Structure and Dynamics of Glassy, Supercooled and Nanoconfined Fluids

• WORKSHOP 2019 •

Buenos Aires | Argentina | July 14 –16

# Program
# &
# Book of Abstracts

**Centro Atómico Constituyentes**

**Comisión Nacional de Energía Atómica**

**- Julio 2019 –**

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# PROGRAM

## Sunday 14th July

Dazzler San Martin, San Martin 920 – Buenos Aires City

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|---|---|
| **12:30 - 13:30** | Registration |
| **13:30 - 13:45** | Welcome |
| SESSION 1 | Chairwoman: Paola Gallo |
| **13:45 - 14:30** | **Pablo Debenedetti**<br>*The phase behavior of supercooled water: a computational perspective* |
| **14:30 - 15:05** | **Thomas Loerting**<br>*Deeply supercooled liquid water below the crystalization line?* |
| **15:05 - 15:30** | **Shankar Das**<br>*Spontaneous breaking of ergodicity and configurational entropy of a supercooled liquid* |
| **15:30 – 16:00** | **Posters presentation (1-6)** |
| **16:00 - 16:30** | Coffee Break |
| SESSION 2 | Chairman: Nicolas Giovambattista |
| **16:30 - 17:15** | **Walter Kob**<br>*On the structure of liquids: More order than expected* |
| **17:15 - 17:50** | **Paola Gallo**<br>*Slow dynamics of supercooled hydration water in biosolutions* |
| **17:50 - 18:15** | **Katrin Amann-Winkel**<br>*Amorphous ices and liquid states of water* |
| **18:15 – 18:45** | **Posters presentation (7-14)** |
| **18:45 - 20:00** | Welcome Cocktail |

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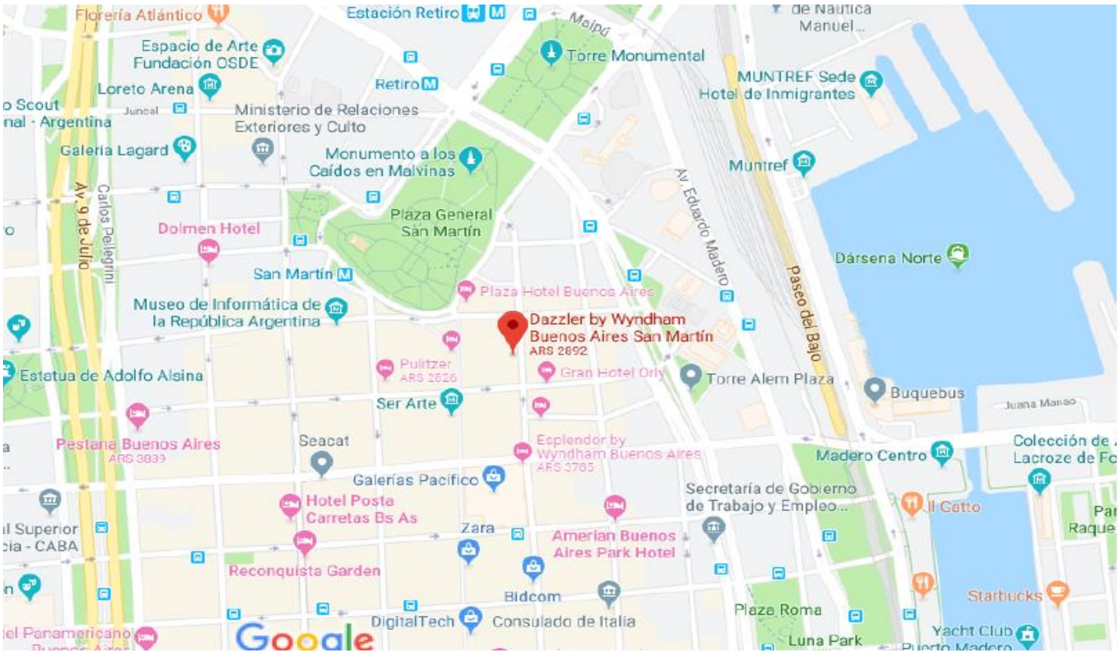

## Monday 15th July

Centro Atómico Constituyentes (CNEA), Avenida General Paz 1650, San Martin, Buenos Aires

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|---|---|
| SESSION 3 | Chairman: Pablo Debenedetti / Horacio Corti |
| **08:30 - 09:15** | **Juan de Pablo**<br>*Liquid Crystals - From simple self assembled constructs to autonomous materials* |
| **09:15 – 09:50** | **Juan P. Garrahan**<br>*Trajectory methods in systems with slow dynamics* |
| **09:50 - 10:15** | **Tsvi Tlusty**<br>*Proteins as evolvable amorphous matter - a nano-rheological view* |
| **10:15 - 10:30** | **Federico Suarez**<br>*Recent progress in the development of the Argentine Neutron Beam Laboratory (LAHN)* |
| **10:30 - 11:00** | Coffee Break |
| SESSION 4 | Chairwoman: Marcia Barbosa |
| **11:00 - 11:35** | **Nicolás Giovambattista**<br>*Water-mediated interactions at the nanoscale* |
| **11:35 - 12:00** | **Mario Del Popolo**<br>*Gas capture properties and mesoscopic organization of nanoporous materials in the liquid phase* |
| **12:00 - 12:25** | **Horacio Corti**<br>*Mass and charge transport under confinement: experimental results* |
| **12:25 - 14:00** | Lunch |
| SESSION 5 | Chairman: Mario Del Popolo |
| **14:00 - 15:00** | **Igal Szleifer**<br>*Complex water and polymer effects in nano-confined systems* |
| **15:00 - 15:25** | **Livia Bove**<br>*Anomalous dynamical phenomena in confined and confining water under high pressure* |
| **15:25 - 15:50** | **Damian Scherlis**<br>*Dissociation of water in nanoscale confinement* |
| **15:50 - 16:20** | Coffee Break |
| SESSION 6 | Chairman: Damian Scherlis |
| **16:20 - 16:55** | **Marcia Barbosa**<br>*Diffusion of water confined in deformed nanotubes* |
| **16:55 - 17:20** | **Paula Longinotti**<br>*Phase transitions of water solutions in mesoconfined systems* |
| **17:20 - 18:00** | **Posters discussion** |
| **18:00 - 18:45** | **Oral presentations (1-3)** |

## Tuesday 16th July

Centro Atómico Constituyentes (CNEA), Avenida General Paz 1650, San Martin, Buenos Aires

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|---|---|
| SESSION 7 | Chairman: Gustavo Appignanesi / Anibal Disalvo |
| **08:30 - 09:15** | **Hajime Tanaka**<br>*Microscopic structural origin of the thermodynamics and dynamics anomalies of water* |
| **09:15 - 09:50** | **Giancarlo Franzese**<br>*Structure and dynamics of water at the interface with phospholipid membranes* |
| **09:50 - 10:15** | **Anibal Disalvo**<br>*Correlation between hydration of carbonyl and phosphate groups in lipid membranes and its effect on electrical, compressibility and defect formation* |
| **10:15 - 10:40** | **Mario Tagliazucchi**<br>*Self-Assembly of Peptide-Amphiphiles in Aqueous Solutions* |
| **10:40 - 11:10** | Coffee Break |
| SESSION 8 | Chairman: Juan Garrahan |
| **11:10 - 11:45** | **Valeria Molinero**<br>*Ice nucleation without supercooling* |
| **11:45 - 12:10** | **Gustavo Appignanesi**<br>*Liquid water: A two-state scenario with "three" molecular classes?* |
| **12:10 - 12:35** | **Yanina Fasano**<br>*Kinetic arrest of vortex-liquid transitions in vortex matter induced by disorder* |
| **12:35 - 14:00** | Lunch & Poster discussion |
| SESSION 9 | Chairwoman: Valeria Molinero |
| **14:00 -14:45** | **Peter Poole**<br>*Phase transitions in fluctuations and their role in two-step nucleation* |
| **14:45 - 15:10** | **Limei Xu**<br>*Liquid-liquid phase transition in glass-forming liquids* |
| **15:10 - 15:55** | **Oral presentations (4-6)** |
| **15:55 - 16:15** | Closure |

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# PLENARY LECTURES

**Pablo G. Debenedetti**

Department of Chemical and Biological Engineering, Princeton University, USA

## *The phase behavior of supercooled water: a computational perspective*

Water affects every aspect of our lives, from agriculture to climate, and from health to geopolitics. It is a key participant in the physical and chemical processes that sustain life as we know it. Its ubiquity and importance notwithstanding, there remain major open questions about water's physical properties, which are anomalous by comparison to those of most other liquids. Examples include the fact that the liquid, if sufficiently cold, expands when cooled and becomes less viscous when compressed. Water's oddities become more pronounced at low temperatures, especially in the supercooled regime, where the liquid is metastable with respect to crystallization.

I will review some the hypotheses that have been proposed to explain experimental observations. Computer simulations have played an important role in research on supercooled water, in large part because they are not subject to the limitations that make experimental probing of deeply metastable states so challenging. I will illustrate the advantages and limitations of computational investigations of supercooled water, focusing on the intriguing possibility of the existence of a liquid-liquid phase transition. The recent resolution of a long-standing controversy on this topic has interesting scientific implications regarding the notion of metastable criticality, as well as important lessons for the practice of science.

**Walter Kob**

University of Montpellier. FRANCE

## *On the structure of liquids: More order than expected*

The structure of liquids and glasses is usually characterized by means of the radial distribution function or the static structure factor. Computer simulations or confocal microscopy experiments on colloidal systems allow also to access the bond angle distributions or the local connectivity of the atoms. However, all these quantities are basically one-dimensional in nature and hence it is hard to infer from them the real three dimensional structure of amorphous systems. As a consequence the structure of liquids and glasses is usually considered to be boring for distances beyond the second/third nearest neighbor. In this talk I will show that this is not the case at all and that by considering simple three dimensional correlation functions one finds a surprisingly ordered arrangement of the particles even at significantly larger distances. This order grows quite quickly if the temperature is lowered, showing that amorphous systems are way more ordered than expected from the study of the usual two-point correlation functions.

**Juan de Pablo**

Pritzker School of Chemical Engineering, University of Chicago, USA

## *Liquid Crystals - From simple self assembled constructs to autonomous materials*

Polymeric materials comprising mechano-chemically active components are able to undergo spontaneous structural rearrangements that generate internal stresses and motion. These stresses can be particularly large in the case of liquid crystalline polymers, where elasticity plays an important role on the structure of the underlying materials. Understanding how internal activity leads to specific behaviors is important for design of autonomous materials systems capable of delivering desired functionalities. This lecture will focus on the relationship between structure, activity, and motion in lyotropic liquid crystalline polymeric systems. More specifically, results will be presented for actin and tubulin suspensions, where activity is generated by protein motors. A distinctive feature of these biopolymers is that characteristic contour lengths can range from hundreds of nanometers to tens of microns, thereby making them amenable for study by optical microscopy. By relying on molecular and meso-scale models, it is possible to arrive at a comprehensive description of these suspensions that helps explain the connections between molecular structure, the formation and shape of distinct topological defects, activity, and defect dynamics. One of the outcomes of such a description is the realization that hydrodynamic interactions can in some cases exacerbate or mitigate the elasticity of the underlying materials, leading to non-intuitive phenomena that do not arise at equilibrium. By balancing such effects, these findings raise the possibility of designing functional materials where specific, macroscopic dynamical responses are engineered into a system to create function.

**Igal Szleifer**

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Mario Tagliazucchi
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Yitzhak Rabin
Department of Physics, Bar-Ilan University, ISRAEL

## *Complex water and polymer effects in nano-confined systems*

Understanding polymer behavior in nano-confined aqueous environment is of great importance for biomedical applications. For example, polymer-coated nanopores in solution can serve as molecular filters that select the passage of molecules based on their water affinity and charge. In this presentation, I will demonstrate the idea of using sequence-designed polymers to construct a smart nano-gate that can respond to external stimuli such as pH and salt concentration. To improve the efficiency of our artificial nano-filter, we seek inspiration from the largest biological channel, i.e., the nuclear pore complex (NPC). The NPC employs intrinsically disordered proteins (IDPs) as its gating materials. Termed FG-Nups, these IDPs carry hydrophobic FG repeats that can bind to molecular cargoes. It has been a long-standing mystery how FG-Nups enable rapid yet selective transport of biomass between cytoplasm and nucleus, due to the experimental difficulty of characterizing the NPC lumen. To address this problem, we have built a computational microscope to visualize the functional structure of FG-Nups. Our model shows that the complex interplay between water and polymer effects, including hydrophobic interaction, specific spacer cohesion, electrostatics, volume exclusion, and conformational entropy, leads to a remarkably elaborate gating structure that features nano-condensates and mosaic FG-territories. These molecular insights can help understand other IDP-involved biological compartmentalization and design better artificial materials based on amphiphilic polymers.

**Hajime Tanaka**

Rui Shi
Department of Fundamental Engineering, Institute of Industrial Science,
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## *Microscopic structural origin of the thermodynamics and dynamics anomalies of water*

Water displays a vast array of unique properties, known as water's anomalies, whose origin remains subject to active debate [1]. With computer simulations of popular water models, we provide a unified picture of water's anomalies in terms of locally favored tetrahedral structures [2], which are characterized by high local tetrahedral and translational symmetries stabilized by four hydrogen bonds. We reveal that such local structural order identified microscopically [3] has different impacts on the thermodynamic and dynamic properties of water [4,5]: The former is controlled directly by the structural order parameter itself, whereas the latter is by the one spatially coarse-grained up to nearest neighbors, reflecting the fact that the dynamics of a molecule crucially depends on its nearest-neighbor environment [5,6]. Based on this, we develop a new hierarchical two-state model. We show that these two structural order parameters intimately linked only via spatial coarse-graining, quantitatively account, respectively, for (i) thermodynamic anomalies and (ii) dynamic anomalies including dynamic slowing down upon cooling, diffusion maximum under compression, “fragile-to-strong” transition, dynamic heterogeneities, and breakdown of the Stokes-Einstein-Debye relation. Our result suggests that the fragile-strong transition is not associated with glass-transition phenomena, but just a consequence of a crossover between the two states. We also show for silica, which also exhibits water-type anomalies, that there is a direct structural signature of the formation of locally favored tetrahedral structure in the scattering function [7]; more specifically, we find that the so-called first sharp diffraction peak (FSDP) originates from density waves characteristic of a tetrahedral unit and its intensity is proportional to the fraction of locally favored tetrahedral structures. We uncover FSDP also in the scattering function of liquid water [8]. This provides strong evidence for the presence of two states, which is experimentally accessible directly. We hope that this will lead to the convergence of the long debate on which of mixture and continuum models is relevant.

**References:**

1. P. Gallo, et al., Chem. Rev. 116, 7463–7500 (2016).
2. H. Tanaka, Eur. Phys. J E, 35, 113 (2012); Phys. Rev. Lett. 80, 5750-5753 (1998); J. Chem. Phys. 112, 799–809 (2000); EPL 50, 340–346 (2000); J. Phys.: Condens. Matter 15, L703–L711 (2003).
3. J. Russo, & H. Tanaka, Nat. Commun. 5, 3556 (2014).
4. J. Russo, & H. Tanaka, Proc. Natl. Acad. Sci. U. S. A. 112, 6920–6924 (2015).
5. R. Shi, J. Russo and H. Tanaka, Proc. Natl. Acad. Sci. USA 115, 9444-9449 (2018).
6. R. Shi, J. Russo and H. Tanaka, J. Chem. Phys. 149, 224502 (2018).
7. R. Shi and H. Tanaka, Sci. Adv. 5, eaav3194 (2019).
8. R. Shi and H. Tanaka, unpublished.

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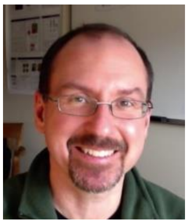

## *Phase transitions in fluctuations and their role in two-step nucleation*

We consider the thermodynamic behavior of local fluctuations occurring in a stable or metastable bulk phase. For a system with three or more phases, we present a simple analysis based on classical nucleation theory that predicts thermodynamic conditions at which small fluctuations resemble the phase having the lowest surface tension with the surrounding bulk phase, even if this phase does not have a lower chemical potential. We also identify the conditions at which a fluctuation may convert to a different phase as its size increases, referred to here as a “fluctuation phase transition” (FPT). We demonstrate these phenomena in simulations of a two dimensional lattice model by evaluating the free energy surface that describes the thermodynamic properties of a fluctuation as a function of its size and phase composition. We show that a FPT can occur in the fluctuations of either a stable or metastable bulk phase and that the transition is first-order. We also find that the FPT is bracketed by well-defined spinodals, which place limits on the size of fluctuations of distinct phases. Furthermore, when the FPT occurs in a metastable bulk phase, we show that the superposition of the FPT on the nucleation process results in two-step nucleation (TSN). We identify distinct regimes of TSN based on the nucleation pathway in the free energy surface and correlate these regimes to the phase diagram of the bulk system. Our results clarify the origin of TSN and elucidate a wide variety of phenomena associated with TSN, including the Ostwald step rule.

**References:**

D. James, et al., J. Chem. Phys. 150, 074501 (2019)

# SEMIPLENARY LECTURES

**Thomas Loerting**

Institute of Physical Chemistry, University of Innsbruck, AUSTRIA

## *Deeply supercooled liquid water below the crystalization line?*

One of the key questions in understanding water's anomalies is whether or not deeply supercooled bulk liquid water can be attained in experiments and be kept stable on the time scales of minutes or hours. The main challenge in these experiments is the interference of crystallization, which needs to be avoided through suitable choice of pressure and temperature. The nature of water just below the crystallization line is intimately linked with the question of the type of molecular motions activated above the glass transitions found upon heating the amorphous ices, namely 136 ± 2 K in case of low-density amorphous ice (LDA) [1] and 116 ± 3 K in case of high-density amorphous ice (HDA) [2]. In my presentation I am going to detail our recent work on this issue, including work about the pressure-temperature path-independence that is required to make the case for equilibrated liquids [3], the effect of doping and isotope substitution on the dynamics in amorphous ices [4] or the long-range diffusion revealed upon heating highly porous vapor-deposited amorphous water [5]. All of our experiments are consistent with the idea that two distinct types of ultraviscous liquid water, high- (HDL) and low-density liquid (LDL), can be accessed below the crystallization line in the pressure range between ambient pressure and 300 MPa. This implies that our observation in 2011 can be understood as the first experimental realization of the first order liquid-liquid phase transition in bulk water: the spontaneous phase separation and formation of a sharp interface between two non-crystalline phases of low- and high-density that progresses with time at 140 K and 70 MPa [6].

1. I. Kohl, L. Bachmann, E. Mayer, A. Hallbrucker, T. Loerting; Nature 435 (2005) E1; M. S. Elsaesser, K. Winkel, E. Mayer, T. Loerting; Phys. Chem. Chem. Phys. 12 (2010) 708–712.
2. K. Amann-Winkel, C. Gainaru, P. H. Handle, M. Seidl, H. Nelson, R. Böhmer, T. Loerting; Proc. Natl. Acad. Sci. U.S.A. 110 (2013) 17720–17725.
3. J. Stern , M. Seidl-Nigsch, T. Loerting; Proc. Natl. Acad. Sci. U.S.A. 116(19) (2019) 9191-9196.
4. V. Fuentes-Landete, L. J. Plaga, M. Keppler, R. Böhmer, T.Loerting; Phys. Rev. X 9 (2019) 011015.
5. C. R. Hill, C. Mitterdorfer, T. G. A. Youngs, D. T. Bowron, H. J. Fraser, T. Loerting; Phys. Rev. Lett. 116 (2016) 215501.
6. K. Winkel, E. Mayer, T. Loerting; J. Phys. Chem. B 115 (2011) 14141–14148.

**Paola Gallo**

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## *Slow dynamics of supercooled hydration water in biosolutions*

I will show results from molecular dynamics simulations on the slow dynamics of hydration water in different biological solutions upon cooling [1-4]. In all these solutions hydration water shows the presence of two different slow relaxations, each one with its dynamic crossovers upon cooling. One of the two relaxations is the alpha-relaxation related to water glassy behavior and present also in the bulk. The other one is characteristic only of hydration water, extremely slow, and it is coupled to the slowprotein motions. I will more extensively focus on recent results of water and trehalose, a disaccharide with cryopreserving hability, and I will show that for concentrations for which trehalose molecules form a cluster in the solution hydration water behaves upon cooling similar to protein hydration water. Trehalose clusters show a Trehalose Dynamical Transition (TDT) similar to the Protein Dynamical Transition (PDT). This TDT is also coupled to the long relaxation that shows a crossover from strong to strong [3] similar to protein hydration water. In the region of mild supercooling, before the well-known fragile to strong crossover takes place, the dynamics of the beta and alpha relaxations of hydration water follows the predictions of the Mode Coupling Theory (MCT) [4].

***References:***

1. G. Camisasca, M. De Marzio and P. Gallo, Two structural relaxations in protein hydration water and their dynamic crossovers, J. Chem. Phys. 145, 044503 (2016)
2. G. Camisasca, M. De Marzio and P. Gallo, Effect of Trehalose on Protein Cryoprotection: Insights on the Mechanism of Slowing Down of Hydration Water, submitted (2019).
3. A. Iorio, G. Camisasca, P. Gallo, Slow dynamics of hydration water and the trehalose dynamical transition, J. Mol. Liq. in press (2019). https://doi.org/10.1016/j.molliq.2019.02.088.
4. A. Iorio, G. Camisasca, P. Gallo, Glassy dynamics of water at interface with biomolecules: a Mode Coupling Theory test, submitted (2019).

**Juan P. Garrahan**

School of Physics and Astronomy, University of Nottingham, UK.

## *Trajectory methods in systems with slow dynamics*

A general theoretical approach to the slow dynamics of glassy systems - the so-called dynamical facilitation theory - is built on the concept of dynamical constraints. From this perspective, the interesting properties of glass formers are to be found in the trajectories of the dynamics, and these properties are uncovered via trajectory ensemble methods. I will describe how these ideas can be used to probe in detail dynamical fluctuation properties, how these fluctuations in turn determine the melting of stable glassy states, and how to describe glassy mixtures within this approach.

**Nicolás Giovambattista**

Department of Physics, City University of New York, USA

## *Water-mediated interactions at the nanoscale*

We perform molecular dynamics simulations to study the effects of temperature and pressure on the water-mediated interaction (WMI) between two nanoscale (apolar) graphene plates at $240 \leq T \leq 400$ K and $-100 \leq P \leq 1200$ MPa. These are thermodynamic conditions relevant to, for example, cooling-, heating-, compression-, and decompression-induced protein denaturation. We find that at all (T,P) studied, the potential of mean force between the graphene plates, as a function of the plates separation r, exhibits local minima at specific plate separations $r = r_n$ that can accommodate $n$ water layers ($n = 0,1,2,3$). In particular, our results show that isobaric cooling and isothermal compression have a similar qualitative effect on the WMI between the plates; both processes tend to suppress the attraction and ultimate collapse of the graphene plates by kinetically trapping the plates at the metastable states with $r = r_n$ ($n > 0$). In addition, isobaric heating and isothermal decompression also have a similar qualitative effect; both processes tend to reduce the range and strength of the interactions between the graphene plates. Interestingly, at low temperatures, the WMI between the plates is affected by crystallization. However, crystallization depends deeply on the water model considered, SPC/E and TIP4P/2005 water models, with the crystallization occurring at different (T, P) conditions, into different forms of ice. In the last part of this talk, we will explore the WMI between hydroxylated graphene surfaces inspired on graphene oxide.  Our results show that WMI between hydrophilic-hydrophilic and hydrophilic-hydrophobic surfaces can play a fundamental role in self-assembly processes in aqueous solutions, alike hydrophobic interactions.

**Marcia C. Barbosa**

Instituto de Física, Universidade Federal do Rio Grande do Sul, BRAZIL

## *Diffusion of water confined in deformed nanotubes*

We use molecular dynamics simulations to study the diffusion of water inside deformed carbon nanotubes with different degrees of eccentricity at 300K. We compared the effect of deformation on zigzag and armchair tubes.

We show that the deformation impacts the diffusion by enhancing the mobility of tubes with small diameter while it suppresses mobility for larger diameters.

**Valeria Molinero**

Department of Chemistry, College of Science, The University of Utah, USA

## *Ice nucleation without supercooling*

Ice usually forms heterogeneously on surfaces. These surfaces bind to ice, lowering the free energy barrier for ice nucleation and promoting the formation of ice at temperatures above the homogeneous nucleation limit. The existing nucleating agents promote ice nucleation up to -1 oC. This poses the question of what is the warmest temperature at which ice can nucleate. We show that the nucleation temperature Tf could reach the equilibrium melting temperature Tm if the surface were able to induce prefreezing, the equilibrium formation of a thin interfacial layer of ice above Tm. To date, prefreezing of ice has not been realized in experiments or simulations. In this work we use molecular simulations and nucleation theory to show that the prefreezing regime is accessible to ice nucleating proteins and long chain alcohol monolayers that are compressed by less than 1% of their equilibrium area. We characterize the evolution of the thickness and structure of the prefrozen ice as a function of temperature and demonstrate that surfaces that induce prefreezing nucleate ice without supercooling. The enticing possibility of accessing the prefreezing regime by small modifications of existing surfaces opens the doors to the design of materials for energy-efficient formation of artificial snow and the induction of precipitation through cloud glaciation.

**Giancarlo Franzese**

Universität de Barcelona, SPAIN

## *Structure and Dynamics of Water at the Interface with Phospholipid Membranes*

An accurate description of the structure and dynamics of interfacial water is essential for phospholipid membranes, since it determines their function and their interaction with other molecules. Here we consider water confined in stacked membranes with hydration from poor to complete, as observed in a number of biological systems [1].

Experiments show that the dynamics of water slows down dramatically when the hydration level is reduced. Our all-atom molecular dynamics simulations identify three (inner, hydration and outer) regions, within a distance of approximately 1 nm from the membrane, where water molecules exhibit different degrees of slowing down in the dynamics [2]. The slow-down is a consequence of the robustness of the hydrogen bonds between water and lipids and the long lifetime of the hydrogen bonds between water molecules near the membrane. The interaction with the interface, therefore, induces a structural change in the water that can be emphasized by calculating its intermediate range order by adopting a sensitive local order metric recently proposed by Martelli et al. [3].

Surprisingly, at distances as far as $\simeq$ 2.5 nm from the interface, although the bulk-like dynamics is recovered, the intermediate range order of water is still slightly higher than that in the bulk at the same thermodynamic conditions [4]. Therefore, the water-membrane interface has a structural effect at ambient conditions that propagates further than the often-invoked 1 nm length scale. Membrane fluctuations smear out this effect macroscopically, but an analysis performed by considering local distances and instantaneous configurations is able to reveal it. We will discuss the implications of our conclusions for multiscale models of hydrated membranes. Our results possibly contribute to the understanding of the role of water at biomembrane interfaces [5].

### *References:*

1. C. Calero, H. E. Stanley and G. Franzese, Materials 2016, 9, 319 (2016).
2. C. Calero and G. Franzese, J. Mol. Liq. 273, 488–496 (2019).
3. F. Martelli, H.-Y. Ko, E. C. Oguz, and R. Car, Phys. Rev. B 97, 064105 (2018).
4. F. Martelli, H-Y. Ko, C. Calero, G. Franzese, Front. Phys. 13, 136801 (2018).
5. S. Samatas, C. Calero, F. Martelli, and G. Franzese, Water Between Membranes: Structure and Dynamics in "Biomembrane Simulations: Computational Studies of Biological Membranes", M. L. Berkowitz ed. (CRC Press, 2019), Chapter 2. ISBN: 9781498799799.

# INVITED LECTURES

**Shankar P. Das**

Prakash Vardhan, Arijt Mondal, Leishangthem Premkumar

School of Physical Sciences, Jawaharlal Nehru University, New Delhi, INDIA

## *Spontaneous breaking of ergodicity and configurational entropy of a supercooled liquid*

The free energy landscape (FEL) description of a supercooled liquid depicts the metastable state as a local minima of an appropriate free energy functional F[$\psi$], where the field $\psi$(x) constitutes a continuum field theoretic description of the many particle system. Unlike potential energy landscape the FEL is dependent on the thermodynamic parameters like temperature of the system. The crystal state is most stable and correspond to the deepest minimum in this free energy landscape (FEL). In the deeply supercooled state distinct basins form in the FEL corresponding to different local minima of the free energy and well below the freezing point, the corresponding FEL breaks up into an exponentially large number of basins with local minima. This transformation in the supercooled liquid has been termed as a spontaneous breakdown of ergodicity. The metastable liquid close to vitrification is viewed as being caught in one of these many possible basins. In case of structural glasses this fragmentation of the FEL occurs without presence of any quenched disorder in the system. Classical density functional theory (DFT) has been an useful tool for understanding the freezing transition of an ergodic liquid in to crystalline state. In this approach the inhomogeneous density field $\rho$(x) characterizing the metastable state is primarily described with a function depicting sharply localized density profiles. We study models of supercooled liquid with the DFT expressions for the free energy functional in terms of the field $\rho$(x). We use a field theoretic approach to understand the spontaneous breaking of ergodicity and calculate the configurational entropy of the supercooled liquid using the same density functional description.

***References:***

1. A. Mondal, L. Premkumar, and S. P. Das. Dependence of the configurational entropy on amorphous structures of a hard-sphere fluid, Phys. Rev. E 96, 012124 (2017)
2. P. Vardhan, S. P. Das. Continuum field theoretic models for supercooled liquids and entropy crisis using density functional approach (unpublished).
3. L. Premkumar, S. P. Das. Fragility index of a simple liquid using structural inputs, J. Stat. Mech., Theory and Expt, JSTAT, P093302 (2016).

**Katrin Amann-Winkel**

Department of Physics, Stockholm University, SWEDEN

## *Amorphous ices and liquid states of water*

Water is the most important liquid for life on earth. However, many properties of water are most anomalous, such as the density maximum at 4°C or the divergence of the heat capacity upon cooling. Although the water molecule is seemingly simple, the hydrogen-bonded network keeping these molecules together and determining the many anomalous macroscopic properties of water, is still a puzzle [1]. Computer-simulations suggest, that the anomalous behaviour of ambient and supercooled water could be explained by a two state model of water. The hypothetical existence of two distinct liquid states, namely high- and low-density liquid (HDL, LDL), is considered controversial. An important role in this ongoing debate plays the amorphous solid states of water. Since the discovery of two distinct amorphous ices with different density (high- and low density amorphous ice, HDA and LDA) it has been discussed whether and how this phenomenon of polyamorphism at high pressures and low temperatures is connected to the occurrence of two distinct liquid phases (HDL and LDL) [2].

X-ray scattering experiments on both supercooled water and amorphous ice are of major importance for our understanding of water [3-5]. This talk will give an overview on our recent experimental findings on amorphous ice and supercooled water. Among other techniques, X-ray correlation spectroscopy (XPCS) was used to study the dynamics in amorphous ice around the hypothesized glass transition temperature [5]. Our experimental results are consistent with the hypothesis of a liquid-liquid transition between HDL and LDL.

***References:***

1. P. Gallo et al., *Water: a Tale of Two Liquids*, Chem. Rev. 116, 7463-7500 (2016)
2. K. Amann-Winkel et al., *Water´s controversial glass transition*, Rev. Mod. Phys. 88, 0110002 (2016) 3. K. H. Kim, A. Spaeh et al., *Maxima in the Thermodynamic Response and Correlation Functions of Deeply Supercooled Water,* Science 358, 1589 (2017)
4. F. Perakis, K. Amann-Winkel et al., *Diffusive dynamics during the high-to-low density transition in amorphous ice*, PNAS 114, 8193 (2017)
5. D. Mariedahl et al., *X-ray Studies of the Transformation from High-to Low-density Amorphous Water,* Phil. Trans. A, doi: 10.1098/rsta.2018.0164 (2019).

**Tsvi Tlusty**

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KOREA

## *Proteins as evolvable amorphous matter - a nano-rheological view*

We will discuss protein as evolvable amorphous matter. In particular, we will examine the link between the internal nanoscale dynamics of the protein and its evolution. We will introduce a mechanical framework, based on Green functions, that explains how mutations shape the cooperative physical interactions in the protein that give rise to its function.

**References:**

1. S. Dutta, J-P. Eckmann, A. Libchaber, T. Tlusty. PNAS 115 (2018) E4559.
2. T. Tlusty, A. Libchaber, J-P. Eckmann. Phys. Rev. X 7 (2017) 021037.

**Mario G. Del Pópolo**

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Universidad Nacional de Cuyo, CONICET, ARGENTINA

## *Gas capture properties and mesoscopic organization of nanoporous material in the liquid phase*

Porous materials share all a common feature, the fact of being solid. Therefore, designing liquids with permanent microporosity is not only of conceptual interest, but could also lead to novel gas separation and storage technologies, as well as to tuneable reaction media. Porosity in liquids is limited to the small interstitial spaces that are transiently generated between the molecules as they move. However, as we will discuss in this talk, it is possible to produce liquids with permanent cavities by dissolving "hollow", or cage-like, molecules in solvents that are too large to enter the intramolecular cavity [1,2]. We refer to such systems as "porous liquids" (PLs). One of the most intriguing features of PLs is that depending on the cages' structure they can selectively dissolve gases and other small molecular guests. We will describe the design principles and discuss the physical characterization of porous liquids by means of molecular simulations, neutron diffraction experiments, and gas solubility measurements. We will also examine the relationship between molecular structure, mesoscopic organization of the cages, fluidity, cavity size distributions, and gas sorption capacity.

**References:**

1. N. Giri, M. G. Del Pópolo, G. Melaugh, R. L. Greenaway, K. Rätzke, T. Koschine, M. F. Costa Gomes, L. Pison, A. I. Cooper, S. L. James. Nature 527 (2015) 216.
2. G. Melaugh, N. Giri, C. E. Davidson, S. L. James, M. G. Del Pópolo. Phys. Chem. Chem. Phys, 16, (2014) 9422.

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## *Diffusion of ions and water confined in mesoporous silica and in water-in-salt electrolytes*

Experimental results on the diffusion of ions in aqueous electrolytes confined within mesoporous materials are scarce.[1,2] We have studied the mass transport of alkaline chlorides aqueous solutions confined in mesoporous silica with pore diameters in the range 2 - 56 nm, by resorting to electrical conductivity for the salts and NMR for water.[1] The diffusion coefficients of all studied electrolytes and water shows a monotonic slight decrease with decreasing diameter for pores larger than 5 nm, while the diffusivity increases markedly for smaller pores. The differences in tortuosity factors among alkaline chlorides and water become negligible for pore sizes larger than 10 nm. The spin−lattice relaxation time measurements of $^1$H-water and Li$^+$ ions confirm this behavior. The effect of the liquid/surface interactions on the determination of tortuosity factors in mesopores was studied using alkanes as a reference in the determination of the true tortuosity.[3] Water is the liquid whose mobility is least affected by the confinement, compared to the alkanes, probably because confinement suppresses the highly directional hydrogen-bonded networks associated with the formation of ice.

The ion and water transport was also studied in superconcentrated (Water-in-Salt) Li triflate aqueous solutions in the concentration range from 7 mol/kg to 21 mol/kg. Redissociation seems to takes place at the highest concentrations and the transport (conductivity  and diffusion) of Li$^+$ and water are decoupled from the bulk relaxation dynamic. Thus, they move faster than expected according to the solution viscosity. The anion is not decoupled from the dynamic of the bulk system. It can be considered as an ionic net  were ion mobility is determined by the size of the anion.

**References:**

1. D. C. Martínez Casillas, M. P. Longinotti, M. M. Bruno, F. Vaca Chávez, R. H. Acosta, H. R. Corti. J. Phys. Chem C 122 (2018) 3638−3647.
2. E. Fuentes Quezada, E. de la Llave, E. Halac, M. Jobbágy, F. A. Viva, M. M. Bruno, H. R. Corti. Chem. Eng. J. 360 (2019) 631-644.
3. L.Garro Linck, S. A. Maldonado Ochoa, H. R. Corti, G. A. Monti, F. Vaca Chávez, R. H. Acosta, J. Phys. Chem. C, submitted.

**Livia E. Bove**

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Dipartimento di Fisica, Università di Roma La Sapienza, Roma, ITALY

# *Anomalous dynamical phenomena in confined and confining water under high pressure*

The ordering of small molecules inserted in nanoporous channel systems is of great interest for applications as molecular sieves, phase separation and heterogeneous catalysts,[1], nanotribology,[2] fabrication of nanomaterials using $H_2O$ as a template[3,4] or even for the durability of concrete.[5]

As water is normally present in most molecular sieves, its order and reaction as a function of temperature and pressure is clearly of particular importance in chemistry, geology, biology, and physics. Whatever the nature of the surface of the porous matrix (hydrophilic or hydrophobic), the restriction of the accessible space induced by nano confinement has important structural, dynamical and thermodynamical consequences. In a specular way, when nano-porous water phases are used as confining matrix of simple molecules, such as hydrogen, methane or carbon dioxide, anomalous structural and dynamical behaviour are observed.

In this talk I will review some of our recent experimental results on "confined" water and "confining" water in zeolites, silica nonopores, ice clathrates cages, and filled ice channels probed by neutron, synchrotron and infrared spectroscopy under low temperature and/or high pressure.[6-11]

***References:***

1. Gelb, L. D. et al., *Rep. Prog. Phys. 62*, 1573, 1999.
2. Deville, et al., *Science 311*, 515, 2006.
3. Klein, J. & Kumacheva, E., Science 269, 816, 1995.
4. Del Rosso, L., Nature Commun. 7, 13394 2017.
5. Alabarse, F. G, J. Am. Chem. Soc. 137, 584-587, 2015.
6. Alabarse, F. G, J. Am. Chem. Soc. Accepted 2019.
7. Ranieri, U.L. et al., Nat. Comm. 8, 1076. 2018.
8. Stefanutti, E. et al., PCCP 21, 4931 2019.
9. Schaack, S., et al., JCP C 122, 11159–11166 2018.
10. Ranieri, U.L., et al., JCP C 123, 1888−1903 2019.
11. Bove L. E., Ranieri, U. L. Phil. Trans. R. Soc. A 20180262 2019, in press.

**Damian Scherlis**

Instituto de Química Física de los Materiales, Medio Ambiente y Energía
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## *Dissociation of water in nanoscale confinement*

The effect of extreme confinement on chemical equilibrium is an open question whose elucidation poses a major challenge to modern laboratory techniques. In particular, no experimental information is available on the equilibrium constant for water dissociation in nanoenvironments, despite speculations suggesting that it may change by orders of magnitude. In the present talk we show results from first principles free-energy simulations indicating that the pKw remains unaletered all the way down to subnanometer confinement, when it is strongly affected by dimensionality and size. All in all, we find the dependence of pKw with pore size is not a monotonical function with a previsible trend, but may vary in sharp and unexpected ways in severely confined environments.

**E. Anibal Disalvo**

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## *Correlation between hydration of carbonyl and phosphate groups in lipid membranes and its effect on electrical, compressibility and defect formation*

Water intercalates at different extents in the acyl chain region of lipid membranes in correlation with the hydration of the phosphate groups. This correlation is sensible to the chain length, the presence of double bonds and the phase state of the lipid membrane.

The lack of carbonyl groups gives the lipid membrane a higher level of free water molecules around the phosphate groups as observed from the comparison of ester PC and diether PC by the correlation between FTIR/ATR and fluorescent analysis. The higher interaction of water with phosphate groups results in a higher disorder, i.e. more polarization propensity of the interphases which is modified by the lateral surface pressure and thus affecting the dipole potential. The absence of CO groups can be in part compensated by the presence of cholesterol that modulates the water level.

The presence of CO groups and its associated water may produce modulation of packing and hydration suitable for the incorporation of aminoacid of peptides and enzymes.

**M. Paula Longinotti**

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M. Florencia Mazzobre.
Departamento de Industrias, Facultad de Ciencias Exactas y Naturales, Universidad de Buenos Aires.

## *Phase transitions of water solutions in mesoconfined systems*

The effect of confinement at the nanoscale on mixtures is a problem with many questions still unsolved. In the last years, several studies were published in the literature considering the nanoscale confinement effect on the dynamics and phase behavior of different mixtures. Among those mixtures aqueous solutions are of great interest. In this talk, I will discuss the effect of confinement at the nano scale on the phase behavior of glycerol and LiCl aqueous solutions. In the former case, we studied the glass transition temperature ($T_g$) of aqueous glycerol solutions over the concentration range between 70 and 100 wt% polyol. In the latter, we analyzed the glass transition of LiCl aqueous solutions with salt concentrations with eutectic ($R$ = 7, with $R$ = moles of water/moles of LiCl) and subeutectic compositions ($7 < R \leq 15$). All aqueous solutions were confined in mesoporous silica matrixes with pore diameters between 2 and 58 nm.

The results show that for glycerol-aqueous solutions $T_g$ decreases with decreasing pore size, being this effect more pronounced with increasing polyol content. This effect will be discussed in terms of the cooperative rearranging regions length scale. In the case of LiCl aqueous solutions, we observed that the glass transition temperature increases by about 2-5 K for the smallest pore diameters, which is equivalent to the effect of applying a pressure of up to 100 MPa on the bulk samples. This behavior is opposite to that observed in aqueous subeutectic NaCl solutions confined within similar silica matrixes.

**Limei Xu**

International Center for Quantum Materials, Peking University, CHINA

## *Liquid-liquid phase transition in glass-forming liquids*

For a group of substances with different chemistry but share similar anomalous properties (e.g., density anomaly, diffusivity anomaly), the existence of a liquid-liquid critical point as well as liquid-liquid phase transition is assumed to be the key. However, the experimental confirmation remains challenging since the hypothesized critical point and phase transition are located in the phase region below the homogeneous crystallization line which is difficulty for experiment to detect directly. In this talk, I will discuss our recent studies on the liquid-liquid phase transitions from supercritical region. Our results shows that the structure change reflected in the potential energy surface is directly connected to the change in dynamic properties, which is important for the investigation of glass transition at low temperature and thus can also be employed to investigate the liquid-liquid transition occurring at in supercritical phase region.

**Gustavo Appignanesi**

Instituto de Química del Sur, Universidad Nacional del Sur,
Bahia Blanca, ARGENTINA

## *Liquid water: A two-state scenario with "three" molecular classes?*

The two-liquids scenario for water assumes the existence of two competing local arrangements: well-structured low-local density molecules and unstructured high-local density ones. By studying molecular dynamics simulations (real dynamics) for different water models combined with potential energy minimizations (inherent-structures scheme) we will show the existence of a “third class” of molecules beyond the two stable states: thermally distorted albeit inherently structured molecules. While the minimization procedure reverts them to a structured IS, these molecules would be classified as belonging to the unstructured state at the real dynamics. This information will be embodied by a new structural indicator we will hereby propose and whose virtues and potentialities will be explored by this talk.

**Mario Tagliazucchi**

Gervasio Zaldivar

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Martin Conda-Sheridan

Department of Pharmaceutical Sciences, College of Pharmacy, University of Nebraska Medical
Center, Omaha (NE), USA

## *Self-Assembly of Peptide-Amphiphiles in Aqueous Solutions*

This talk will present our recent advances in understanding the self-assembly behavior of peptide amphiphiles, which are molecules composed of a hydrophilic peptidic headgroup and a hydrophobic alkyl tail. These surfactants can self-assemble in solution into different types of structures, such as spherical micelles, long fibers, lamellar ribbons, vesicles, etc. We used a molecular theory to predict the relative stability of these structures as a function of the molecular structure of the peptide amphiphile and solution composition (pH and ionic strength). I will briefly discuss how these parameters direct the outcome of the self-assembly process and compare the morphology of the self-assembled aggregates and the apparent pKa of the peptide amphiphiles within the aggregates with experimental data. I will also show that self-assembled lamellar ribbons can twist with a period that depends on the width of the ribbon and provide a possible theoretical explanation for this interesting phenomenon. Finally, I will present some preliminary experimental results for the electrochemically-driven self-assembly of peptide amphiphiles.

**Yanina Fasano**

Centro Atómico Bariloche (CNEA), CONICET
Instituto Balseiro, Universidad Nacional de Cuyo, Bariloche, ARGENTINA

## *Disorder-induced kinetic arrest in the vortex melting transition in type-II superconductors*

First order phase transitions (FOT) are ubiquitous in nature, including structural, magnetic, electric and electronic transformations with a discontinuous entropy-change and its associated latent heat. In real experimental systems, the unavoidable quenched disorder affects the thermodynamic properties of FOT. Even a slight amount of disorder can produce a rounding of the FOT entailing a depletion of the latent heat.[1] Contrasting this rounding prediction with experimental results is challenging since the unavoidable static finite size and dynamic kinetic arrest effects smear an unified description for a wide variety of condensed matter systems. Although no general picture is still available, most condensed-matter systems share the stabilization of glassy phases on increasing disorder.[2] Vortex matter in type-II superconductors is a model system to study how on increasing the disorder strength, density, and spatial correlation, a FOT in pristine samples can mute to a continuous second order transformation or even a crossover line. We study vortex matter in the high--temperature $Bi_2Sr_2CaCu_2O_{8+\delta}$ superconductor with correlated disorder in the form of a low dose of columnar defects, focusing on the changes induced in the FOT vortex melting transition on increasing disorder. We use dc and ac local Hall magnetometry to track the melting temperature and entropy-jumps. We found that the transition remains first-order up to a critical density of disorder, but for larger densities of columnar defects the transition is smeared and the typical features are frequency dependent, a phenomenology typically observed in kinetically-arrested phase transitions.

**References:**

1. Y. Imry, M. Wortis, Phys. Rev. B 19 (1979) 3580.
2. J. A. Mydosh, *Spin glassses: An experimental introduction*, Taylor and Francis, London (1993).

# ORAL PRESENTATIONS

## LAHN PRESENTATION

### Recent progress in the development of the Argentine Neutron Beam Laboratory ¨LAHN¨

Federico Suarez

Laboratorio Argentino de Haces de Neutrones, Comisión Nacional de Energía Atómica

The RA-10 is a 30 MW multi-purpose reactor under construction in Buenos Aires, designed to satisfy national and regional demand for radioisotopes, nuclear materials testing and neutron beams research. It is foreseen to start operations by the end of 2022. Within the National Atomic Energy Commission we are working on the development of the ”Argentine neutron beam laboratory for the RA-10 reactor” (LAHN); aimed at implementing state-of-the-art instruments, developing a user community and the laboratory staff. The LAHN will be a world class facility available to users from all around the world, and will become the first large-scale neutron laboratory in South America.
During the first stage, two instruments are being designed: (i) a neutron imaging instrument placed on a cold beam; and (ii) a multi-purpose diffractometer placed on a thermal beam, optimized for non-destructive studies on large objects. Both instruments will be located at the reactor face, in order to exploit very intense, undisturbed, neutron beams. A second phase for the project has already been launched too, taking into consideration the demands of the local and regional scientific community. Some instruments will be installed at the guides hall of LAHN coming from laboratories in Europe which are closing their reactors. Besides this, an ambitious program is running to popularize neutron techniques in Argentina and create new users. In this talk, the current state of the project will described, providing details of the instruments to be installed, training opportunities and strategies to develop the Argentine user community.

## ORAL 1

### Structure and dynamics in glassy nanoparticles

Richard K. Bowles

Department of Chemistry, University of Saskatchewan, Canada

Molecular dynamics simulation is used to study the relationship between structure and dynamics in supercooled binary Lennard--Jones nanoparticles over a range of particle sizes. The glass transition temperature of the nanoparticles is found to be significantly lowered relative to the bulk, decreasing as $N^{-1/3}$ with decreasing particle size. This allows the nanoparticles to sample low energy states on the potential energy landscape and we are able to study their relaxation times, measured in terms of the intermediate scattering function, and their structure, measured in terms of locally favoured structures, to low temperatures. We show that the growing relaxation times in the supercooled nanoparticles are coupled with the growth of physical clusters formed from favoured local structures in a way that is well described by the Random First Order Transition entropic droplet model.

**ORAL 2**

## On the effect of an external field in barrier-tunneling in glassy dynamics

Isidoro González-Adalid Pemartín,[1] V. Martin-Mayor,[1,2] G. Parisi,[3] J. J. Ruiz-Lorenzo[4]

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[4]Departamento de Física, Universidad de Extremadura and Instituto de Computación Científica Avanzada (ICCAEx), Universidad de Extremadura, Badajoz, Spain, and Instituto de Biocomputación y Física de Sistemas Complejos (BIFI), Zaragoza, Spain.

The extremely slow glassy dynamics is a major problem in the investigation of the glass transition. Albeit powerful sampling methods (such as swap dynamics or parallel tempering) produce equilibrium configurations at low temperatures, the problem remains that the glass transition is essentially a dynamic problem. This means that, even if a smart algorithm has provided us with well-equilibrated starting configurations, we still need to follow from that start point a (extremely slow) nature-mimicking dynamics in order to obtain meaningful results.

I shall explain here an alternative approach in which we overcome free-energy barriers by switching on an external field. Furthermore, the reaction of the dynamics to the external field carries information on the free-energy landscape. The proposal has been put to practice in the numerical investigation of small spin-glass samples in three dimensions.[1] Incidentally, a similar approach is being pursued in the experimental investigation of the dielectric response of glycerol.[2]

In this way, we have achieved reduction of tunneling times by some three orders of magnitude. Furthermore, the external field strongly enhances the homogeneity of the relaxation times for different samples and/or initial conditions. However, the expected reduction of tunneling times is quantitatively achieved only for very small values of the external field. The dynamic response to the field is strongly non-linear, something that we have partly explained though the concept of virtual states.

**Referencias:**

1. I. Gonzalez-Adalid Pemartin, V. Martin-Mayor, G. Parisi and J. J. Ruiz-Lorenzo, J. Phys. A: Math. Theor (2019). publish online https://doi.org/10.1088/1751-8121/ab08d9.

2. D. L'Hôte, R. Tourbot, F. Ladieu and P. Gadige, Phys. Rev. B 90 (2014) 104202.

**ORAL 3**

# Hyperuniform vortex patterns at the surface of type II superconductors

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A many particle system must possess long-range interactions in order to be hyperuniform[1,2] (i.e. suppress long-wavelength density fluctuations) at thermal equilibrium. Hydrodynamic arguments and numerical simulations show, nevertheless, that a three-dimensional elastic-line array with short-ranged repulsive interactions, such as the one induced and directed by a magnetic field along the *z* direction in a type-II superconductor, form at equilibrium a class-II hyperuniform two-dimensional point pattern in any constant-*z* cross section, with point density fluctuations vanishing isotropically as $q^{\alpha}$ with $\alpha=1$ at small wavevectors q. This prediction includes both the solid and liquid vortex phases in the absence of disorder, and also the liquid phase in presence of weak uncorrelated quenched disorder. We show that the three-dimensional Bragg-glass phase is marginally hyperuniform, while the Bose-glass and the liquid-phase with columnar disorder are expected to be non-hyperuniform at equilibrium, with the exception of the Mott-Glass phase. Motivated by these theoretical predictions and recent simulations we experimentally study large wavelength vortex density fluctuations of magnetically decorated vortex structures in pristine, electron irradiated and heavy-ion irradiated $Bi_2Sr_2CaCu_2O_{8+\delta}$ superconducting samples in the mixed state. For most of our samples we find nearly hyperuniform two-dimensional point patterns at the superconductor surface with an effective exponent $\alpha_{eff}$ close to 1. We interpret these results in terms of a large-scale memory of the high-temperature line-liquid phase due to glassy dynamics during the field-cooling protocol. We also discuss the crossovers expected from the dispersivity of the elastic constants at intermediate length-scales, and how finite-size effects in the $z$-direction should kill the hyperuniformity in the x-y plane for lengths $q^{-1}$ above the superconductor thickness. We argue these predictions may be observable and propose further experiments to independently test them.

**References:**

1. S. Torquato & F. H. Stillinger, Phys. Rev. E 68, 041113 (2003).
2. S. Torquato, Phys. Rev. E 68, 022122 (2016).

## ORAL 4

### Memory functions of supercooled liquids: from mean field to three dimensions

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We develop a hybrid numerical approach to extract the exact memory function K(t) of a tagged particle in three-dimensional glass-forming liquids. We compare the behavior of the exact memory function to two mean-field approaches, namely the standard mode-coupling theory and a recently proposed ansatz for the memory function that forms the basis of a new derivation of the exact form of K(t) for a fluid with short-ranged interactions in infinite dimensions. Each of the mean-field functions qualitatively and quantitatively share traits with the exact K(t), although several important quantitative differences are manifest.

## ORAL 5

### Correlations of local relaxation rates in glassy systems

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We discuss a quantitative description of dynamical heterogeneity in glass-forming liquids in terms of fluctuations in the local relaxation rates. We extract a recently defined two-point correlator, s(q,t), that probes those fluctuations. We define a two-point dynamical susceptibility $\chi^{\phi}{}_2(t)$ as the long wavelength limit of s(q,t). We find that the long time decay of this susceptibility is characterized by a certain time scale, the exchange time $\tau_{ex}$, which we identify as the typical lifetime of the heterogeneities.

We numerically study the equilibrium dynamics of a 3D binary hard-sphere (HARD) mixture and a 3D Kob-Andersen Lennard-Jones (KALJ) mixture. We extract the exchange time $\tau_{ex}$ as a function of either the temperature (for KALJ) or packing fraction (for HARD). In HARD systems, close to the glass transition, $\tau_{ex}$ is an order of magnitude longer than the α-relaxation time $\tau_\alpha$, and $\tau_{ex}/\tau_\alpha$ increases with packing fraction. However, in KALJ systems, $\tau_{ex}$ is approximately twice $\tau_\alpha$, and $\tau_{ex}/\tau_\alpha$ has no significant dependency on temperature. We also measure a two-point correlation length for the local relaxation rates. We find that for HARD near the glass transition this correlation length grows approximately logarithmically with time up to a maximum at time around $2\tau_\alpha$, and it decays at long times.

**ORAL 6**

# Ice growth kinetics and quasi-liquid layer thickness as revealed by environmental AFM

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The existence of a QLL at the ice–air interface has been thoroughly discussed in the literature, mainly considering that this layer plays an important role in the flow behavior of ice and snow, the adsorption of substances onto ice, and the low friction of solids on ice. Moreover, ice crystal growth from the vapor phase is an important microphysical process critical for the evolution of atmospheric cold clouds. The rates of water condensation and evaporation on atmospheric particles influence the lifetime properties of clouds, and thereby affect the atmosphere's radiation budget, the formation of precipitation and weather. Quantitative understanding of both the QLL thickness and the kinetics of ice growth are extremely important for atmospheric science, being also crucial for many industrial processes. For example, multiphase modeling of gas phase reactions, photolysis reactions and aqueous reactions in suspended aerosol particles depend on the QLL thickness.

Therefore, in this work we will show an environmental atomic force microscopy (AFM) technique developed in our group to measure the quasi-liquid layer (QLL) thickness of ice and the kinetics of ice growth from the vapor phase under humidity and temperature-controlled conditions. Both quantities were studied by measuring force curves over pure ice deposited on mica under equilibrium and non-equilibrium conditions, respectively.

Our results suggest a QLL thickness lower than 1 nm in the temperature range between -7 and -2 ºC, that is in the lower-bound of other experimental determinations. This suggests that other authors overestimated the QLL thicknesses due to temperature gradients and indentation of ice. In addition, this experimental technique allowed obtaining more reliable ice growth rates, since temperature and humidity were controlled close to the ice nucleation centers avoiding systematic errors.

# POSTERS

## POSTER 1

### Is the HDL-LDL equilibrium a thermally distorted version of HDA-LDA?

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Several indicators of local structure have been devised to evidence the two hypothetical types of coexisting liquid water molecules: high and low local density ones (HDL and LDL, respectively). In general, all of them have focused on detecting the more-ordered lower density species assuming its structural similarity with ice, mainly overlooking the molecules comprising the high-density state. However, we will show that the latter are not as unstructured as it was originally supposed[1]. We will also evidence that locally structured molecules are prone to exhibit slow dynamic propensity and, more conspicuously, that locally unstructured molecules are extremely subject to high dynamic propensity[2]. Moreover, we shall develop a new structure indicator (producing neat bimodal behavior) that will re-emphasize the relevance of the unstructured fraction of liquid water. We will show that truly HDL-like molecules are much scarcer than formerly supposed and easily confused with inherently structured but thermally distorted molecules. Finally, comparing radial distribution functions as obtained from trajectories of molecular dynamics simulations with those from their respective inherent (thermally quenched) structures, we shall furnish new support for the theory of HDL-LDL coexistence. Furthermore, our results postulate the possibility of considering the HDL-LDL equilibrium as the thermally distorted version of the equilibrium between their solid amorphous relatives, HDA-LDA.

**References:**

1. Montes de Oca, J. M., Accordino, S. R., Verde, A. R., Alarcón, L. M. & Appignanesi, G. A. Phys. Rev. E 99 (2019) 062601.
2. Verde, A. R., Montes de Oca, J. M., Accordino, S. R., Alarcón, L. M. & Appignanesi, G. A. J. Chem. Phys., a*ccepted* (2019).

## POSTER 2

# Estimating the length-scale of influence of dynamical heterogeneities in glassy relaxation: Size-dependence study of localized dynamic fluctuations

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We study the evolution of dynamic fluctuations averaged over different space lengths and time scales to characterize spatially and temporally heterogeneous behavior in glassy systems: simulated binary Lennard-Jones systems [1], TIP4P/2005 water [2] and experimental dense colloidal suspensions [1]. We show that the onset of glassiness is marked by a significant enhancement of spatially localized dynamic fluctuations originating in regions of correlated mobile particles. Thus, these regions are responsible for a clear initial departure of the fluctuations from the usual power law decay (the inverse of the square root of N trivial system-size dependence expected for an uncorrelated system) until the behavior reverts to such regime at large system sizes. Both the magnitude of the departure and the molecular size at which the trivial regime is finally recovered grow as we increase glassiness. Thus, this method provides a means to estimate an upper limit to the maximum length scale of influence of the regions of correlated mobile molecules that, notably, we show to increase significantly as we move deeper within the glassy regime.

**References:**

1. J. A. Rodriguez Fris, E.R. Weeks, F. Sciortino, and G.A. Appignanesi, Phys. Rev. E Rapid Comm. 97, 060601 (2018).
2. J. M. Montes de Oca, S. R. Accordino, G. A. Appignanesi, P. H. Handle, and F. Sciortino, J. Chem. Phys. 150, 144505 (2019).

**POSTER 3**

# Hydration studies on lipid membranes: Classification of water molecules in different bilayers in gel and liquid phase.

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We use molecular dynamics simulations to study the hydration properties of lipid membranes, both in the gel state and in the crystalline fluid. We show that while the hydration centers remain significantly hydrated in both phases, the gel-fluid transition implies significant changes in the second hydration layer, particularly in the region of the hydrocarbon tails. Thus, while the water population is almost nil in the hydrocarbon tails zone in gel state, this region becomes partially accessible to water in crystalline liquid state in membranes of dipalmitoylphosphatidylcholine (DPPC). In the case of membranes composed of unsaturated lipids, the presence of double bonds modifies the hydration map particularly in the hydrocarbon zone. We also show that the water molecules located in the non-polar zone have a lower hydrogen bond (HB) coordination in comparison with the molecules of the first hydration layer, which are arranged in relatively compact 'nanoclusters'. The water molecules in the hydrocarbon tails zone tend to be organized in less compact structures, like branched chains, with a small population of isolated molecules. This behavior is similar to that observed in other hydration contexts, such as water penetrating carbon nanotubes or hydrophobic pores, reflecting the reluctance of water to sacrifice HB coordination.

**POSTER 4**

# Effective models for grafted polymer nanoparticles in two dimensions

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In this work, a two dimensional system of polymer grafted nanoparticles is > analyzed using large-scale Langevin Dymanics simulations. Effective core-softened potentials were obtained from the Ornstein–Zernike equation for two cases: one where the polymers are free to rotate around the nanoparticle core and a second where the polymers are fixed, with a 45º angle between them. The use of effective core-softened potentials allow us to explore the complete system phase space. In this way, the PT, T$\rho$ and P$\rho$ phase diagrams for each potential were obtained, with all fluid and solid phases. The phase boundaries were defined analyzing the specific heat at constant pressure, the system mean square displacement, the radial distribution function and the discontinuities in the density-pressure phase diagram. Also, due the competition in the system we have observed the presence of water-like anomalies, such as the temperature of maximum density - in addition with a tendency of the tmd to move to lower temperatures (negative slope)- and the diffusion anomaly. It was observed different structural morphologies (worms, stripes, amorphous) for each nanoparticle case. We observed that for the fixed polymers case the waterlike anomalies are originated by the competition between the potential characteristic length scales, while for the free to rotate case the anomalies arises due a smaller region of stability in the phase diagram and no competiton between the scales was observed.

**POSTER 5**

# Self-assembled nanostructures of peptide-amphiphiles: Charge regulation by size regulation

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Self-assembled nanostructures of peptide amphiphiles (PAs) with molecular structures $C_{16}K_2$ and $C_{16}K_3$ (where C indicates the number of carbon atoms in the alkyl chain and K are lysines in the head group) were studied by a combination of theoretical modeling, transmission electron and atomic force microscopies and acid-base titration experiments. The supramolecular morphology of the PAs (micelles, fibers or lamellas) was dependent on the pH and ionic strength of the solution. Theoretical modeling was performed using a molecular theory that allows to determine the equilibrium morphology, the size and the charge of the soft nanoassemblies as a function of the molecular structure of the PA, and the pH and salt concentration of the solution. Theoretical predictions showed good agreement with experimental data for the pH-dependent morphology and size of the nanoassemblies and their apparent pKas. Two interesting effects associated with charge-regulation mechanisms were found: firstly, ionic strength plays a dual role in the modulation of the electrostatic interactions in the system, which leads to complex dependencies of the aggregation numbers with salt concentration; secondly, the aggregation number of the nanostructures decreases upon increasing the charge per PA. The second mechanism, charge regulation by size regulation, tunes the net charge of the assemblies in order to decrease the electrostatic repulsions. A remarkable consequence of this behavior is that adding an extra lysine residue to the charged region of the PAs can lead to an unexpected decrease in the total charge of the micelles.

**POSTER 6**

# Anomalous diffusion of water molecules at grain boundaries in ice Ih

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Water ice is one of the most abundant crystalline solids on Earth and its properties on a macroscopic scale are of interest in a variety of contexts [1]. One of these involves the mechanical behavior, which controls, for instance, the flow dynamics of large ice masses. Given the polycrystalline character of crystalline ice, it is important to assess the role of grain boundaries. Using *ab initio* and classical molecular dynamics simulations, we study the pre-melting phenomena [2] of a number of symmetric tilt boundaries in proton-disordered hexagonal ice Ih. With regard to molecular mobility, the translational motion of the water molecules is found to be subdiffusive for time scales longer than 10 ns. Furthermore, it is well-described by a continuous-time random walk model characterized by a waiting-time distribution with a power-law decay, suggesting that the dynamics in the pre-melt layers at GBs in ice Ih is glassy in nature, even at temperatures close to Tm [3].

**References:**

1. V. F. Petrenko and R. W. Whitworth, Physics of Ice, Oxford University Press, (1999).
2. J. G. Dash et. al, Rev. of Mod. Phys, 78, 695 (2006).
3. P. A. F. P. Moreira, R. G. de Aguiar Veiga, I. de Almeida Ribeiro, R. Freitas, J. Helfferich, and M. de Koning. (2018). Phys. Chem. Chem. Phys., 20, 13944 (2018).

**POSTER 7**

# An alternative approach to study the heterogeneous dynamics and its consequent dynamic correlation length that emerges in a glass-former.

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In this work, the relaxation of a simple bead-spring polymer system was investigated with Molecular Dynamic simulations near its glass transition temperature. We developed a new way to look at the dynamic length scale in this glass-former, focusing on the correlated motion of particles in an isoconfigurational ensemble (ICE). We found that, while the usual dynamic four-point correlation length deviates from the structural length scale (calculated with the point-to-set method) at low temperatures, Pearson's length behaves similar to the static length in the whole temperature range [1]. Moreover, similar to what happens with mobility, monomers with higher correlation are clustered, and the clustering increases with decreasing temperature. An interesting result is that regions with high Pearson dynamic correlation are not coincident with highly mobile or immobile regions. This results represents a new approach to the study of dynamic heterogeneity that emerges in glass forming liquids, complementing the more traditional characterization in terms of mobility [2]. Additionally we studied the breakdown of the Stokes-Einstein Relation (SER) in connection with dynamic susceptibility calculated for an ICE, such that the effect of the structure on dynamics can be discriminated. We found that the onset of structure effects on the dynamical behavior is coincident with the onset of SER breakdown [3].

**References:**

1. C. Balbuena, M. M. Gianetti, and E. R. Soulé, J. Chem. Phys., **150**, 234508 (2019).
2. C. Balbuena, M. M. Gianetti, and E. R. Soulé, J. Chem. Phys., **149**, 094506 (2018).
3. C. Balbuena and E. R. Soulé, J. Phys.: Condens. Matter, submitted.

**POSTER 8**

## Determination of $K_w$ in nanoconfined water in nanodroplets

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In this work we use DFT Car-Parrinello molecular dynamics combined with the Umbrella sampling scheme to study the self dissociation constant of water ($K_w$) confined in nanodroplets. Three systems of different size were used to analyze the effect of confinement: aggregates of 20, 12, and 6 water molecules. We show that in the largest droplets (20 and 12 molecules) there is no difference in the value of the dissociation free energy with respect to bulk water, whereas for the hexamer we observe an increase in said energy (or equally that the Kw value drops for the smallest system). It is also shown that, for the larger droplets, the solvation shells of the hydronium and the hydroxide ions resulting from the dissociation is similar to those in the bulk but the solvation changes for the 6 molecules cluster. Thus, most of the energetics of this reaction appears to be already captured by the first hydration core, present in the two bigger clusters. The structural constraints in the hexamer, on the other hand, result in a deficient solvation of the products, which seems to be the reason as to why the free energy of dissociation becomes unfavored in this case.

**POSTER 9**

## Study of the efficiency of nanostructures for water desalination. From macroscale to atomistic approach

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In order to evaluate how membrane permeability affects the performance in desalination, in this work the reverse osmosis desalination system was simulated by using a macroscale flow model and the potential is quantified in terms of improvements in membrane permeability. We find that increasing the membrane permeability by one order of magnitude would allow for 15% (SWRO - seawater) and 24% (BWRO - brackish water) less energy or 60% (SWRO) and 68% (BWRO) fewer pressure vessels with a given capacity. Besides that, the ultrafast water transport in nanoconfinement environmental such as carbon nanotubes, graphene and molybdenum disulfide nanopores hold the greatest potential to cause a reduction in desalination operating costs. In addition, the water flux throgouth a nanoporous membrane based on a 2D material was evaluated using molecular dynamics approach.

**POSTER 10**

# Preferential adsorption in ethane/carbon dioxide fluid mixtures confined within silica nanopores

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Molecular Dynamics simulations have been performed to investigate the structural and dynamical properties of fluid ethane confined within cylindrical silica nanopores of diameters 4 and 1 nm. The pure ethane phase and equimolar mixtures of ethane and $CO_2$ were considered for the analysis. Computer simulations were carried out along the isotherm T = 320 K, exploring densities in the range $\rho/\rho_c = 0.05 - 2.18$. Density profiles along the axial and radial directions of the pore, orientational distribution functions, analysis of interfacial dynamic properties and estimations of diffusion coefficients are presented. In agreement with experimental data, the results show the formation of a dense adsorption layer, with densities that may be up to 10 times larger than those of the bulk phase, particularly at low densities and under sub-nanometer confinement. The incorporation of $CO_2$ within the pores, in equimolar mixtures, modifies the structure of ethane species, leading to preferential adsorption of $CO_2$ over ethane species. This preference is reflected in adsorption layers having mole fractions of $CO_2$ above 50%, that rise up to 80% at the highest bulk density studied. The dynamics of ethane is also modified in mixtures, as manifested by the increments measured in the diffusion coefficients of confined ethane in presence of $CO_2$, in accordance with experiments. In contrast with experimental data, our results for the diffusion coefficients of confined ethane showed a monotonic decreasing behavior with increasing bulk density. Possible reasons for this discrepancy between experiments and simulations are discussed.

**POSTER 11**

## Response of water behavior at the interphase of phospholipid matrixes assessed by FTIR

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Lipid membranes are one of the most important biological matrixes in which biochemical processes take place. This particular lipid arrangement is driven by different water disposition interacting with it, which is related to different water states with different energy levels at this interphase.

In our work we report changes in water content and distinctive water states by FTIR when this self assembled matrix changes its physicochemical properties. The OH stretching band in the liquid water IR spectrum is the principal region used to understand its molecular organization (4000-3000cm$^{-1}$). The strength of the hydrogen-bonding depends on the cooperative/anti-cooperative nature of the surrounding hydrogen bonds, with strongest hydrogen bonds giving the lowest vibrational frequencies.

Thus, we can use water as mirror of the membrane state in this kind of biological systems. Different phospholipids associate water at particular modes according to their structure; this may produce modulation of packing and hydration suitable for the incorporation of aminoacid of peptides and enzymes.

**POSTER 12**

## Modelling of surface nanobubbles

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We present a Molecular Dynamics - Kinetic Monte Carlo simulation scheme to describe the electrochemical production of gas in nanometric electrodes. This method, combined with coarse grained potentials, allows to study the nucleation, growth and stability of surface nanobubbles in controlled electrochemical conditions. The computed current-voltage response shows agreement with measurements from single nanobubble experiments [1]. At steady state, the electrochemical reaction proceeds on the three-phase interface, resulting in a diffusionaly controlled current. The effect of the applied potential on nanobubble's size and shape is discussed. This scheme turns out to be a unique approach for the microscopic investigation of redox processes dynamics at nanoelectrodes

**References:**

1. Luo, Langmuir, 29(35), 11169-11175.

**POSTER 13**

# Water can vitrify on the nanoscale at ambient conditions

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Nanoconfined water is commonly present at ambient conditions naturally and industrially, for example, it appears within and around biopolymers in cells, in clay mineral, and in nano electromechanical systems. To what extent does the configurational change of water molecules slow down with decreasing the confinement size, and does it cease eventually, that is, a liquid-glass transition occurs?. To answer the questions is important not only to understand the mechanism underlying the properties and functions of systems where nanoconfined water resides but also to learn about unusual characteristics of water. There are, however, few studies exploring the questions and no one has observed a liquid-glass transition of water with changing the confinement at ambient conditions. In the present study, we have conducted persistent spectral hole-burning spectroscopy of probe molecules within reverse micelles at the water-droplet sizes from ~1.6 nm to ~7 nm. The results demonstrate that the water is in a glassy state below ~4 nm on a laboratory time scale, that is, the liquid-glass transition takes place at 4 nm owing to the confinement. For bulk water, the configurational change is on the picosecond scale at ambient conditions, and the glass-transition temperature is commonly believed to be ~136 K. Hence, our results are surprising because nanoconfined water exhibits drastic changes in the relaxation time and in the glass-transition temperature compared with bulk water. Furthermore, our findings for soft matter will shed new light on understanding the biological function in cytoplasm with molecular crowding and glassy behaviors

**References:**

1. Murakami et al., J. Phys. Chem. B 115 (2011) 5877; Phys. Rev. E 88 (2013) 052304; J. Chem. Phys. 148 (2018) 144505.
2. Zhoua et al., Proc. Natl. Acad. Sci. USA, 106 (2009), 10632.

**POSTER 14**

## Computer simulation of liquids with permanent porosity

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Porosity is commonly associated with solid materials like zeolites, MOFs, COFs, among others. However, in recent years, the concept of porous liquids (PLs) has emerged as a counter-intuitive new class of materials that combines the fluidity of liquids with some properties of porous solids [1]. These PLs consist of hollow organic cages -discrete organic molecules that contain a permanent, empty cavity- dissolved at high concentrations in solvents that are too large to occupy the cavities. This combination results in materials that flow freely at room temperature, and carry some of the physical properties of the cage molecules, such as gas uptake capacity and guest binding selectivity, into the liquid phase [2]. Our research is focused on the characterization of porous liquids with widely different chemical compositions [3,4]. In the present work, we resort to a combination of experimental and computational tools, namely neutron diffraction experiments and Molecular Dynamics simulations, to analyze the structure and dynamical properties of highly concentrated mixtures of iminospherand cages in organic solvents of appropriate size. We also report gas uptake simulation results for a variety of hydrocarbons, and discuss the transport dynamics of different guest molecules inside the PL.

**References:**

1. N. O'Reilly, N. Giri, S. L. James, *Chem. Eur. J.* 13 (2007) 3020.
2. N. Giri, C. E. Davidson, G. Melaugh, M. G. Del Pópolo, J. T. Jones, T. Hasell, A. I. Cooper, P. N. Horton, M. B. Hurst-house, S. L. James, S. L. *Chem. Sci.* 3 (2012) 2153.
3. G. Melaugh, N. Giri, C. Davidson, S. James, M. Del Pópolo, Phys. Chem. Chem. Phys. 16 (2014) 9422.
4. N. Giri, M. G. Del Pópolo, G. Melaugh, R. L. Greenaway, K. Rätzke, T. Koschine, L. Pison, M. F. C. Gomes, A. I. Cooper, S. L. James, *Nature* 218 (2015) 216.

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Many of you have largely helped us to shape our scientific view, our scientific gaze. And this is very important for us, not only on a scientific basis, but also on human grounds. Thus, we expect this to be a new beautiful opportunity in this regard.

Science is capable of producing beauty, just like art. And precisely for this ability to uncover beauty, the scientific view holds, like the poetic word, a powerful philosophical breath, even a deep ethical imprint. Since by discovering beauty in its object of study, it rescues the unseen, the everyday, the apparently trivial and irrelevant; in short, the ignored. And thus it re-signifies it. It forces us to discover the innate beauty that it deeply holds.

The scientist's vision, like that of the poet, is thus sometimes the lover's gaze, that vision that compels us to also behold the world like lovers. An antidote against apathy, a plea to remain aware of that inherent incommensurable beauty that embodies each one of our neighbors, that is evident in each corner of the world, that inhabits everything.

This is why we believe that scientists, like poets, will always be essential.

**Thank you for coming!**

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