymorphism” in that water under appropriate conditions could exist in two different phases. We will also discuss recent work on nanoconfined water anomalies as well as the apparently related, and highly unusual, behavior of water in biological environments. Finally, we will discuss how the general concept of liquid polymorphism is proving useful in understanding anomalies in other liquids, such as silicon, silica, and carbon, as well as metallic glasses, which have in common that they are characterized by two characteristic length scales in their interactions.

Atomistic Origin of Viscosity and Shear Thinning

Takeshi Egami

University of Tennessee and Oak Ridge National Laboratory

The theory of liquids usually starts with the Langevin equation of motion assuming local force and random thermal excitation. In this mean-field picture each atom basically sees the same environment except for statistical fluctuations. However, the results of recent molecular dynamics study suggest the importance of *intermediate-scale* dynamics which cannot be captured by the conventional mean-field approach. The equivalence of stress and temperature in liquid flow results in a simple scaling relationship between stress and temperature [1], leading to a formal explanation of shear thinning. The study of local dynamic atomic correlation during the steady state flow demonstrated the presence of local elastic correlation [2]. The spatial extent of elastic correlation depends on shear rate, resulting in the microscopic explanation of shear thinning. In high-temperature metallic liquids the elementary excitations are local topological excitations in the atomic connectivity network, rather than phonons, and they determine viscosity [3]. In supercooled liquids and in glasses collective excitations in local topology govern the mechanical behavior [4]. These observations suggest, instead of the mean-field approach, we should choose a new approach based on topological description of atomic connectivity and its dynamics [5].

The Boson Peak in Confined Water

Francesco Mallamace

Università di Messina

The Boson peak (BP) of deeply cooled confined water is studied by using inelastic neutron scattering (INS) in a large interval of the (P, T) phase plane. By taking into account the different behavior of such a collective vibrational mode in both strong and fragile glasses as well as in glass-forming materials, we were able to determine the Widom line that characterizes supercooled bulk water within the frame of the liquid-liquid phase transition (LLPT) hypothesis. The peak frequency and width of the BP correlated with the water polymorphism of the LLPT scenario, allowing us to distinguish the “low-density liquid” (LDL) and “high-density liquid” (HDL) phases in deeply cooled bulk water. Moreover, the BP properties afford a further confirmation of the Widom line temperature T_w as the (P, T) locus in which the local structure of water transforms from a predominately LDL form to a predominately HDL form.

References

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- [2] Iwashita, T. and Egami, T. Phys. Rev. Lett. **2012**, 108, 196001.
- [3] Iwashita, T., Nicholson, D. M. and Egami, T. Phys. Rev. Lett. **2013**, 110, 205504.
- [4] Fan, Y., Iwashita, T. and Egami, T. Nature Commun. **2014**, 5, 5083.
- [5] Egami, T. Mod. Phys. Lett. B **2014**, 28, 1430006 (2014).

25 Years of Science with Sow-Hsin

Piero Baglioni

Università di Firenze, Italy

I will review twenty-five years of scientific interaction with Sow-Hsin, from the early days to more recent advances.

Microscopic Dynamics of Hydration Water in aqueous solutions of Lysozyme and Trehalose upon Cooling

Paola Gallo

Università Roma Tre, Italy

Aqueous solutions of carbohydrates have an extraordinary cryopreservation ability and among them trehalose is particularly effective [1]. I will present results from molecular dynamics simulations of two aqueous solutions, water and lysozyme, and water, lysozyme and trehalose. The dynamics of the system is studied upon cooling. In particular relaxation times are analyzed and a comparison between the two system is done. Water in the ternary system is slowed down, especially around the lysozyme and this is due to the influence of trehalose [2].

I show here a detailed study of hydration water around lysozyme [3]. Hydration water shows two distinct slow relaxations in both solutions. One is the alpha relaxation typical of glass formers, the longer one is specific of hydration water. Two crossovers are found, one is the fragile to strong crossover already detected in hydration water [4], the other is related to the enhancement of the protein mean square displacement.

When trehalose is added to the aqueous solution the relaxation times of hydration water molecules become substantially higher especially at lower temperatures and especially for the longer relaxation time. These results point to a cryoprotective action exerted by packing slow water between the protein and a layer of trehalose molecules.

References

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- [2] Corradini, D., Strekalova, E.G., Stanley H.E. and Gallo, P. Sci. Rep. **2013**,3, 1218.
- [3] Camisasca G., Corradini D., De Marzio M. and Gallo P., in preparation.
- [4] Chen, S. -H., Liu, L., Fratini, E., Baglioni, P., Faraone, A., Mamontov, E., P. Natl. Acad. Sci. USA **2006**, 24, 9012; Mallamace, F., Chen, S.-H., Broccio, M., Corsaro, C., Crupi, V., Majolino, D., Venuti, V., Baglioni, P., Fratini, E., Vannucci, C. and Stanley, H.E. J. Chem. Phys. **2007**, 127, 045104; Lagi, M., Chu, X.-Q., Kim, C., Mallamace, F., Baglioni, P. and Chen, S.-H. J. Phys. Chem. B **2008**, 112, 1571

How much time does it take to measure slow relaxations in water?

Eugene Mamontov

Oak Ridge National Laboratory

“Unique neutron scattering strength of the signal dependence on the scattering momentum transfer can be applied to probe the characteristic length scale of the dynamic entities associated with slow dynamics in liquids, which at present cannot be studied by other experimental techniques.” Statements of this kind are abundant in the scientific literature, and, unfortunately, there is some truth in them. Regrettably, in the absence of viable experimental technique alternatives, this implies stiff competition for a limited resource (beam time at neutron scattering facilities), which may render demanding experiments impossible to accommodate. We describe our attempts, which spanned several years, to measure slow relaxation dynamics in an aqueous system using incoherent neutron spin-echo. In the end, the data has been measured, analyzed, and published, even though it took several years. The characteristic length scales associated with slow relaxations in liquids can indeed be probed in this manner. The approach that we adopted has proved itself, but, for the future measurements, how can we do better?

Neutron spectroscopy of ultra-confined water

Alexander I. Kolesnikov

Oak Ridge National Laboratory

Water confined within microporous silicate minerals hemimorphite and beryl, presents an extreme case of confinement, where the restricted, hydrogen-bonded (HB) and non-HB water molecules are situated in channels whose diameter is not much larger than the water molecule itself. Inelastic, quasi-elastic and deep inelastic neutron scattering (QENS, INS and DINS, respectively) were used to studies the dynamics of the confined water.

The QENS experimental data for hemimorphite provide clear evidence that the confined water molecules undergo local diffusive motion with characteristic times ranging from less than picosecond to hundreds of picoseconds even when the mineral is cooled to cryogenic temperatures. In particular, the spectra exhibit two thermally activated relaxation processes, a faster one on the subpicosecond time scale and a slower one on the 10-100 ps time scale at temperatures between 70-130 K, at which other hydrogen bonded confined water is usually immobile. The origin of the high mobility of the confined water will be discussed in relation to planar configuration of HB-network in hemimorphite.

The INS study of water in beryl reveals a new “quantum tunneling state” of the water molecule. A number of peaks were observed in the INS spectra which were uniquely assigned to water quantum tunneling. In addition, the water proton momentum distribution measured with DINS directly showed coherent delocalization of the water protons in the ground state. *The average kinetic energy of the water protons, directly-obtained from the experiment, is a measure of their quantum-mechanical zero point motion, and is found to be ~30% less than it is in bulk liquid or solid water, in complete disagreement with accepted models based on the energies of its vibrational modes.* A combination of *ab initio* DFT calculations and path integral MD simulations supports the experimental data and show large delocalization of water protons and electrons across the channel of mineral.

Structure and phase transitions in metallic glasses

Xun-Li Wang

City University of Hong Kong

I first met Professor Sow-Hsin Chen in the late 1980s at the Intense Pulsed Neutron Source, Argonne National Laboratory, where I spent two years working on my PhD thesis project. At the time, I was working on magnetism in high-T_c superconductors, and Professor Chen was an authority on water. My research and career have evolved since then. Although Professor Chen and I never had a joint publication, my interactions with him through the years have left an indelible mark in my neutron scattering career, e.g., in my current research on liquid-to-liquid phase transition in metallic liquids. In this talk, I will use my research on metallic glasses and metallic liquids to recall my interactions with Professor Chen and his influence in my career development.

7.00 pm *Chinese Banquet*

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