Abstract Book

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Brillouin and Neutrons: Assessing New Advances

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THz excitations in disordered condensed matter and in biological and magnetic systems

Neutron and photon inelastic scattering at low Q
Modeling
Simulation techniques
Instrumentation
Perspectives

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Orals
Introduction to and some history of Neutron Brillouin Scattering

Jens-Boie Suck

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In the first part of the talk an introduction into the conditions and challenges of Neutron Brillouin Scattering will be given. In the second part a brief look at the history of Neutron Brillouin Scattering with some weight on the instrumental development, leading to the building of the BRISP spectrometer at the High Flux Reactor of the Institut Laue-Langevin, will be presented.

Brillouin scattering using X-rays: examples from recent experiments

Giulio Monaco

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A selection of results obtained using synchrotron-based inelastic X-ray scattering with meV energy resolution to study the high-frequency vibrational dynamics in glasses will be here reviewed. The combination of momentum-resolved and momentum-integrated experiments allows probing in detail both the vibrational density of states and the density-density fluctuations down to energies of the order of ~1 meV, i.e. down to the energy range corresponding to the low-temperature thermal anomalies of glasses. These results provide compelling evidence of a transition in the glass from the isotropic elastic response at long wavelengths to a microscopic regime as the wavelength decreases below a characteristic length of a few nanometers, corresponding to about 20 inter-atomic distances. In the microscopic regime the glass vibrations closely resemble those of the corresponding polycrystal, with excitations related to the acoustic and optic modes of the crystal.

More recent applications of inelastic X-ray scattering at resonance conditions to probe magnetic excitations in correlated electron systems will be also discussed, highlighting in particular their complementarity to neutron scattering experiments.

Finally, the emerging possibilities for inelastic X-ray scattering experiments at free electron laser sources will be briefly presented, therefore clarifying why they are expected to open new scientific areas in the near future.
An alternative theory for time dependent correlations: recurrence relation and the exponential expansion

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We discuss the general theory of time correlation functions and their spectra as given by Mori and Zwanzig, and present an alternative formulation based on recurrence relations of new basis vectors in the Hilbert space of the analyzed property. We also show that, in a very general way, spectra can be decomposed in series of Lorentzian functions. Consequently, the correlation functions can always, on a theoretical basis, be represented by a series of exponentials. Examples of application of the theory are given.

A perspective on scattering by monoatomic liquids: deep down, are they all the same?

Wouter Montfrooij

University of Missouri, USA

We will review how disturbances in monoatomic fluids relax back to equilibrium starting with the most simple liquid, superfluid helium, and proceeding to the most complex ones, namely liquid metals. We discuss the (unexpected) commonalities between all of these liquids, and we focus on some decades-old open questions, as well as on some new ones. We also review methods of analysis for scattering spectra and computer simulations, discuss how to avoid common pitfalls in data analysis, and how to test models down to the 1%-level of agreement.

Transverse modes in the spectra of density fluctuations of simple liquids.

Renzo Vallauri

ISC-CNR Sesto Fiorentino, Italy

We report the results of transverse and longitudinal current correlation functions evaluated in liquid and amorphous sodium by computer simulation. The analysis of the corresponding spectra allows the evaluation of a dispersion curve position as a function of the wavevector. The results are compared with the experimental findings of Giordano-Monaco (PNAS \textbf{107}, 21985 (2010)), obtained by new analysis of $S(k,\omega)$ measured by inelastic X-Ray scattering in liquid and polycrystalline sodium. A substantial agreement between experimental and computer simulation results is found. We studied also the behaviour of the line width in terms of the wavevector which gives hints for the correct interpretation of the experimental results. An attempt of a theoretical explanation of the whole set of data is performed by making use of the Vibration-Transit theory developed for the study of the dynamical processes in simple liquids.
Liquid Au studied by neutron Brillouin scattering and \textit{ab initio} simulations: an insight on collective and self dynamics

**Stefano Bellissima**

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We performed a neutron Brillouin scattering determination of the dynamic structure factor of liquid gold in the wave-vector range \(6 \text{ nm}^{-1} < Q < 16 \text{ nm}^{-1}\), accessed by the Brisp spectrometer at ILL. Despite the experimental difficulties due to the considerable neutron absorption and the high melting temperature of the sample, a non-negligible coherent signal could successfully be extracted and revealed the presence of underdamped ion density-fluctuation modes in the whole \(Q\) range of the experiment. The quality of the data further enabled a significant comparison with the results of \textit{ab initio} simulations for liquid gold. The agreement found between neutron and simulation data not only provides a necessary test of \textit{ab initio} methods still limited to the use of few hundreds atoms, but also reasonably justifies a thorough analysis of the simulated dynamics in the extended \(Q\) range that can be more comfortably accessed by calculations. A viscoelastic modeling of the simulated spectra proves to be appropriate and, surprisingly, the so-derived salient dynamic features of liquid gold show an overall \(Q\) behavior globally similar to that already found for insulating liquids. In addition to the analysis of the coherent spectra, we also applied a recently proposed general theory for the functionality of correlation functions to the study of simulated self spectra, which are successfully represented in terms of real/complex lorentzian modes. We also show how self spectra permit a reliable determination of the modes that describe the velocity autocorrelation spectrum \(Z(\omega)\), which is related to the vibrational density of states of the fluid.

**Force matching: a method to obtain local pseudopotentials for complex systems.**

**Beatriz G. del Rio** and **Luis E. González**

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The first principles calculations along with Molecular Dynamics (MD) simulation techniques have become an essential way to study different properties of condensed matter. Most of the first principles methods are based on the Density Functional Theory. Since the microscopic details of the orbital are involved in the Kohn-Sham \textit{ab initio} molecular dynamics (KS-AIMD), calculations are computationally very expensive, and this limits the size of the systems (one or two hundred particles) under study. However, some of these constraints may be greatly reduced by the so called orbital free \textit{ab initio} MD (OF-AIMD) simulation method. In the OF-AIMD method the electronic orbitals of KS formulation are disposed of by using only the valence electron density which greatly reduces the number of variables describing the electronic state, and thus enables to study larger samples (tens of thousands of particles) and for longer simulation time (tens of picoseconds).

A key ingredient in the OF-AIMD method is the local ionic pseudopotential describing the ion-valence electrons interaction. We have developed a forcematching method [1] to derive a local ionic pseudopotential suitable to account for complex systems, such as a free liquid surface or transition metals, for which no adequate local pseudopotentials were available.

Simulation of moderately quantum fluids

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Fluids such as neon and hydrogen/deuterium above the triple point, and helium at not too low temperatures, are often categorized as "moderately quantum mechanical" in the sense that, although the constituent particles may still be assumed to obey Boltzmann statistics, a purely classical or even semiclassical treatment is out of the question. As far as the prediction of static properties of these systems by simulation is concerned, Path Integral Monte Carlo (PIMC) is in principle a rigorous numerical method. In the so-called classical isomorphism, each quantum particle is replaced by a classical ring polymer of P copies of itself, and as the Trotter number P goes to infinity, the exact quantum mechanical results are recovered. With respect to dynamical properties, the situation is much less satisfactory, especially if one is interested in simulating realistic many-body systems. Of the half a dozen or a dozen approximate methods that have been proposed, we will here focus on three that are based on the integration of classical equations of motion in some kind of quantum mechanical force field. In Centroid Molecular Dynamics (CMD) it is the centers (centroids) of the ring polymers that move classically, and the forces on the centroids are given by the average force on a polymer if a PIMC simulation were done with fixed polymer centers at each time step. Ring Polymer Molecular Dynamics (RPMD) goes one step further in that it is now the individual monomers that are propagated classically under the influence of the (classical) intra- as well as inter-polymer forces. Unfortunately, the applicability of both CMD and RPMD is limited to the calculation of time correlation functions of operators that are linear in position or momentum. Thus the only straightforward way to simulate the self intermediate scattering function is via the velocity correlation function and the Gaussian approximation. A very recently proposed alternative that may overcome the linearity problem is the Feynman-Kleinert Quasi-Classical Wigner approach (FKQCW). As with CMD, the centroids evolve in an effective force field now given by the self-consistently smeared quadratic approximation to the instantaneous n-body potential. However, they are now accompanied by a bunch of "satellites" whose (classical) equations of motion are formulated in terms of the normal coordinates of the system. It is along these satellite trajectories that time correlation functions are calculated as correlations between Wigner transforms involving the corresponding operators. There is some arbitrariness in the choice of the satellites’ equations of motion, but in principle this method seems to provide a direct path to the full F(k,t).
Pressure induced changes in the structural and dynamic properties of bulk liquid Fe close to the melting line. A first principles study.

David J. González

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The static and dynamic properties of liquid Fe at high pressure and temperature have been studied using an ab initio molecular dynamics method. Results are reported for four thermodynamic states at pressures of 27, 42, 50 and 58 GPa for which xray scattering data are available. The calculated static structure shows very good agreement with the available experimental data, including an asymmetric second peak which becomes more marked with increasing pressure. The dynamical structure reveals the existence of propagating density fluctuations and the associated dispersion relation has also been calculated. The relaxation mechanisms for the density fluctuations are analyzed in terms of a model with two decay channels (fast and slow, respectively). We found that the thermal relaxation proceeds along the slow decaying channel whereas the fast one is that of the viscoelastic relaxation. The possible coupling between longitudinal and transverse excitation modes has been investigated by looking at specific signatures in two wavevector regions: the first one is located around the position of the main peak of the structure factor, \(Q_p\), as suggested by the recently reported appearance of high frequency transverse waves in liquid Li under high pressures; the second region is around \(Q_p/2\), as suggested by the recent finding of transverse acoustic modes in inelastic xray scattering intensities of liquid Fe at ambient pressure. Finally, results are also reported for several transport coefficients.
Metal to Nonmetal Phase Separation in Expanded liquid Alkali metals

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Different quantum mechanical calculations of the ground state energy for pure electron gases at different densities reveal a mechanical instability in terms of a negative compressibility if the gas is sufficiently expanded (see [1] and references therein). Appropriate scaling [1,2] allows relating the corresponding gas densities to mass densities in real liquid alkali metals that are regarded as perfect model systems for free electron gases. It is found that the region of instability corresponds to about 3-4 times the critical density (blue shaded area in the attached figure). This however contradicts the common view of the density induced metal to non-metal transitions in liquid alkali metals which are believed to set in at about 2-3 times the critical density, i.e. at considerably lower density (see e.g. [3]). The figure depicts this region by the grey shaded area. We have investigated the density dependence of the collective modes in liquid Rubidium to understand this discrepancy. The density range explored is indicated in the figure by the dashed red line. Collective modes were chosen as suitable probes for this investigation because their properties are highly sensitive to variations in the interatomic interactions which are expected to occur if the electron gas ceases to exist.

All experiments were carried out on the time of flight instruments IN4 and BRISP which are situated at the Institute Laue Langevin in Grenoble and which are perfectly suited to investigate the properties of collective excitations in liquids due to their small angle capabilities in combination with the use of thermal neutrons. As a result, we find that the density dependence of the collective mode properties shows a distinctive variation when the density range is approached where the electron gas instability is predicted (blue shaded area). Variation of intensity, widths and mode frequencies will be discussed and it is shown that the observed behaviour can be understood within a scenario recently suggested for the metal to non-metal transition in liquid mercury [4]. There the localization of the conduction electrons forces the formation of a micro emulsion consisting of a dense metallic and less dense non-metallic phase. The densities of these phases remain constant with further expansion but the volume fraction of the non-metallic phase increases causing the observed overall density reduction. This scenario explains the observed density dependence of the collective mode parameters along the explored density range and throws new light on the mechanisms of the density-induced metal to non-metal transition in liquid metals.

Structure and dynamics of high-pressure Na close to the melting line. An ab-initio molecular dynamics study.

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The melting curve of sodium for a pressure range up to 120 GPa has been evaluated by the orbital free ab initio molecular dynamics method. This method uses the electronic density as the basic variable combined with an approximate electronic kinetic energy functional and a local ionic pseudopotential. It scales almost linearly with system size, which allows to perform simulations with a large number of particles and for long simulation times. The calculated melting curve shows a maximum melting temperature at 30 GPa followed by a steep decrease lasting up to 110 GPa. For various pressures and temperatures we have evaluated some static properties, including average and local structure, electronic properties, like the electron localization function (ELF), and dynamic properties, both single-particle and collective ones, from which some transport coefficients are deduced. Despite the accurate reproduction of experimentally measured quantities [1], we do not observe any indication of an early transition from a bcc-like to an fcc-like liquid, as suggested previously [2], but rather a change in the pressure variation of icosahedral-like order and bcc-like order, with no sign of fcc-like structures in the whole liquid range studied. We also consider the evolution of the ELF within this type of local arrangements upon pressurization. In the dynamic realm we find an enlarged wavevector region where interatomic collisions are important as pressure is increased and temperature decreased along the melting line, leading to a peculiar behaviour of the dynamic properties.


Collective excitations in liquid metals. Relationship to the electron-electron interactions

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After the seminal paper of Copley and Rowe on liquid rubidium there have been numerous investigations of the collective dynamics of liquid metals. Here we present the high frequency, low momentum dynamics of liquid metals to correlate the collective mode velocity to the screened electron-electron interaction described in the homogeneous electron gas approximation. We present a way to determine the appropriate electron density which controls the screening of the ion potential though the electron gas dielectric function. It is shown that contrary to a simple expectation this model is not adequate only in the case of simple metals like alkali metals but it works correctly also in the case of polyvalent metals and even in the case of early transition metal like Ti. Some guess on the behaviour of the damping of collective modes is also proposed.
How to reveal the anomalous vibrational spectrum of glasses by different techniques

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Based on a theory of vibrational excitation in a system with spatially fluctuating elastic coefficients, developed recently by the author and collaborators, a scheme is developed for relating spectroscopic data obtained by inelastic neutron, X-ray, nuclear, light and Raman scattering, as well as by THz infrared absorption to each other and to information from molecular-dynamics (MD) simulations. The key quantity is the longitudinal and transverse dynamic susceptibility, which is related to the various scattering intensities (and MD spectra) by the fluctuation-dissipation theorem. It is shown that the deviation from the wave-like Debye spectrum (“boson peak”) is governed by the disorder-induced sound attenuation coefficient. It is further shown how this quantity dominates the spectral features obtained by the different spectroscopic methods.

Elastic heterogeneities and anharmonic damping of THz vibrations in glasses

Giacomo Baldi

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I will report on experiments performed in recent years by means of high resolution inelastic x-rays scattering (IXS) on a series of glasses of different structure and composition. I will discuss in some details two main results. The first is the striking similarity observed between the spectrum of the glass and that of the corresponding polycrystal [1], suggesting that the glass vibrations closely resemble those of the polycrystal, with excitations related to the acoustic and optic modes of the crystal. I will then describe a second important observation, namely the unusually large temperature dependence of the sound attenuation of a sodium silicate glass at terahertz frequency [2], an unprecedentedly observed phenomenon. The anharmonicity can be ascribed to the interaction between the propagating acoustic wave and the bath of thermal vibrations. At low temperatures the sound attenuation follows a Rayleigh-Gans scattering law. As the temperature is increased the anharmonic process sets in, resulting in an almost quadratic frequency dependence of the damping in the entire frequency range.

Vibrational excitations and elastic response in disordered solids

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Advanced inelastic scattering techniques have substantially improved our understanding of high frequency excitations at mesoscopic length scales in disordered solids. These are vibrational modes sensibly more complex than phonons propagating in the corresponding crystals, and show intriguing features like negative dispersion curves or Raleigh-like line broadening, proportional to the fourth power of frequency. This latter observation, in particular, has been recently rationalized in terms of interaction of the excitations with some form of local elastic heterogeneity: the system mechanical response does not conform to the expected macroscopic limit at all scales, as it is in the case of crystals, and elastic constants distributions [1] must be considered at the mesoscale. By using Molecular Dynamics simulation we have addressed these issues in the case of a toy model that, at fixed density and temperature, shows a phase behavior encompassing the perfect crystal, increasingly defective ordered phases, and the fully developed amorphous state. On one side [2], we have quantitatively characterized the extent of the elastic heterogeneity, directly calculating the local elastic constant distributions. On the other side, we have fully characterized the high frequency spectrum of all phases in terms of dynamic structure factors [3] and normal modes [2], continuously following the metamorphosis of plain phonons in the perfect crystals into acoustic-like excitations in the glass. In this talk I will describe in details the numerical results, and provide convincing evidence of a direct correlation between these two sets of data, for all investigated solid states of matter.

Neutron Brillouin scattering experiments on HRC and spin waves in metallic ferromagnet SrRuO$_3$

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The High Resolution Chopper Spectrometer (HRC) was installed at the BL12 beam port in the Materials and Life Science Experimental Facility (MLF), the Japan Proton Accelerator Research Complex (J-PARC), in order to study dynamics in condensed matters with high-resolutions and relatively high-energy neutrons [1-3]. On the HRC, neutron Brillouin scattering (NBS) experiments became feasible by reducing the background noise at low scattering angles down to 0.6°. NBS is the most promising way to observe excitations in the forward direction from powders, polycrystals, or liquids. Owing to the kinematical constraints of neutron spectroscopy, the incident neutron energy $E_i$ in the sub-eV region is necessary for measuring scattering in the meV range of the energy transfer $E$ near to (000), and with a high energy resolution of $\Delta E/E_i$, further, the scattered neutrons need to be detected at very low angles.

SrRuO$_3$ has a pseudo-cubic perovskite crystal structure and is a rare example of 4d band ferromagnetic metal with the ferromagnetic transition temperature, $T_c = 165$ K. This system exhibits an anomalous Hall effect, where the anomalous Hall conductivity shows a nonmonotonous dependence on the magnetization [4]. This anomalous behavior can be well reproduced by a first-principle calculation based on the Weyl fermion / magnetic monopole picture originating from the band crossing due to the spin-orbit interaction [4]. Recently, a large spin wave gap was observed in SrRuO$_3$, while La$_{0.8}$Sr$_{0.2}$MnO$_3$, a ferromagnet having a similar pseudo-cubic structure, showed no spin wave gap [5]. Here, we demonstrate the spin dynamics of the metallic ferromagnet SrRuO$_3$ by NBS experiments on the HRC. The observed spin wave dispersion in a polycrystalline sample of SrRuO$_3$ below $T_c$ is well described by $E(Q) = E_g + DQ^2$ ($Q$: wavenumber). The temperature ($T$) dependence of the spin wave gap $E_g$ shows nonmonotonous behavior, which can be related to the nonmonotonous $T$ dependence of the anomalous Hall conductivity. This work shows for the first time that the spin dynamics directly reflects the role of Weyl fermions [6].

Spin-orbit coupling and magnetic frustration in iridates

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I will present our study of two families of iridate magnetic materials, prone to magnetic frustration. The nowadays strong interest in iridates comes from the interplay between a large spin-orbit coupling, inherent to the presence of magnetic Ir⁴⁺, and moderate electronic interactions that can produce a new highly entangled spin-orbitronic state for the Ir⁴⁺. This novel quantum state may lead to unprecedented electronic transport and magnetic properties. The two systems of interest are first the pyrochlores R₂Ir₂O₇ (R = rare-earth element), where both the rare-earth and the Ir atoms lie on interpenetrated frustrated pyrochlore lattices. The magnetic interactions between the two magnetic species enrich the physics of these materials. The second system is Sr₃NiIrO₆ consisting of frustrated ferrimagnetic Ni-Ir chains on a triangular lattice with an unconventional magnetic ordering. I will show how the study of the electronic/magnetic excitations, combining Resonant Inelastic X-ray scattering and inelastic neutron scattering on various Energy-momentum transfer ranges, can bring important information and reveal new physics in these materials.

Observation of spin-wave dispersion in Nd-Fe-B magnets using neutron Brillouin scattering


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The low-energy spin-wave dispersion in polycrystalline Nd-Fe-B magnets was observed using neutron Brillouin scattering (NBS). Low-energy spin-wave excitations for the lowest acoustic spin-wave mode were clearly observed. From the spin-wave dispersion, we were able to determine the spinwave stiffness constant Dsw (100.0 meV Å²) and the exchange stiffness constant A (6.6 J/m).

H-bonding dynamics and macromolecular hydration in biology

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The essential, vital role of liquid water in molecular biology is so well established that its search guides the pioneering exploration for extra-terrestrial Life. Yet the H-bonding properties of water and their hydration interactions with biological molecules are still not fully understood. Water dynamics occurs on a range of time scales from the picosecond (THz range) to the nanosecond of diffusional motions. Because of the strong H/D isotope effect, neutron diffraction and neutron spectroscopy are particularly powerful experimental methods for the study of water in biology. In order to contribute to the discussion on Brillouin scattering, I shall briefly review data from neutrons on water interactions with biological molecules, with a particular emphasis on the role of H-bond dynamics in the THz range.
An integrated approach to investigate the collective dynamics of the phospholipid bilayer

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The collective dynamics of 1,2-dimyristoyl-sn-glycero-3-phosphatidylcholine (DMPC) bilayers in the gel and liquid phases have been investigated by Brillouin neutron scattering and far-infrared (far-IR) spectroscopy. A comprehensive picture of the vibrational features of phospholipid membranes at THz frequencies on the basis of the identified inelastic excitations is proposed. The typical excitation energies of both the gel and liquid phases are described in terms of three branches. A model consisting of three interacting damped harmonic oscillators is introduced to describe the system. A strong similarity between the THz vibrational dynamics of lipid membranes and those of water is found.
Inelastic light scattering techniques for monitoring the phase behaviour in hydrogel materials

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Hydrogels are a class of soft materials that resemble natural living tissues more than any other type of biomaterials due to their high water content and their elastic texture. Thanks to their biocompatibility and biodegradability, hydrogels appear very attractive for a wide range of applications, including drug delivery and controlled-release systems and scaffolding matrices for 3D culturing cells in the field of tissue engineering and regenerative medicine. The recently growing use of hydrogels especially in technological fields of high social impact has led to the need of precise control of mechanical and physico-chemical properties of these polymer networks starting from their molecular architecture. Besides these practical aspects, hydrogel materials are a good model system to be used to addressing the more fundamental question of how the behaviour of water in the proximity of hydrophobic and hydrophilic groups of complex molecules can drive gelation phenomena. This is particularly true in the case of polysaccharide hydrogels, due to the presence of both hydrogen-bond donor/acceptor groups in the polymeric backbone of the hydrogel matrix.

In this contribution, we show how the joint use of UV Raman scattering and Brillouin spectroscopy can be implemented for monitoring the phase changes in polysaccharide hydrogel materials and their influence on the localized and collective vibrational dynamics of the system. We investigate the molecular mechanism responsible of the temperature and pH-responsive behaviour exhibited by natural and biodegradable cyclodextrin-based hydrogels, by exploiting high capability of UV Raman scattering experiments, performed by taking advantage of the tuning of excitation sources of appropriate intensity of the synchrotron facility. Changes in the gel structure can be probed by following the intensity, frequency position and spectral shape of Raman peaks that represent the vibrational signatures of the reorganization of hydrogen-bond network of water molecules and of the solvation-effects in the proximity of hydrophobic/hydrophilic groups of the polymer backbone in the hydrogel state at variable temperature, hydration level and pH. In a complementary way, Brillouin scattering is used to probe the collective dynamics of the system, by measuring the propagation velocity and the attenuation of longitudinal acoustic modes at different conditions of temperature and pH. Characteristic phase transitions of the materials can be linked to the observed changes in the temperature and pH dependence of the acoustic velocity, providing information on the evolution of the elastic properties of the hydrogels.

The overall results presented here corroborate the potentiality of the joint use of inelastic scattering techniques to provide a comprehensive molecular view of complex macroscopic phenomena exhibited in hydrogel phases.
Low-frequency inelastic features of proteins and their role in biological functionality

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Protein biological function depends on structural flexibility and change. The opening and closing of ion channels for signaling currents depends on the structural changes of the transmembrane protein forming the pore. In the case of haemoglobin, the oxygen-transporting protein in blood, global structural change with initial binding of oxygen in a single domain biases the structure towards binding in additional domains, increasing delivery efficiency. It has been suggested that long-range vibrations involving dynamical networks that extend through the protein play a crucial role in controlling these structural changes [1]. Recent OKE experiments have revealed the existence of underdamped delocalized vibrational motions in the THz domain which are related to the binding of lysozyme with the inhibitor triacetylchitotriose in solution [2]. These results have reopened a quite debated discussion on the role played by low-frequency modes for functional activity of biomolecules. In this talk we will present neutron scattering results that shed some light on the characteristics and the nature of protein low-frequency inelastic features, their relationship with water dynamical properties and the possible connection with biological functionality. A particular attention will be drawn on the collective vibrational properties of maltose binding protein, one of the few perdeuterated proteins measured by Brillouin neutron scattering.


Hydration vs. Bulk Water: what we have learnt from Neutron Brillouin Spectroscopy

Andrea Orecchini

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During the last decade, the development of new instrumentation for neutron Brillouin spectroscopy has allowed to observe the coherent collective dynamics of hydration water in biomolecules. With an appropriate use of isotope substitution and contrast methods, THz modes propagating in hydration water could be singled out and studied for biological systems of increasing complexity, spanning from the model protein Ribonuclease A, to the more complex DNA, up to entire living cells of Escherichia coli. The experimental results revealed both similarities and differences with respect to the previously known behavior of bulk water. The emerging differences have been further confirmed by new data measured in similar experimental conditions for bulk water. In this talk, we will summarize what we have learnt from the comparison between bulk and hydration water, which pointed out the effects on water collective dynamics produced by a perturbation of the hydrogen-bond network.
Inelastic x-ray scattering in the ID28 beamline of the ESRF

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The ID28 high resolution inelastic x-ray scattering (IXS) beamline at the ESRF is dedicated to the study of vibrational properties of condensed matter. The first part of this talk will include an overview of the instrumentation and the recent scientific activity on the beamline. Moreover, the (thermal) diffuse scattering (TDS) side-station, which is currently under construction, as well as the unique capabilities arising from the combination of TDS and IXS studies will be briefly described. In the second part of the talk, a few recent experimental results highlighting the complementarity of Brillouin and IXS will be given. Finally, recent IXS results on the lattice dynamics of superconducting and charge-density-wave systems will be briefly presented.


Brillouin microscopy for subcellular 3D mechanical imaging

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Brillouin spectroscopy has shown great potential to become a reliable diagnostic tool due to its capability of measuring viscoelastic properties of materials in a non-contact manner. The recent development of high-sensitivity CCD cameras and Virtually Imaged Phase Array (VIPA) etalons has dramatically reduced data acquisition time to ~0.1 sec per spectrum. This has brought Brillouin spectroscopy from a point sampling technique to a new imaging modality. We describe the characterisation of a confocal Brillouin microscope designed to measure mechanical properties of biological tissues. The frequency broadening of the Brillouin spectrum due to high illumination and collection apertures has been investigated in order to determine the optimal geometry that maximises both the spectral and the optical resolution [1]. A high extinction ratio was achieved in a Michelson interferometer to suppress strong specular reflections [2]. Sub-micron resolution Brillouin images of single cells and arterial wall tissues have been acquired, in particular when atherosclerotic plaques were formed [3]. These results might encourage the application of Brillouin microscopy as a tool of choice in clinical practice.

Ray-tracing simulations for a high-flux upgrade of the BRISP spectrometer

Marco Zanatta

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To date, the BRISP spectrometer represents the state-of-the-art for every instrument aiming to perform Brillouin neutron scattering. Exploiting accurate ray-tracing McStas simulations, we will investigate an improved configuration of BRISP primary spectrometer to provide a higher flux at the sample position, while preserving all the present capabilities of the instrument. This configuration is based on a neutron guide system, and it is designed to fit the available room on the instrument platform with no modification of the secondary spectrometer. Preliminary evaluations show that this setup can achieve a flux gain factor ranging from 3 to 6, depending on the wavelength. This can expand the experimental possibilities of BRISP towards small samples, possibly using also complex sample environments. Moreover, recent works on the collective dynamics of glasses highlighted the crucial importance of mapping a wide $Q$-$E$ region with an appropriate resolution, to properly understand the dynamics of these systems. This versatility is natively implemented on BRISP, and it could be improved by properly expanding the available choices for the monochromator crystals.
T-REX: A Time-of-flight Reciprocal space Explorer for the future ESS source

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We present the concept of a time-of-flight chopper spectrometer for the future ESS neutron source. The instrument is designed to yield a dynamic range that extends from 20 μeV to 150 meV in energy transfer and from 0.01 Å⁻¹ to 17 Å⁻¹ in wavevector transfer. The elastic energy resolution (FWHM) can be freely adjusted in the range from 1% to 3% at 3meV and from 4% to 8% at 100 meV. T-REX has been benchmarked against existing state-of-the-art neutron TOF-spectrometers and shows flux gain factors between one and two orders of magnitude and possible solutions are foreseen to increase the signal to noise ratio. These features make the instrument a real game changer in wide areas of scientific research: magnetism, strongly correlated electron materials, functional materials, soft-matter, biophysics and disordered systems. Independent of the specific application, it will allow parametric studies with acquisition times below one hour for a full data set from single crystals, the application of extreme conditions, in-situ or in-operando studies on timescales of seconds or spectroscopy from sub 100 mg single crystals.

It will implement time-of-flight spectroscopy with Polarization Analysis as a standard tool, e.g. for studying the effect of confinement on the magnetic excitations in nano-particles or to uniquely derive the vibrational hydrogen excitations in soft matter through separation of the nuclear spin incoherent scattering.

The wide energy transfer range available enables studies of picosecond diffusional and relaxational dynamics of biosystems as well as their vibrational counterpart, in one experiment, while keeping the sample under controlled environmental conditions.

Parametric studies (e.g. under variable T or p) of the vibrational density of state will become routine, while the polychromatic character of T-REX (Repetition Rate Multiplication) will open new ways for studies of vibrations characterized by damped phonon-like excitations in disordered systems, which are nowadays possible at dedicated Brillouin Neutron spectrometers.
Focusing adaptive-optics for Brillouin neutron spectroscopy at extreme conditions

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Neutron Spectroscopy employing extreme-conditions sample environments is nowadays a crucial tool for the understanding of fundamental scientific questions as well as for the investigation of materials and chemical-physical properties. For all these kinds of studies, an increased neutron flux over a small sample area is needed. The prototype of a focusing neutron guide component, developed and produced completely at the FRM II, has been installed at the time-of-flight (TOF) disc-chopper neutron spectrometer TOFTOF and came into routine-operation. The design is based on the compressed Archimedes’ mirror concept for finite-size divergent sources. It represents a unique device combining the supermirror technology with Adaptive Optics, suitable for broad-bandwidth thermal-cold TOF neutron spectroscopy (here optimized for 1.4 – 10 Å). It is able to squeeze the beam cross section down to a square centimeter, with a more than doubled signal-to-background ratio, increased efficiency at high scattering angles and improved symmetry of the elastic resolution function. We present a comparison between the simulated and measured beam cross sections, as well as the performance of the instrument within real experiments. This work intends to show the unprecedented opportunities achievable at already existing instruments, along with useful guidelines for the design and construction of next-generation neutron spectrometers.

FEL-based four wave mixing experiments stimulated by EUV transient gratings

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Four wave mixing (FWM) is a third order coherent non-linear process, which permits to study a large array of dynamical processes in real time. An important step forward in this technique is represented by its extension to the extreme ultra violet (EUV) and soft x-ray regime [1,2]. EUV/soft x-ray FWM may enable elemental selectivity and nano to atomic spatial resolution, as well as probing an energy-wavevector range inacessible by optical methods. In particular, EUV/soft x-ray transient gratings (TGs) can be used to impulsively excite vibrational modes in the “mesoscopic” wavevector range (0.1-1 nm⁻¹), which is of the greatest relevance for the study of disordered systems and nanostructures [3]. We hereby report on the experimental demonstration of EUV/soft x-ray FWM [4], obtained by combining a porposely designed setup and the unique coherence properties of the ultrafast EUV pulses delivered by the seeded free electron laser (FEL) source FERMI (Elettra, Italy).

The combination of the FWM approach and the two-colour operation mode of FERMI have allowed the realization of more complex FWM experiments, namely coherent Raman scattering [5]. This approach can be in principle used to detect the ultrafast dynamics of high-energy excitations, such as valence band excitons, as well as their correlations with vibrational dynamics; preliminary results on this side will be also reported.

Analysis of dynamic structure factors of liquids outside hydrodynamic regime

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The origin of collective propagating and relaxing processes contributing to dynamic structure factor is well understood only in hydrodynamic regime when the liquid is treated as continuum. Outside the hydrodynamic regime there exist non-hydrodynamic collective modes like structural relaxation, shear and heat waves which can contribute to collective dynamics. In particular, recently several groups claimed visible contributions to the experimentally measured dynamic structure factors coming from transverse collective excitations. In my talk I will present a scheme of analysis of time correlation functions (obtained either from classical or ab initio simulations) and dynamic structure factors which enables to separate contributions coming from different collective modes outside the hydrodynamic regime with the purpose of estimation of their origin. A comparison with a DHO fit will be shown.

Structure and dynamics in liquids and fluids at extreme conditions

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Physics of hot dense fluids and liquids is one of the hot topics of modern condensed matter physics. Unexpected liquid-like behavior of dense fluids or liquid liquid transitions in hot dense liquids are some examples of this fascinating scenario emerging recently. I will describe our advances in this research showing some recent IXS experiments on fluid Ar and liquid Rb at high pressures and temperatures, together with ab initio simulations, able to give a deeper physical insight and to investigate to even more extreme conditions, still difficult to achieve experimentally.
The measurement of time-dependent correlation functions can be achieved using different experimental techniques. During the last years, the time-resolved non-linear spectroscopy has proven to give valuable and complementary information on the dynamics of complex liquids [1]. The time-resolved optical Kerr effect (OKE) performed by femtosecond laser enables to measure the very fast dynamics, up to about 10 THz; the Transient Grating technique, realized with pulsed and cw laser sources, covers the dynamic window from GHz to KHz enabling the measurement of the slow dynamics inclusive of the Brillouin spectrum.

Here we report on OKE investigation of the vibrational dynamics and relaxation processes in supercooled bulk [2,3]. The experiment measures the water intermolecular vibrations and the structural relaxation process in an extended temperature range, down to 247 K. We performed a comparative and detailed data analysis based on different theoretical models. We found the mode-coupling models provide the more rigorous and complete model for water dynamics. These results on the water dynamics support the existence of two local structures coexisting in the liquid and supercooled phase. The comparison of the present data with that in the amorphous ice suggests to attribute the local structures to high-density and low-density water forms.

Moreover, using the OKE technique we investigated the dynamics of water confined in silica pores of 4 nm at different levels of hydration and temperatures [4,5]. The dynamic scenario is relatively complex and it can be rationalized considering the presence of two water types inside the hydrophilic nanopores: the outer water, close to the pore surfaces, and the inner water, contained in the internal part of pores.

Brillouin neutron and x-ray scattering from an aqueous system at low temperatures

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In the first part of the talk we discuss our studies of collective excitations in an aqueous solution of lithium chloride that is remarkably similar in many properties to pure water, but does not freeze easily. The salt addition allowed us to circumvent water freezing and perform neutron and x-ray Brillouin scattering experiments in the temperature range of 270 K to 205 K. Both series of experiments revealed the presence of lower- and higher-frequency excitations, similar to those observed in pure water above freezing temperature. These two excitations were detectable through the entire temperature range of the experiment, at all measured values of the scattering momentum transfer (2 nm⁻¹ < Q < 18 nm⁻¹). Our initial analysis of the fit parameters of the Brillouin spectra did not reveal the presence of the expected “dynamic crossover” in the collective dynamics of water. In order to probe a wider temperature range using a quasicontinuous set of temperature points, we collected the elastic scattering intensities as a function of temperature using both neutrons and x-rays. We performed elastic intensity temperature scan measurements on different spectrometers to separate the effects due to the coherent vs. incoherent dynamics from those due to the difference in the energy resolution (that is, the probed time scale). A clear evidence of the crossover in the dynamics of the water molecules in the solution was observed in the single-particle relaxation dynamics on the micro-eV (nanosecond) time scale, but not in the collective dynamics on the milli-eV (picosecond) time scale. Furthermore, the neutron incoherent elastic intensity scans with milli-eV resolution showed largely regular harmonic behavior as a function of temperature, indicating that the dynamic crossover, clearly visible on the micro-eV (nanosecond) energy scale in the neutron data is not accompanied by any pronounced change in the elastic constants. In the second part of the talk we discuss the present and future possibilities of Brillouin scattering at Spallation Neutron Source, Oak Ridge National Laboratory.
Methanol is the simplest molecule containing a polar hydroxyl group and a non-polar methyl group, thus offering the possibility of studying the properties of small amphiphile systems in aqueous environments, as well as self-assembly phenomena. As for other alcohols it shows non-ideal behaviour when mixed with water and a large negative excess entropy that has been often attributed to an enhanced water ordering in the vicinity of hydrophobic groups. However the validity of such model has been questioned and alternative suggestions proposed. We have recently explored by means of quasielastic neutron scattering and MD simulations the possibility of observing anomalies in the dynamics of water and methanol in the mixtures that could be related with any of the available propositions. As part of this effort, the collective dynamics of pure methanol have also been measured using the BRISP spectrometer at the Institut Laue-Langevin (Grenoble, France). The experimental results have been analyzed in combination with the MD and show striking similarities with the collective dynamics of water. In particular a third high-frequency mode that in recent simulation results on supercooled water has been associated to four-coordinated molecules is also visible in methanol, suggesting that tetrahedrality is not an essential requisite for this mode to appear.
Coherent Brillouin spectroscopy with femtosecond light pulses

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We recently developed a novel pump-probe approach (Broadband Photo-Acoustics, BPA) based on the generation and detection of coherent vibrational excitations, which enables to simultaneously determine sound velocity, acoustic damping and refractive index dispersion in a wide momentum-energy region, bridging the gap between traditional inelastic light scattering and neutrons/X-ray domains.

I will describe the working principle of the method and the experimental setup, along with two recent applications. The first [1] aims to answer to a central question for glass transition theories: does a glass cease to flow at finite temperature?. The second [2] deals with the dynamical transition between the elastic continuum and the microscopic regime in glasses, as visualized by the attenuation of hypersonic waves and by the vibrational density of states (Boson Peak). Finally, perspectives for the application of BPA to liquids will be discussed.


Water dynamics under high pressure: the hydrogen bond role

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The access to hydrogen dynamics under high pressures (GPa) is of paramount importance for several issues spanning from volcanology to planetary science and allows to probe the existence of new exotic phases in water predicted \cite{1-3} to show up at high pressure and characterized by an anomalous proton mobility.

We recently investigated the THz dynamics of liquid H\textsubscript{2}O along high temperature isotherms (400 \textdegree{}K-500 \textdegree{}K) in the GP regime, by coupled inelastic neutron and x-ray scattering experiments \cite{4-6}. We observed dynamical anomalies both in the diffusive and collective dynamics of water, revealing the persistence of the hydrogen bond network in hot dense water up to ice VII crystallization. Both the relaxation strength of the hydrogen bond network and the molecular reorientation time are shown to be pressure-independent. Our results strongly question the long-standing belief that hydrogen bonds are destroyed in hot liquid water under the effect of compression and suggest an alternative mechanism for water densification at high temperature.

\cite{6} U.L. Ranieri, L.E. Bove et al. submitted.
The method of Neutron Brillouin Scattering (NBS) is not quite new but has been developed from the initial time of neutron beam research. In 1970s it was revived again mainly for the research of spin wave measurements from either powdered or glassy (or amorphous ferromagnets) materials. On the 3 axis spectrometers installed at the reactor facilities the measurable energy transferred energy is limited below 3 meV due to the kinematical constraint. On the other hand, once the single crystal is available the spin wave dispersion curves are readily determined with a wide range of the momentum–energy space. Currently, numbers of new ferromagnets can be synthesized, for instance, of transition metal oxides, doped semiconductors, or multi layered materials for the spintronics devices. These materials are in many cases are difficult to be synthesized to the single crystalline form so that the NBS becomes the important method in order to determine the necessary parameters of the ferromagnet. Particularly the determination of the spin wave stiffness constant is very important for obtaining strong ferromagnetism. The NBS measurement combined with pulsed neutron sources is mandatory for the microscopic (mostly spin wave measurement) studies for these materials. I will emphasize such an importance that NBS should be more recognized in future as the fundamental tool for the materials science development by taking several realistic examples.
Although predicted by L. Brillouin since 1922, Brillouin scattering became a usual technique only at the advent of coherent light sources (lasers). Depending on the wavelength of the radiation, Brillouin light scattering reaches typically the GHz domain and momentum transfers of the order of 10,000 cm$^{-1}$, where matter is well described as continuous, although not necessarily homogeneous. Most of the studies focused on dispersion curves that relate energy and momentum transfers of the probed collective excitations, eventually identifying structural relaxation times. The case of heterogeneous systems was also extensively studied particularly because of the many potential applications. Generally, the contribution of Brillouin light scattering to the knowledge of liquids and glass dynamics has been very important.

More generally, collective excitations in condensed matter are potentially observed at very different time scales; consequently probing different phenomena. From the KHz-MHz region covered by ultrasound techniques, till the THz domain studied by X-rays or neutron scattering, the different couplings give information about large heterogeneities of the matter, mechanisms of mechanical absorption, rigidity, intermolecular forces, bulk and shear moduli or localised vibrations. Coherent neutron scattering, because interacting only with the massive part of the atoms is a rich approach although, in practice, it is a technique penalized by the relatively poor energy resolution of instruments which allows measurements only of a few systems, in cases where both the elastic and incoherent quasi-elastic contributions are sufficiently weak. Yet, whenever possible, neutron Brillouin scattering in the THz domain allowed the observation of high frequency branches of the dispersion relation in cases where structural relation takes place at high energy. Some examples issued from the complex physics of water and amorphous ices will be presented. Also the complex problem of the transition from the continuum (hydrodynamic) description to the individual particle models remains a domain open to research. It is worth mentioning that the transition towards localized modes or the merging of collective excitations in inelastic low frequency vibrations of amorphous materials are currently studied, particularly at the dedicated instrument BRISP at ILL.

A natural extension of Brillouin scattering is the study of the density of the vibrational density of states of particularly interest in the case of glasses. We will present shortly the main aspects of the propagation of collective excitations and its relation with the density of states, focusing on possibilities and perspectives of neutron scattering.
Posters
Spectral modes from the expansion in Lorentzian lines: the case of spectra of current autocorrelations

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The general theory of exponential expansion of time correlation functions in many-body statistical mechanics [1] has a very simple but remarkable consequence for the second derivative of a time correlation function \(a(t)\). The frequency spectrum of \(a(t)\) is made of central Lorentzians and/or distorted inelastic Lorentzians, characterized by parameters \(z_j\) (where \(j\) numbers the spectral modes) which are either real widths or complex quantities representing damping and frequencies of oscillatory modes, respectively. Then, \(d^2 a/dt^2\) has a spectrum composed by lines with the same set of \(z_j\)'s. Any spectral mode appears in both spectra with different amplitudes only, and a simple and explicit relation holds between corresponding amplitudes. This result (i) proves that the time autocorrelation of a dynamical variable and that of its derivative of any order contain the same information on the system considered, and (ii) has an immediate application to the case of current autocorrelations. The dynamic structure factor and the longitudinal-current correlation spectrum form the best-known pair of frequency distributions corresponding to time correlations related to each other by a double-derivative operation. Both are commonly used for the description of the collective dynamics of a system. We illustrate the above mentioned property with examples in the case of simple dynamical models and draw some conclusions relevant to the analysis and interpretation of collective dynamics spectra.


The collective dynamics of a hydrogen-bonded liquid: methanol

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Methanol is a hydrogen-bonded liquid of enormous importance in pure and applied physics and chemistry, and is the object of innumerable studies. Yet, the fundamental aspects of its molecular dynamics are still known only to a very poor extent. The study of the collective dynamics is hindered by the weakness of the acoustic excitations, which has led to the wrong conclusion that sound modes propagate only in a surprisingly narrow range of small wave vector values. Combined molecular dynamics simulations and neutron Brillouin scattering measurements reveal, however, quite a different situation. Methanol is shown, for the first time, to feature the normal viscoelastic behaviour typical of a large variety of liquids, including the arrest of acoustic propagation near the peak of the static structure factor. Besides this, however, two more excitations are detected in the molecular centre-of-mass dynamics structure factor, at frequencies, respectively, one lower and one higher than the acoustic frequency, both with negligible dispersion. The rich translational dynamics revealed by this study classifies methanol as a fluid partly similar to the most important hydrogen-bonded liquid: water.
A Bayesian Approach in the analysis of Inelastic Neutron Brillouin Spectra

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In this work we describe a Bayesian approach to the analysis of Neutron Brillouin Scattering (NBS) data. Specifically, when dealing with spectra related to liquids and disordered systems, which are typically characterized by a poor definition of the excitation lines and, generally speaking, of spectroscopic features, the central issue is to establish how many excitation modes are justified by the experimental data and to support the related choices about model parameters on a probability basis. Furthermore when overdamped excitations are present, commonly used and widespread fitting algorithms are particularly affected by the initial values of the parameters that may lead to an inefficient exploration of the parameters space and, consequently, to output results corresponding to a local minimum the algorithm is not able to escape from. The statistical method we discuss here could turn out of great importance in determining the reliability and significance of physical information extracted from experimental data, especially in those cases where the measurement of spectral features is rendered difficult not only by the kind of sample, but also by the limited instrumental resolution and count statistics.

An algorithm based on Markov chain Monte Carlo and Reversible Jump techniques has been applied to model simulated data generated by different combination of several Damped Harmonic Oscillator functions. The output will be the most probable number of components in the simulated neutron Brillouin scattering spectra (and of course the posterior distribution function for the variable “number of lines”) together with a posterior distribution function of all the relevant dynamical parameters. Finally we show how the algorithm manage to fit on real experimental data on liquid gold, collected on the Brillouin Spectrometer Brisip at the Institut Laue Langevin and how the results are consistent with the rigorous analysis already assessed in literature.

We also envisage the very interesting possibility, offered by this approach, to apply physical constraints to fitting models, such as theoretically known sum rules or the finiteness of higher-order frequency moments of the dynamic structure factor.
New experimental results for the scattering cross sections of liquid and solid deuterium for ultracold neutrons and an approach to their calculation

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We present the scattering cross sections (σ⁰scatt) of liquid deuterium at T = 20.6 K and solid deuterium at 14.8 K and 16.5 K for ultracold neutrons (UCN) for various sample preparation methods [1,2]. The measurements on a mixture of ortho- and para-deuterium (c_ortho=0.8) were taken at the Institute Laue-Langevin by means of a UCN transmission experiment. The indispensable thorough raw data treatment procedure, including quantum-mechanical effects, is explained.

For the first time, a calculation model for coherent and incoherent scattering in liquid deuterium in the hydrodynamic limit based on appropriate physical concepts is provided and shown to fit the experimental data well. The applicability of the incoherent approximation for UCN scattering in liquid deuterium was tested and found to deliver acceptable results.

In addition to liquid deuterium, transmission measurements were also carried out for solid deuterium of various temperatures and preparation methods (i.e. freezing speeds and temperature cyclings). The results for crystals frozen from the liquid down to 14.8 K and 16.5 K show that the scattering cross sections of deuterium increase with temperature. The influence of the preparation method on scattering cross sections was studied by preparing solid deuterium crystals in two different ways – fast frozen (from the liquid phase) and slowly frozen (from the gaseous phase). The fast frozen crystals exhibit larger UCN scattering cross sections than the slowly frozen ones. The two were measured at 16.5 K and 16.1 K, respectively.

Regardless of the preparation method, both crystal types show a much larger than expected elastic cross section at UCN energies. The expected incoherent elastic cross section, which arises from molecular properties, was calculated to be 4.5 barn (for c_ortho=0.8), leaving – depending on the crystal preparation method – 9 to 43 barn unexplained. We believe that voids and cracks in the deuterium crystal are the cause of this elastic cross section, as proposed earlier. We applied the Guinier approximation in order to extract new information about those crystal defects from our data.

Collective excitations in liquid and glassy 3-methylpentane

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The existence of high-energy propagating collective excitations in disordered condensed matter systems, such as amorphous glasses and liquids, has been widely demonstrated by inelastic x-ray scattering and by neutron scattering experiments. Here we report on a detailed investigation of the two common types of disorder, static and dynamic, as can be determined at the terahertz frequencies in 3-methylpentane, by means of high-resolution inelastic x-ray scattering. We probed the vibrational dynamics in a large temperature range, including the glass, the supercooled liquid, and the liquid phases and we measured the static structure factor $S(q)$ at all temperatures. We found that the characteristic frequency of the excitations follows a well-defined dispersion curve, miming the longitudinal modes of a crystalline solid, extending beyond $8 \text{ nm}^{-1}$ at all the investigated temperatures. This indicates the persistence of a solid like behavior also in the liquid phase. Experimental results from the dynamic structure factor $S(q, \omega)$ suggest the existence of a pseudo-Brillouin zone parameter $a_0$ whose size compares surprisingly well with the periodicity inferred from the first sharp diffraction peak $a_m$ in the $S(q)$. Furthermore, in the whole investigated temperature range, both length scales exhibit a weak temperature dependence, about $15\%–20\%$, and well compare to the average intermolecular distance $a$, obtained from molecular number density.

We finally show that the IXS sound velocity coincides with the infinite frequency sound velocity previously inferred from visible and ultraviolet Brillouin spectroscopy data. This analysis confirms the role of the shear relaxation processes in determining the variation with frequency of the apparent sound velocity, and supports the profound role attributed to the interatomic separation in governing the thermodynamic properties of disordered systems.
Monte Carlo estimates of multiple scattering in neutron spectroscopy: an efficient algorithm even with large detection areas

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The substantial upgrade in new generation reactor-based time-of-flight (ToF) spectrometers lies in their hugely increased detection area. For such instruments, the strong improvement is clearly the high neutron-collection power, and with this the count statistics achievable in relatively short times. Dealing with thousands of time channels and several tens of thousands of detection pixels is, however, quite punishing as soon as data handling and correction for various effects in real-geometry conditions are considered. Anisotropic multiple scattering evaluation, even in an approximate way, is surely the most demanding step in the general treatment of inelastic neutron data, and becomes a very hard task when “extreme” conditions are further imposed by a widely-extended detection geometry, as that typical of new or upgraded neutron ToF spectrometers such as BRISP, IN4C or IN5 at the Institut Laue Langevin in Grenoble. For this reason, we refreshed our approach to multiple scattering calculations, in order to obtain reasonably accurate real-geometry results in nearly real-time conditions. Our new code, though conceptually originating from a long standing experience of Monte Carlo (MC) integration techniques to extract (unnormalized) double and single scattering intensities, is now made particularly efficient in computing time both by a careful application of the “importance sampling” method used to calculate some of the required MC integrals, and by the choice of programming languages which allow for a heavy but efficient use of matrix algebra. The fast matrix manipulation performances offered by some languages thus allow to avoid the (far slower) nested-loop logic required by more traditional languages. The concepts at the basis of the algorithm and several implementation details are presented.
The exotic behavior of Pr$_{0.5}$Ca$_{0.5}$MnO$_3$: a complete structural and dynamical analysis

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Strongly correlated systems are subject of intense research nowadays because of their uncommon properties. Most results suggest that intrinsic inhomogeneities with a tendency to phase-separation play a key role in these compounds. We studied the nature either static or dynamic of the short ranged correlations evidenced in a polycrystalline sample of Pr$_{0.5}$Ca$_{0.5}$MnO$_3$.

The sample was studied by means of several techniques as magnetic susceptibility, neutron powder diffraction (NPD) and inelastic neutron scattering (INS). The complete magnetic structure was determined by NPD at the D20 diffractometer at ILL with a temperature ranging from 10 K to 300 K. Furthermore we carried on measurement with XYZ polarization analysis on D7 at ILL and we evidenced the existence of a large magnetic peak centred at 1.6 Å$^{-1}$, persisting well above the transition temperature of the charge ordering (Fig. 1). This peak evidences the presence of short ranged correlations still present above the charge ordering. In order to discern the possible dynamic nature of this signal, we performed measurements on BRISP at ILL (Fig. 2). We followed the evolution of magnetic correlations at the charge ordering transition; moreover, it was possible to analyse a quasi-elastic component emerging at higher temperatures.
Physical gels made of amphiphilic molecules, namely in this study Methyl-4,6-O-benzylidene-Î±-D-mannopyranoside, can be formed either in polar and protic liquid like water or in organic apolar solvent such as toluene. The solvent, that influences the supramolecular organisation of the gelators, plays an important role in the stability and formation of the gel phase. Gelator-solvent interactions govern not only the assembly but also the solvent diffusion in the material. We present here our first measurements of Transient Grating spectroscopy, that provide valuable information on the gel characteristics through the monitoring of the longitudinal acoustic wave propagating in the system. Opposite effects on the speed of sound in the gels are observed for the two solvents investigated, being relevant of the interactions between the gelators and the surrounding liquid.

Collective dynamics in a disparate mass amorphous alloy Zr\textsubscript{40}Be\textsubscript{60}

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Experiment at IN4 spectrometer \cite{1} with neutron incident wavelength of 2.2 Å on Zr-Be metallic glasses reveal two distinct vibrational modes. The collective character of the low-energy and high-energy excitations, i.e. a dispersion relations, are observed beyond the first pseudo-Brillouin zone boundary up to 7 Å\textsuperscript{-1}. Aiming at investigating whether the high-energy excitations in solid disordered systems and binary liquids are the same physical origin, we performed a neutron scattering experiment at the BRISP spectrometer \cite{2} on the Zr\textsubscript{40}Be\textsubscript{60} metallic glass in vicinity of the first pseudo-Brillouin zone boundary. Experiment was made with neutron incident wavelength of 1.03 Å.

Independent time-of-flight spectra were converted into the dynamic structure factor, \(S(q, E)\). An alternative approach to discuss the dynamic structure on qualitative level makes use the correlation function of the longitudinal currents, \(J_l(E, q) = (E/q)^2 S(E, q)\). When the maxima of \(J_l(E, q)\) are plotted as a function of \(q\), a dispersion relation \(E(q)\) is obtained (see Fig. 1). With increasing momentum transfer the dispersion relation of low-energy mode goes through a maximum and then through a minimum at values of \(q\) close to \(q_p/2\) and \(q_p\) (the position of the first peak in the static structure factor, \(S(q)\)). It is well established behavior of the acoustic modes in disordered systems \cite{3}. A much more pronounced dispersion is found for the high-frequency mode. Energy of this excitation drastically decreases towards \(q_p/2\) (the maximum of dispersion relation of acoustic mode).

In order to achieve a first pseudo-Brillouin zone for high-energy mode it is desirable to commission a Cu(220) monochromator, which provide us with neutron incident wavelength of 0.769 Å.

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THz dynamics of glasses studied by means of inelastic neutron scattering

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The THz dynamics of two different glasses has been investigated by means of Inelastic Neutron Scattering: the glucose (C₆H₁₂O₆) [1] and the germanium diselenide (GeSe₂) [2], an amorphous glass belonging to the family of continuous random network forming glasses. In both cases two well-defined peaks are resolved in the dynamic structure factor in a broad $Q$ region, below and above the position of the first sharp diffraction peak (FSDP) in the static structure factor. One can be interpreted as the THz-regime evolution of the longitudinal acoustic mode sustained at lower frequencies. In both systems, the apparent propagation speed, which has been deduced from the INS experiment, shows a minimum at the wavevector corresponding to the position of the FSDP. The second branch can be either an optic- or transverse-like mode. By comparing the dispersion relation of this branch to the experimental density of states of glucose, a possible relation has been identified between the low frequency mode and the well-known excess of low frequency modes, that is, the Boson Peak.


The Neutron Brillouin Spectrometer BRISP

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The spectrometer BRISP is presented together with some examples of its recent activity of research.