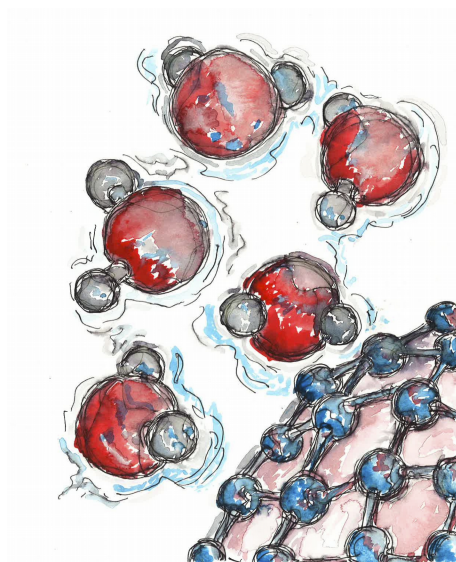# Water at interfaces: from proteins to devices

November 29, 2016 - December 02, 2016
CECAM-AT

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1 Description

Due to the importance of water in many fields of science among which there are physics, biology, medicine, water treatment, atmospheric science, engineering, there is growing interest towards the behaviour of water at interfaces. Such field of study been largely emphasized in recent publications but, contrary to bulk water, it was subject only lately of few meetings. The goal of this workshop is to gather leading scientists, from a wide spectrum of disciplines ranging from biophysics to material sciences, working on modeling and experimental aspects of water at interfaces and at different length scales. We believe that the state of the art in water modeling is mature to address important applications in the fields aforementioned, hence the timeliness of this meeting is optimal and the location ideal with many local groups working on related problems. A meeting such as the one we propose will be an ideal opportunity to gather scientists, both with a theoretical as well as experimental background, that are interested and willing to discuss different approaches and issues to make an advance in this relevant field. The long term goal of this workshop is to catalyze interdisciplinary collaborations that integrate our knowledge on different length scales and coarse-grain modelling to address the study of large bio-molecular/polymeric systems and of the design of new functionalized materials.

The key points of the workshop that we propose are the following:
1) Importance of water behaviour (dynamical properties) close to inorganic interfaces, with emphasis on the properties of recent filtration membranes applied in water desalination and sanitation.

2) Accurate description of the solvation of bio-molecules. In particular, during the workshop the current problems and future perspectives of water models on both equilibrium and dynamical properties of proteins will be discussed.

3) Current computational and conceptual challenges related to the ice nucleation and ice inhibition materials, important in fields like cryo-preservation of tissues and frozen food storage, among which proteins play an important role that we will highlight in the workshop.

Oral contribution submitted before the 15th of October 2016 will be considered for addition into the program

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2 Program

Day 1 - Tuesday November 29, 2016

Session 1
- 14:00 to 14:40 - Registration

- 14:40 to 15:25 - **Rahul Raveendran Nair**
  Molecules at two-dimensional interfaces and capillaries

- 15:25 to 16:10 - **Sandrine Lyonnard**
  Water confined in fuel cell membranes

- 16:10 to 16:40 - Coffee Break

- 16:40 to 17:25 - **Laurent Joly**
  Water friction on graphene and boron nitride surfaces: insight from ab initio molecular dynamics

- 17:25 to 18:10 - **Maarten Biesheuvel**
  Water desalination using capacitive electrodes: from carbon nanotube membranes to activated carbon suspensions -- concepts and mean-field transport theory

Day 2 - Wednesday November 30, 2016

Session 2
- 9:00 to 9:45 - **Huib Bakker**
  Structure and dynamics of water molecules at protein surfaces

- 9:45 to 10:30 - **Giancarlo Franzese**
  A multiscale approach for water at bio-nano interfaces

- 10:30 to 11:00 - Coffee Break / Poster Session

- 11:00 to 11:45 - **Valentino Bianco**
  The role of water in the selection of stable proteins at ambient and extreme thermodynamic conditions

- 11:45 to 12:30 - **Giorgio Schirò**
  Unveiling the role of hydration water in the onset of protein structural dynamics

- 12:30 to 15:00 - Lunch

Session 3
- 15:00 to 15:45 - **Biman Bagchi**
  Protein hydration dynamics and hydrophobic force law

- 15:45 to 16:30 - **Xavier Barril**

The quasi-bound state of protein-ligand complexes. implications in drug discovery.

- 16:30 to 17:00 - Coffee Break / Poster Session

- 17:00 to 17:45 - **Max Paoli**
  The role of science for sustainable development

Social dinner
- 20:00 to 23:00 - Social Dinner

Day 3 - Thursday December 1, 2016

Session 4
- 9:00 to 9:45 - **S. Roke**
  Snapshots of ion induced long-range structure in aquous systems

- 9:45 to 10:30 - **Francesco Mallamace**
  An nmr study on the hydrophilic and hydrophobic interactions

- 10:30 to 11:00 - Coffee Break / Poster Session

- 11:00 to 11:45 - **Simone Meloni**
  The salvinia paradox: how the hydrophilic patches help keeping the plant's surface dry

- 11:45 to 12:30 - **Fabio Bruni**
  The putative liquid-liquid transition is a liquid-solid transition in water confined in mcm

- 12:30 to 14:30 - Lunch

Session 5
- 14:30 to 15:15 - **Martin Chaplin**
  Just how anomalous is interfacial water?

- 15:15 to 16:00 - **Othmar Steinhauser**
  Water dynamics near biological surfaces studied by shell-resolved computational spectroscopy

- 16:00 to 16:30 - Coffee Break / Poster Session

- 16:30 to 17:15 - **Martina Havenith**
  Thz spectroscopy and solvation science

- 17:15 to 18:00 - **Ana Vila Verde**
  Supra-additive slowdown of water rotation by outer-sphere ion pairs

Day 4 - Friday December 2, 2016

Session 6

- 9:00 to 9:45 - **Hinrich Grothe**
  Biological ice nucleation in the atmosphere and the biosphere

- 9:45 to 10:30 - **Klaus Liedl**
  Water ordering at protein interfaces and consequences for biomolecular recognition

- 10:30 to 11:00 - Coffee Break

- 11:00 to 11:45 - **Eduardo Sanz**
  The role of the ice-water interface in homogeneous ice nucleation

- 11:45 to 12:30 - **Alexander Bittner**
  Imaging thin water layers on single viruses

- 12:30 to 12:45 - Closing Word

3 Abstracts

Molecules at two-dimensional interfaces and capillaries

Rahul Raveendran Nair

National Graphene Institute and School of Chemical Engineering and Analytical Science, University of Manchester, Manchester, UK

Van der Waals assembly of two-dimensional crystals continue attract intense interestdue to the prospect of designing novel materials with on-demand properties. One ofthe unique features of this technology is the possibility of trapping moleculesbetween two-dimensional crystals. The trapped molecules are predicted toexperience pressures as high as 1 GPa. In my talk, I will discuss the experimentaldemonstration of the huge van der Waals pressure inside the nano-enclosures madefrom 2D crystals. Another feature of van der Waals material assembly is the uniquepossibility of designing two-dimensional channels for molecular transport.Permeation through nanometre-pore materials has been attracting unwaveringinterest due to fundamental differences in governing mechanisms at macroscopicand molecular scales, the importance of water permeation in living systems, andrelevance for filtration and separation techniques. Graphene-based materials canhave well-defined nanometer pores and can exhibit low frictional water flow insidethem, making their properties of interest for filtration and separation. In my talk, I willdiscuss our recent results on molecular and ionic permeation properties of various2D materials based membranes and its prospect for several applications.

Water Confined in Fuel Cell Membranes

Sandrine Lyonnard

CEA-INAC, UMR 5819 (CEA-CNRS-UGA) Grenoble, France

In this talk, we will discuss the role and state of water in hydrogen fuel cells, with particular focus on the dynamical behavior of water (and protons) confined within nanostructured proton-conducting polymer electrolyte membranes (PEMs). The PEM is the key component of low temperature fuel cells, which are one of the most promising technologies for powering small portable devices and automobiles. It acts as a separator between the electrodes, electronic insulator and proton conductor. A microscopic understanding of the relation between the chemical structure of the polymer membrane, its morphology, and the proton transfer is essential to improve fuel cell performances. Benchmark materials are PerfluoroSulfonic Acids (PFSA), composed of a hydrophobic PTFE matrix bearing side-chains terminated by hydrophilic SO_3- groups. In the presence of water, interconnected nanoscale ionic domains are formed upon swelling. The efficiency of proton conduction is directly impacted by the total amount of water and the microscopic properties of the adsorbed molecules. Herein, we will review our recent efforts to probe the water and proton multi-scale dynamics by combining multi-resolution Quasi Elastic Neutron Scattering (QENS) [1,5], PFG-NMR [6] and Molecular Dynamics simulations [7,8]. We demonstrate that the mobility of species is controlled by the balance between short range interactions and connectivity. Confinement at the nano-scale and direct interaction with the interfaces are shown to produce anomalous sub-diffusion within the ionic nano-channels. In conclusion, we elucidate the true nature of water confined in soft ionic nano-structures and provide a general understanding of the complex multi-scale structure-function interplay in advanced materials for use as electrolytes in energy devices.

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Water friction on graphene and boron nitride surfaces: insight from ab initio molecular dynamics

Laurent Joly, Gabriele Tocci, Samy Merabia, Angelos Michaelides
Université Lyon, France

Nanofluidic systems (i.e. natural and artificial systems where fluids are confined at the nanoscale) play a key role in water treatment and sustainable energies, where materials based on carbon and boron nitride (BN) offer promise of radical performance improvement. New behaviors arise in nanoconfined liquids due to the predominant role of surfaces. In particular, friction is the most important interface property that limits nanofluidic transport, and its understanding is therefore crucial for the design of efficient nanofluidic devices. Here, I will illustrate with recent work how ab initio molecular dynamics (AIMD) can help unravel the molecular mechanisms underlying liquid-solid friction.We started by comparing the structure and friction of water on graphene and BN using AIMD [1]. While friction is usually correlated with wetting, we found that although the water/graphene and water/BN interfaces present a very similar structure, the friction of water is ca. 3 times larger on BN than on graphene. We traced back the difference in friction to the greater corrugation of the energy landscape on BN arising from specific electronic structure effects. We then turned to studying the role of defects, which are inevitably present in real nanofluidic systems and play a crucial role in nanofluidic energy harvesting [2]. The simulations show that water dissociates at certain defects, and that these "reactive" defects have a strong influence on friction. Furthermore, friction is extremely sensitive to the chemical structure of reactive defects and to the number of hydrogen bonds they can partake in with the liquid. Overall this work shows the crucial role of surface chemistry on the efficiency of real nanofluidic systems.

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Water desalination using Capacitive Electrodes: from Carbon Nanotube Membranes to Activated Carbon Suspensions -- Concepts and Mean-field Transport Theory

Maarten Biesheuvel
Wetsus, European Centre of Excellence for Sustainable Water Technology

Carbon can desalinate water in several ways. In the first approach, a carbon material with thin pores such as carbon nanotubes (CNTs), with pores of a few nm in diameter, is used to selectively separate water from ions [1-3]. CNTs acquire a surface charge by interaction with water (charging negatively), but can also be electrically addressed so the charge can be changed at will. In the second approach, porous carbon electrodes are cyclically charged and discharged, resulting in the periodic adsorption and desorption of ions [4,5]. Here, one electrode preferentially adsorbs cations, and the other anions. Data for carbon charging and desalination help to validate models of the structure of the electrical double layer in the water and in the carbon [6]. For micropores in activated carbon, the Donnan approach (effectively smearing out the potential) is shown to describe data accurately [6]. We describe experimental approaches for capacitive water desalination using film electrodes as well as electrodes in the form of a slurry and fluidized bed, where carbon beads are pumped around a circuit, charging in one part of the system, and discharging elsewhere [7]. We show how upon contact, oppositely charged beads release salt. Finally we discuss a new method where a single carbon bead falling through an electric field absorbs salt [8].

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Structure and dynamics of water molecules at protein surfaces

Huib Bakker

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Femtosecond nonlinear optical techniques are ideally suited to study the structure and dynamics of water molecules near protein surfaces. We used polarization-resolved femtosecond infrared spectroscopy to study the reorientation dynamics of water molecules near the surface of several globular protein surfaces. [1] We observed that water molecules near the protein surface strongly slowdown in their reorientation in comparison to water molecules in bulk liquid water. When we added an increasing amount of urea to the protein solution, we observed that the water-exposed surface increases by ~50% before the secondary structure of the protein changes. This finding indicates that protein unfolding starts with the protein structure becoming less tight, thereby allowing water to enter. With surface vibrational sum frequency generation (VSFG) spectroscopy, we studied the structure of water at the surface of antifreeze protein III.[2] The measured VSFG spectra showed the presence of ice-like water layers at the ice-binding site of the protein in aqueous solution, at temperatures well above the freezing point. The observations suggest that not the protein itself, but rather the ordered ice-like hydration layers at the protein surface are responsible for the specific recognition and binding to nascent ice crystallites.

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A Multiscale Approach for Water at Bio-Nano Interfaces

Giancarlo Franzese

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I will present recent simulations and theoretical results about the dynamics and structural properties of water at the interface with biomolecules, such as proteins and bio-membranes, and inorganic metals, such as nanoparticles and graphene sheets. By all-atoms simulations I will analyze how hydrophobic and hydrophilic interfaces affect the properties of the vicinal water [1] and we will use the results in a multi scale approach to develop coarse-grained models for water that allow us to extend our investigations to timescales and/or length-scales that would be unreachable in atomistic simulations. We will consider applications to nano confined ice formation, protein folding and design, protein crystallization, self-assembly of bio-membranes and nanoparticles-protein-corona [2-7].

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The role of water in the selection of stable proteins at ambient and extreme thermodynamic conditions

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Proteins that are functional at ambient conditions do not necessarily work at extreme conditions of temperature T and pressure P. Furthermore, there are limits of T and P above which no protein hasa stable functional state. Here we show that these limits and the selection mechanisms for working proteins depend on how the properties of the surrounding water change with T and P. We find that proteins selected at high T are super-stable and are characterized by an optimal segregation of ahydrophilic surface and a hydrophobic core. Surprisingly, a larger segregation reduces the stability range in T and P. Our computer simulations, based on a new protein design protocol, explain the hydropathy profile of proteins as a consequence of evolutionary pressure exerted by water. Our results are potentially useful for engineering proteins and drugs working at extreme conditions.

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Unveiling the Role of Hydration Water in the Onset of Protein Structural Dynamics

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Proteins are the nano-machines of living systems. At the molecular level, proteins need to be covered by water in order to be biologically active. This so-called hydration water is generally acknowledged to enable the internal motions of proteins that are so fundamental for their capacity to fulfill a specific biological function. Yet, the molecular mechanism behind water's importance for protein dynamics has remained elusive. We used a combined approach by neutron scattering and MD simulations on wild type and perdeuterated proteins and on model systems to provide insights into the nature of water thermodynamics and dynamics promoting functional protein motions [1-5]. The same approach on pathological states of a protein related to the Alzheimer disease and on protein-polymer nano-hybrids revealed the importance of a detailed understanding of hydration water dynamics for biological and nanotechnological applications [6-9].

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Protein Hydration Dynamics and Hydrophobic Force Law

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Dynamics of the hydration layer surrounding a protein has drawn intense attention in the last few years, even giving rise to considerable controversies and debates. The central issue seems to be the time scale of relaxation [1-3]. The estimates vary from system to system and are found to depend on the technique used to measure relaxation of the hydration layer.. For example, dielectric relaxation provides a time scale of the order of 40-100 ps while solvation dynamics gives time scales as long as ns. Similarly, uncertainties exist about the width of hydration layer. We shall first provide an overview of the area and subsequently combine simulations performed on different proteins to interrogate these issues. Particular attention shall be paid to the role of amino acid side chains in hydration layer dynamics. We shall even discuss the nature of hydrophobic force experienced by two parallel plates immersed inside water. We find a sharp cross-over in the distance dependence of the hydrophobic force law [4].
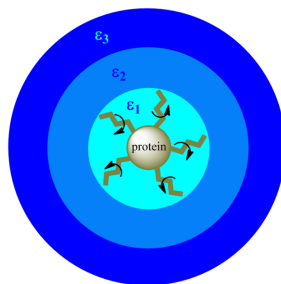
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The Quasi-Bound State of Protein-Ligand Complexes. Implications in Drug Discovery

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The main goal of structure-based drug design is to predict the thermodynamic parameters that define the behaviour of protein-ligand complexes. However, these systems also present a well-defined binding mode, which leads us to postulate that structural stability is a fundamental property of biologically-relevant non-covalent complexes. Based on this hypothesis, we have devised Dynamic Undocking (DUck), a fast computational method to calculate the work necessary to reach a quasi-bound state (WQB), where the ligand has just broken the most important native contact with the receptor. Though WQB is an out of equilibrium property that is irrelevant from a thermodynamic perspective, we find it surprisingly effective in virtual screening because true ligands form more resilient interactions than decoys. Water plays a major role in the process to reach the quasi-bound state, as it involves the rupture of a key intermolecular hydrogen bond. In this talk I will introduce the quasi-bound concept, demonstrate the effectiveness of DUck to discover new ligands and illustrate the fundamental role of re-hydration in the early stages of protein-ligand dissociation.[1]

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The Role of Science for Sustainable Development

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Human societies worldwide are facing global challenges that are affecting life on Earth. The United Nations' new Sustainable Development Goals (SDGs) represent the best overall attempt to date to bring to the forefront of international agendas key issues such as population growth, energy demands, food security, climate adaptation, environmental degradation, biodiversity loss, water and health. Because most people appear to be "carrying on as usual", raising awareness and education are critical to achieve a change of mind/behavior. Scientists can and must take a proactive role because of their involvement in academia and research. In particular, they ought to strive to communicate whenever possible with both policy makers as well as the wider lay audience. Sustainability-related sciences can provide solutions but, if not implemented, these will remain examples of human ingenuity and not serve their full real purpose. A shift in direction may be happening with many funding bodies such that there is increasing support for projects focusing on sustainable development and on building the basis for a more sustainable future.

Snapshots of Ion Induced Long-range Structure in Aquous Systems

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Ions and ionic groups interact with water in many ways, changing dipole orientation, inducing charge transfer, and distorting the hydrogen-bonding network. These effects have been studied in experiments probing the vibrational dynamics, THz time domain response, and dielectric response, as well as in computer simulations. The ions' influence is shown to be short-range, affecting the structure of water in the first, second and at most the third hydration shell. However, such experiments and simulations are biased towards detecting short-range perturbations. Here, we probe the interaction of ions with water with elastic second harmonic scattering, a background-free method that is biased towards ordering effects on the nanometer length scale. We observe the ions of 20 different electrolytes inducing long-range structural

perturbations that are interpreted as an increase in the orienational order of in the H-bonding network. The measured perturbations appear at ionic concentrations as low as

An NMR study on the hydrophilic and hydrophobic interactions

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Using nuclear magnetic resonance (NMR) we study the dynamics of the hydrogen bond (HB) sub-domains in bulk and emulsified water across a wide temperature range that includes the super-cooled regime. We measure the proton spin-lattice Tâ, and spin-spin Tâ,, relaxation times to understand the hydrophilic interactions that determine the properties of water. We use (i) the Bloembergen, Purcell and Pound (BPP) approach that focuses on a characteristic correlation time tC, and (ii) the Powles and Hubbard (PH) approach that measures the proton rotational time tR. We find that when the temperature is low both relaxation times are strongly correlated when the HB lifetime is long, and that when the temperature is high a decrease in the HB lifetime destroys the water clusters and decouples the dynamic modes of the system. Lately in the same theoretical approaches we study pure bulk methanol and the corresponding water solutions in a large concentration range (methanol molar fractions) from the stable to the super-cooled regimes. Our findings are: i) a separate characterization of the hydrophilic and hydrophobic interactions and the corresponding dynamics; ii) whereas the hydrophilicity dominates the solution chemical physics in the water super-cooled phase, the hydrophobicity governs it by increasing temperature from a crossover value located at about 265K, where the HB lifetime is of the order of some picoseconds. Such a situation is of strong relevance for more complex materials such as polymeric systems and biological macromolecules like peptides and proteins especially for their thermal configuration changes.

The Salvinia Paradox: how the hydrophilic patches help keeping the plant's surface dry

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Plants and animals have developed very diverse strategies to prevent the wetting by water and other liquids. Very interesting is the case of Salvinia Molesta, which leaves are decorates with hydrophobic eggbeater-like hairs. What makes special this case is the fact that these hairs feature hydrophilic patches at their top [1]. One question to address is how these hydrophilic patches help keeping non-wet the surface of this plant. We investigated this problem by combining atomistic simulations and continuum models [2]. Focusing on a simplified system, we show that this combination of hydrophobic and hydrophilic domains improves the stability of the contact between the liquid/air interafce and the top of the hairs. This is achieved through a modification of the free energy profile of the wetting/cavitation process from the hydrophilic patches. In practice, the liquid/air interface stays pinned at chemical or geometrical discontinuities, which helps plants resisting to changes of thermodynamic and mechanical conditions, such as local changes of temperature and pressure, or the action of external forces, which can trigger the wetting of the surface.A continuum theory interpreting atomistic results is used to design surfaces of improved stability. Our findings suggest possible routes to design surfaces of interest for technological applications requiring non-wettable submerged surfaces: energy related applications [3], marine applications [4], medical application [5], and many others.

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The putative liquid-liquid transition is a liquid-solid transition in water confined in MCM

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Neutron diffraction experiment have been performed on water confined in MCM-41/C10 (pore size 2.8 nm) as a function of temperature, in the range 165 - 300 K. TheNIMROD diffractometer used allows to investigate a wide Q range, extending from10^{-2} to 30 Å^{-1} ; this is of particular relevance, as it is possible to look at the sametime at the peak (around 0.21 Å^{-1}) resulting from the (10) plane of the2D hexagonal arrangement of the water cylinders in the silica matrix and at waterpeak (around 1.7 Å^{-1}). The intensity of the first peak has been taken as an indicatorof the average mass density of water in the pores, and the observed intensitychanges as a function of temperature as an indication of a liquid-liquid transitionbelow 210 K. The shape of the water peak, at around 1.7 Å^{-1}, suggests theformation of hexagonal ice below 240 K, as shown by the characteristic structurefactor features typically present in crystalline water. Based on these observations theproposed liquid-liquid transition, determined with small angle neutron scatteringtechnique , is really a crystallization transition of at least a fraction of watermolecules confined in the MCM material.

Just how anomalous is interfacial water?

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Liquid water is commonly cited as being a most anomalous liquid. Although it is an apparently simple molecule (H2O), it has a highly complex and anomalous character due primarily to its inter-molecular hydrogen bonding. Notable amongst the anomalies of water are the opposite properties of hot and cold water. As cold liquid water is heated individual molecules shrink, bulk water shrinks and becomes less easy to compress, its refractive index increases, the speed of sound within it increases, gases become less soluble and it is easier to heat and conducts heat better. In contrast as hot liquid water is heated it expands, it becomes easier to compress, its refractive index reduces, the speed of sound within it decreases, gases become more soluble and it is harder to heat and a poorer conductor of heat. With increasing pressure, individual molecules expand, cold water molecules move faster but hot water molecules move slower. These properties can be explained if liquid water consists of a two-state mixture of differently hydrogen-bonded water clusters, and such two-state models for water are becoming widely accepted.Investigations of water at interfaces often treat water as a molecular substance using models that are incapable of showing dissociation, electron delocalisation or that it consists of rapidly equilibrating extensively hydrogen bonded chains and clusters. Because of this they are generally incapable of showing the breadth of liquid water's anomalies even if especially designed to fit with one or two of them.The curiosity that the interface studies generally show no anomalies, suggested the script below from Sir Arthur Conan Doyle's short story "Silver Blaze" and is the subject of this paper.Scotland Yard detective: "Is there any other point to which you would wish to draw my attention?"Sherlock Holmes: "To the curious incident of the dog in the night-time."Scotland Yard detective: "The dog did nothing in the night-time."Sherlock Holmes: "That was the curious incident."Also is it important or relevant that many studies of the aqueous interface use methods that are highly localised when the effects of the interface can probably extend over depths of several micrometres?Important papers:

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Water Dynamics near biological surfaces studied by Shell-Resolved Computational Spectroscopy

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Dynamics, i.e. the time evolution of molecular motion, has itsexperimental counterpart in spectroscopy. Originally developed forisolated molecules and the gas phase spectroscopy of bulk mediahas achieved considerable progress during last fifteen years.Nevertheless, the spectra contain complex information composedof various contributions. Computational spectroscopy based onsimulation can disentangle these contributions. Even more, nearsurfaces a shell-resolved decomposition is possible revealing therange of influence of the surface.Altogether we are interpreting four types experimentalspectroscopy: Nuclear Quadrupole Relaxation NQR, NuclearMagnetic Relaxation (NMR), Dielectric Relaxation (DRS) andStokes-Shift Spectroscopy. For the present talk we have selectedNQR and DRS for a shell-resolved interpretation. Concerning thesurfaces we have selected an outer, essentially convex proteinsurface as well as the inner, concave surface of a reverse micelle.

THz spectroscopy and Solvation Science

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The majority of chemical reactions – among those many that are central to important industrial processes – and virtually all biological processes, take place in a liquid-state environment. Solvents – with water being the most prominent – are used to "solvate" molecular species from reagents to proteins and thereby transfer these as "solutes" into the liquid state. Understanding "the role of water in the myriad of processes – from catalysis to molecular recognition "was addressed as one of the main challenges for chemistry in next century. Now, we witness the emergence of Solvation Science as a new interdisciplinary field to understand the influence of solvation on reactions, the function of biomolecules, and processes at liquid-solid interfaces. Water's flexible network enables it to adapt its structure and dynamics. Hydration water makes significant contributions to the structure and energy of proteins and provides a responsive surrounding which allows for conformational changes. In particular, water may hold the key to the way proteins interact, fold, bind substrates, and aggregate. Water at protein interfaces (hydration water or interfacial water) has been shown to thermodynamically stabilize the native structure of bio-macromolecules, to affect protein flexibility, and to contribute to molecular recognition in enzyme catalysis. Protein-water interactions are now known to shape the "free energy folding funnel" that drives protein folding. We could show that THz absorption spectroscopy is a powerful tool to probe hydration dynamics of biomolecules. Under ambient, physiologically relevant conditions, 90% of the modes which contribute to the total entropy of the solvated protein are captured by the low frequency modes of the protein/solvent, i.e. the vibrational density of states (VDOS) between 0 and 10 THz (300cm-1). I will present examples for low frequency spectra of hydration water around solutes and explain how these provide sensitive probes of hydration dynamics. THz calorimetry will be introduced as a new tool for water mapping which gives access to spatially resolved values of ΔCp, ΔS, ΔH and ΔG. Transient THz spectroscopy can be used to record snapshots of the low frequency spectrum of a solvated proteins subsequent to initiation of the protein folding, thus capturing changes during hydrophobic collapse. We propose that water is not just a passive spectator solvent in biological processes, but has a vital function in most biomolecular and cellular processes.

Supra-additive slowdown of water rotation by outer-sphere ion pairs

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Results from femtosecond infra-red spectroscopy experiments have suggested that some ions (e.g., magnesium, sulfate) supra-additively slow down the rotation of water molecules much beyond their first hydration shell. This suggestion is challenged by results from other experiments and simulations, which do not see marked effects of ions on water dynamics beyond the ions' first hydration shell, and which suggest that these effects are mostly additive. To resolve this controversy, we use molecular dynamics simulations and classical, polarizable, models, to investigate water dynamics in solutions of salts of high and low charge density: MgSO4 and CsCl, respectively. The models reproduce the free energy of hydration of the ions at infinite dilution and the activity derivative of solutions at 2.5 m concentration, so they adequately capture both ion-water and ion-ion interactions. Our results [1,2] suggest that static solvent-shared ion pairs of CsCl may already weakly alter water rotation relative to that observed near isolated ions. However, the average water dynamics in CsCl solutions up to 2.5 m can still be understood as the sum of contributions of water near isolated ions, because the perturbation induced in water dynamics by the ion pairs is small and these pairs are short-lived. In contrast, solvent-shared ion pairs of MgSO4 can intensely slow down water rotation, with supra-additive effects being observed for the water molecules that simultaneously belong to the first hydration layer of both ions. Water rotational dynamics in MgSO4 solutions of concentration near 1.5 m includes sizeable contributions from non-additive effects by ion pairs; at concentrations near the solubility limit, even slower water dynamics arises because of non-additive effects in ion clusters. These results show that the concentration of solvent-shared ion pairs - a static property - directly affects the water dynamics of a solution.

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Biological Ice Nucleation in the Atmosphere and the Biosphere

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From the thermodynamic point of view, ice and snow can form already at temperatures slightly below its melting point, i.e. below zero degrees Celsius. Actually, ultrapure, liquid water can be supercooled down to minus forty degrees Celsius without freezing. The reason is a kinetic activation barrier, which hinders the phase transition. However, water impurities, e.g. biological material or organic particles, can lower the activation barrier and can thus catalyze the phase transition. This process is called heterogeneous ice nucleation and it plays an important role in many biological, meteorological and technical processes, e.g. in the formation of atmospheric ice clouds. The most effective ice nucleus is ice itself, since it provides the own hexagonal structure, on which water molecules from the liquid phase can be oriented in order to form further ice phase. The most effective heterogeneous ice nucleus is the bacterium Pseudomonas Syringae. The reason is a protein at its outer cell membrane, which exhibits a hexagonal, ice-like structure. Furthermore, fungal spores, pollen, and carbonaceous particles are also very effective ice nuclei [1, 2]. In many cases, the physical and chemical reasons for the ice nucleation activity are understood only rudimentary. Thus, the search for the perfect ice nucleus is still an open issue [3].The talk will explain the fundamentals of heterogeneous ice nucleation and will give examples from the field and the laboratory.

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Water Ordering at Protein Interfaces and Consequences for Biomolecular Recognition

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Water molecules at interfaces play decisive roles in biomolecular recognition. We investigated two rather different protein interfaces and the roles of water molecules at these interfaces.On the one hand we studied the recognition of ice by proteins, which is of major importance for ice nucleation proteins as well as for anti freezing proteins. The recognition of ice depends on a layer of water molecules at the protein interface. We developed a hydrophobicity scale, which calculates hydrophobicity from enthalpic and entropic contributions directly from the phase space occupied by these water molecules. This method also allows for the definition of localized hydrophobicity on a biomolecular surface. Surprisingly we find a rather weakly bound and disordered layer of water molecules to be optimal for ice binding. This layer seems to mediate between the ice structure and the protein surface.On the other hand we investigated the recognition of peptides by proteases. We developed a method to quantitatively localize the specificity and promiscuity of this recognition processes in the binding interface of proteases. In this way we can study these localized regions of specificity and promiscuity in the binding interface and analyze their thermodynamic properties. Differences in specificity and promiscuity obviously originate from differences in free energy of binding. Surprisingly we find little correlation of specificity with enthalpy and enthalpy related quantities. Especially we find neither strong correlation with the binding side potential nor with the enthalpy of water molecules in the first solvation layer. In contrast, we find strong correlations of specificity and promiscuity with rigidity and flexibility of the binding pocket. This correlation translates into ordering processes of water molecules in the interface. Regions of specificity show ordered water molecules in the interface, whereas regions of promiscuity tend to have more disordered water molecules in the first solvation sphere.

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The Role of the Ice-water Interface in Homogeneous Ice Nucleation

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The avoidance of water freezing is the holy grail in the cryopreservation ofbiologycal samples,

food and organs [1]. Fast cooling rates are used to beatice nucleation and avoid cell damage. This strategy can be enhanced byapplying high pressures to decrease the nucleation rate [2], but the physics behindthis approach has not been fully understood yet. I present a computersimulation study to investigate ice nucleation at high pressures using acombination of simulation methods to obtain nucleation rates and interfacialfree energies [3,4,5]. We find that the slowing down of the nucleation rate ismainly due to an increase of the ice I-water interfacial free energy withpressure [6]. Our work also clarifies the molecular mechanism of icenucleation for a wide pressure range. This study is not only relevant tocryopreservation, but also to water amorphization and climate change modelling [7].

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Imaging thin water layers on single viruses

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The interaction of water with the coat proteins of viruses is key to understanding virus transmission and infectivity. The first step towards understanding more complex viruses is investigating a simple case: The well-characterised Tobacco Mosaic Virus (TMV). Its highly regular helical coat protein arrangement is a textbook example for perfect self-assembly, but was as yet probed by methods that average over many particles (XRD, cryoEM). Local probing of single particles can be achieved with AFM and low voltage SEM, which show virus particles in the dry state with unexpected nanoscale surface features at irregular axial spacings. It is as yet unknown whether such morphological changes are reversible, and whether they occur also in other viruses. The stepwise wetting by water can be studied by AFM in a humidity chamber, and by environmental electron microscopy (SEM and STEM) in up to 10 mbar water. The observed scenarios comprise wet wedges (around TMV), layers (on TMV), and water pools (confined by TMV). Except for ultrathin layers (≥1 nm), the water topography is completely compatible with macroscale wetting scenarios.

4 Posters

The role of water in the beginnings of material-driven fibronectin fibrillogenesis

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Fibronectin (FN), an extracellular matrix protein, is known to undergo important quaternary conformational changes upon adhesion to the substrate made of ethyl acrylate (EA) polymer chains [1]. Surprisingly, an almost identical polymer composed of methyl acrylate (MA) chains, which differs by a single carbon, does not trigger the transition. This conformational transition is necessary to start FN fibrillogenesis or self-association which influences several processes including cell differentiation and migration [2]. Understanding the cause of the conformational transition will aid the design of better materials to drive fibrillogenesis, which has potential applications in tissue engineering and cell culture [3]. In order to understand the difference in interactions between the EA and MA functional groups and fibronectin, we have used molecular dynamics simulations of self-assembled monolayers (SAMs), which have been functionalised with the EA or MA side chains. In our simulations, we investigate the interaction of two crucial domains, FNIII 9-10 that are key to the process [1], with these two surfaces. In doing so, we have set out to investigate first whether or not we find evidence which would be consistent with the experimental observation from Prof. Salmeron-Sanchez's group that fibronectin forms fibrils when in contact with EA functionalised SAMs but not when in contact with MA functionalised SAMs. Additionally, we aim to understand the interactions that are unique to the EA functionalised and MA functionalised systems, and how these interactions may affect fibrillogenesis. Our initial results are consistent with the latest experimental results. Water, described by the TIP3P model [4], is suspected to play an important role in the adhesion and consequently in the FN transition. The radial distribution function indicates a higher water density around SAM MA and we are currently analysing the spatial preference of the water around the functional groups. Our aim is to find out if the structure of water around the functional groups acts as a barrier to adhesion in the case of MA side chain.

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VASP Multigrid Continuum Model (VASP-MGCM): An Implicit Solvation Model for Density Functional Theory Calculations

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Continuum models have become a widespread technique for the study of solvent effects in Density Functional Theory (DFT) methods [1]. Adding solvation contributions relies on the calculation of the electrostatic potential of the system through the solution of the Generalized Poisson Equation (GPE). In this context, multigrid methods are especially appropriate for the solution of partial differential equations, but their use, up to now, is not much extended in DFT-based codes because of their high memory demand. We have implemented a multigrid solver-

based approach for the treatment of solvation effects in the Vienna ab initio Simulation Package (VASP) [2]. The stated implicit solvation model, named VASP-MGCM (VASP-Multigrid Continuum Model) [3], uses an efficient and transferable algorithm for the product of sparse matrices that highly outperforms serial multigrid solvers. The model has been validated using a large set of neutral and ionic species, as well as adsorbed molecules on metallic surfaces, whose solvation energies in water have been compared to both experimental and simulation data. Once validated, the present model has been applied to the study of liquid-solid reactions of interest in industrial and energy-related problems [4].

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Low temperature friction of water on a crystalline surface

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Switching from macroscopic to nanoscopic scale in hydrodynamics gives rise to striking properties like fast transport or nonlinear electrokinetic effects, which open up new perspectives in the development of devices and sensors at nanoscale [Bocquet & Tabeling (2014), Abgrall & Nguyen (2008), Sparreboom et al. (2009)]. Generally speaking, sensors are usually used with various temperature conditions. However, to this day, one knows very few about the evolution of interfacial water properties with temperature. In the present work, we analyse by classical molecular dynamics water friction in the vicinity of a Lennard-Jones crystal surface from 225 to 360 K, using the TIP4P/2005f forcefield [Gonzalez et al. (2011)], that we recently proved to be very efficient to reproduce bulk transport properties (shear viscosity, self-diffusion) over this range of temperatures~[submitted]. Surprisingly, non-equilibrium shear calculations reveal a non-monotonic evolution of the friction with temperature, whereas equilibrium calculations within the Green-Kubo formalism [Bocquet & Barrat (2007)] show a monotonous decrease of this quantity with the temperature. Previous studies on graphene and carbon nanotubes showed a very good agreement between both methods at room temperature [Falk et al. (2010)]. With the crystaline surface used in the present work, the two methods matches only at high temperatures. This means the Green-Kubo formalism might not be valid at low temperatures, with a possible phase transition of the surface water. Calculations are currently under way to control this hypothesis.

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Cavitation in water confined in nanotubes

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In this contribution, we study via the nucleation of a vapor cavity inside nanometer-sized hydrophobic pores. The extreme confinement of water within these pores promotes spontaneous cavitation even at positive liquid pressures, a phenomenon which cannot be explained solely via the classical nucleation theory. The current interpretation of such experimental finding invokes the contribution of a negative line tension [1]. In this context, we employ molecular dynamics to obtain molecular-level insights into nanoconfined cavitation and develop a theoretical description going beyond classical nucleation theory. The water model used in simulations is TIP4P/2005 [2], which captures the most important features of liquid water and interfaces. In order to reconstruct the free energy barriers and the cavitation mechanism we employ a specialized rare event technique, the string method in collective variables [3]. By employing the coarse-grained density field in order to describe this thermally activated process, we avoid simulation artifacts due to the use of a single collective variable, which is one of the fundamental assumptions of classical nucleation theory [4]. Results demonstrate the existence of several cavitation pathways and indicate deviations from both the classical picture and from its extension based on line tension.

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Exploring the role of interfacial water layer in the bio-nano assembly of an integrated protein/graphene interface

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Bio-nanotechnology has advanced to the point at which it can now focus on understanding and controlling bio-nano interface interactions. The basic biomolecule and the star nanomaterial, protein and graphene, play significant roles in the revolution of science and industry. Water is the universal solvent in biological systems and also the substance with the largest number of anomalies in its properties. Understanding the behavior of interfacial water between a protein and graphene is fundamental to our ability to regulate protein and graphene interactions.

Using molecular dynamics simulations, we investigated a bio-nano interface consisting of a β-sheet antifreeze protein stacked onto the extended surface of graphene [1]. In this setup, the behaviors of an integrated interfacial water layer would be observed during the bio-nano assembly process. In contrary to the enhanced hydrogen-bonds (HBs) network of the interlayer water confined between two hydrophobic graphene surfaces [2], we found that the intermolecular coupling of interlayer water would be relaxed by the relative motion of protein upon graphene. Meantime, the interlayer water filled within interstices of the bio-nano interface would suppress the molecular vibration of surface groups on protein, and would impair the CH…π interaction driving the attraction of the protein and graphene. From the transition of the intermolecular coupling between protein-bound and unbound water at temperatures of 300 K and 250

K, we found that the CH…π attraction between protein and graphene was regulated by the HBs between water molecules in the interlayer. On the other hand, the involvement of interlayer water also contributed to a larger walking range of the protein on graphene, to search for a suitable binding status with lower free energy traps. Therefore, the formation of a bio-nano interface by the adsorption of the bulk β-sheet proteins on hydrophobic graphene surface in water environment will be regulated by the hindrance of interlayer water in both aspects of their interfacial interactions and their assembly process. This finding should be lawful for the assembly of other bulk proteins owning a stable structure on graphene surface because most of the stable proteins in water have a strong coupling with water molecules on their surfaces.

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The interplay of water, cationic protein sites and polyatomic anions depends critically on sodium

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Polyatomic ions such as sulfates, phosphates or sulfonates are key players in biological processes but the molecular mechanisms by which these ions act are currently incompletely understood. We use molecular simulations and classical, atomistic models with fixed-charge and explicit solvent representation to clarify the molecular mechanisms of interaction between cationic amino acids and sulfates, phosphates and sulfonates, in their methylated and non-methylated forms and in the presence of excess counterions. This works goes beyond prior reports on the topic in that it uses a newly developed, internally consistent force field for all ions, which correctly captures the energy magnitude and length scale of anion-cation interactions. Our results suggest a possible molecular origin of previously unexplained experimental observations [1]: anions that, according to experiment, differ strongly in the magnitude of their interaction with cationic amino acids have in fact very similar interactions with those amino acids, but different interactions with sodium; the presence of excess sodium in the experiments thus determines the experimental outcome due to competition with the cationic sites on the protein.

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Water dynamics depends strongly on curvature for small hydrophobic molecules but not for hydrophobic protein sites

Ana Vila Verde, João Robalo

The dynamics of waters of hydration in proteins impacts the kinetics of protein folding and function. Achieving control over the dynamics of waters of hydration is thus a promising avenue to optimize proteins for biomedical and biotechnological applications. Recent experimental results [1] suggest that fluorination – replacing C-H by C-F groups – can be a useful tool to control the dynamics of water near proteins, but the molecular scale origin of the experimentally observed trends is not yet understood and the

general applicability of this procedure is unproven. We use molecular simulations and classical, atomistic, fixed-charged models to systematically investigate how changes in solute polarity and curvature brought by fluorination alter the structure and dynamics of water around small hydrophobic molecules and around the side chains of hydrophobic amino acids in proteins. Our results indicate that fluorination of small hydrophobic molecules markedly slows down their waters of hydration, and that this effect results from the different curvature of fluorinated vs. non-fluorinated groups instead of, as suggested by others, from changes in polarity. In contrast, water dynamics around hydrophobic amino acids in proteins is largely insensitive to fluorination because the effect of nearby protein backbone and neighboring amino acids on water dynamics overcomes the effect of fluorination. Our results indicate that the experimentally measured effect of protein fluorination on the dynamics of waters of hydration result from an interaction between the fluorinated amino acids and their immediate neighbors that strongly depends on the details of protein conformation and composition, rather than being a generalized effect arising from the interactions between water and fluorinated sites.

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5 Participant List

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6 Venue

The workshop will be held at the [Erwin Schrödinger Institute](#)

From the airport the cheapest way to reach the faculty is the **S7** train and get down at **Handelskai** U-Bahn (metro) station. From there take the **U6** line direction **Siebenhirten** and exit at **Währinger Straße-Volksoper**. At that point you can either walk for about 500 m or take the 40, 41 or 42 tramline direction
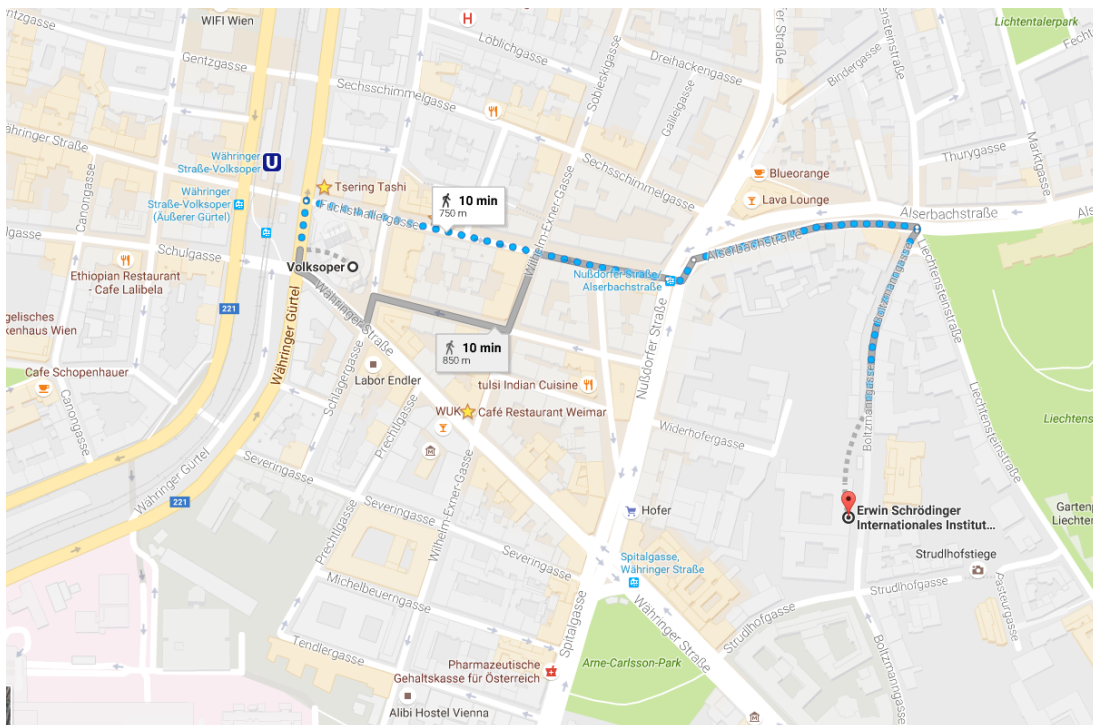
Schottentor and exit at the second stop. The cost will be around 5 Euros.

Alternatively you can get a taxi (get it inside not outside) for about 35/40 euros. Maybe you have the [car2go](#) subscription, if so remember that it works also in Vienna and it will cost you around 10 euros to drive to the University.

Coffee breaks and poster sessions will take place at institute.

7 Hotels

The following hotels that are close to the venue of the workshop:

- **Boltzmann Hotel:**
  http://www.hotelboltzmann.at/

- **Hotel Harmonie:**
  http://www.tripadvisor.com/Hotel_Review-g190454-d227177-Reviews-BEST_WESTERN_Hotel_Harmonie-Vienna.html

- **Appartement Pension 700m zum Ring:**
  http://www.booking.com/hotel/at/700mzumring.en.html

- **Hotel & Palais Strudlhof:**
  http://www.sotour.at/wien/hotel-palais-strudlhof/

- **Ibis Styles Hotel:**
  http://ibisstyleswien.com/en/

Also good sources are websites such as:
- http://www.tripadvisor.com/
- http://www.booking.com/
- http://www.venere.it/
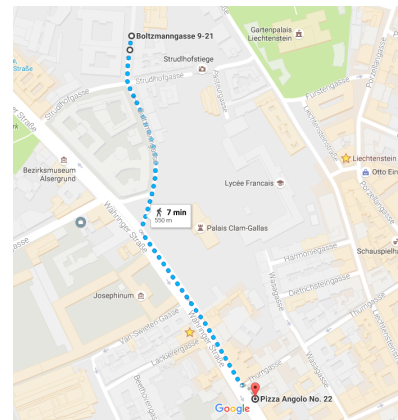- https://www.airbnb.com

8 Lunch

Lunch options located near the faculty of physics are available at the addresses below. In addition there are several places that offer quicker options like sandwiches or take away.
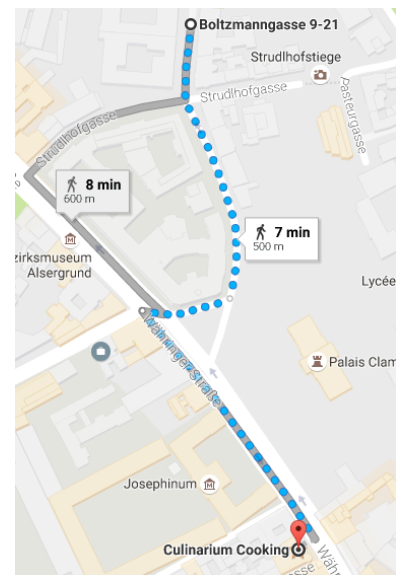

- **Hotel & Palais Strudlhof** Pasteurgasse 1
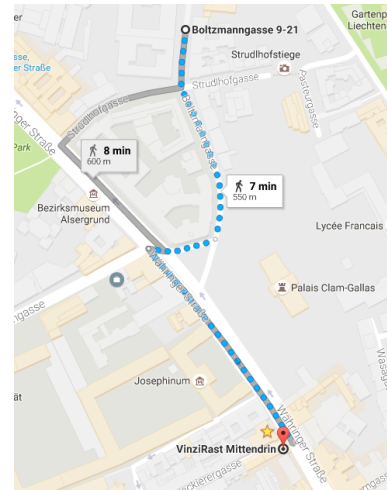 (Austrian ~20 Euros)
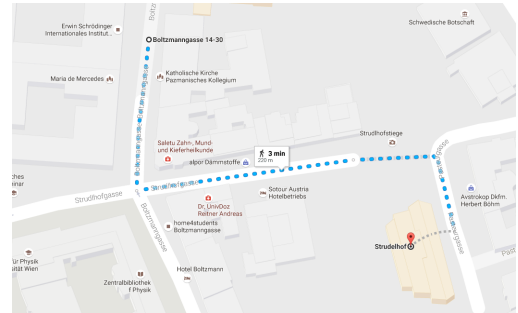
- **Pizzeria Angolo 22** Währinger Straße 22
  (Italian ~10 Euros)

- **Culinarium Cooking** Währinger Strasse 9
  (Asian ~ 10 Euros)

- **MIttendrin** Währinger Strasse 19
  (European ~ 10 Euros)

- **Flein** Boltzmanngasse 2
  (European ~ 20 Euro)