HIRES 2023

Synergies in HIgh RESolution Spectroscopy

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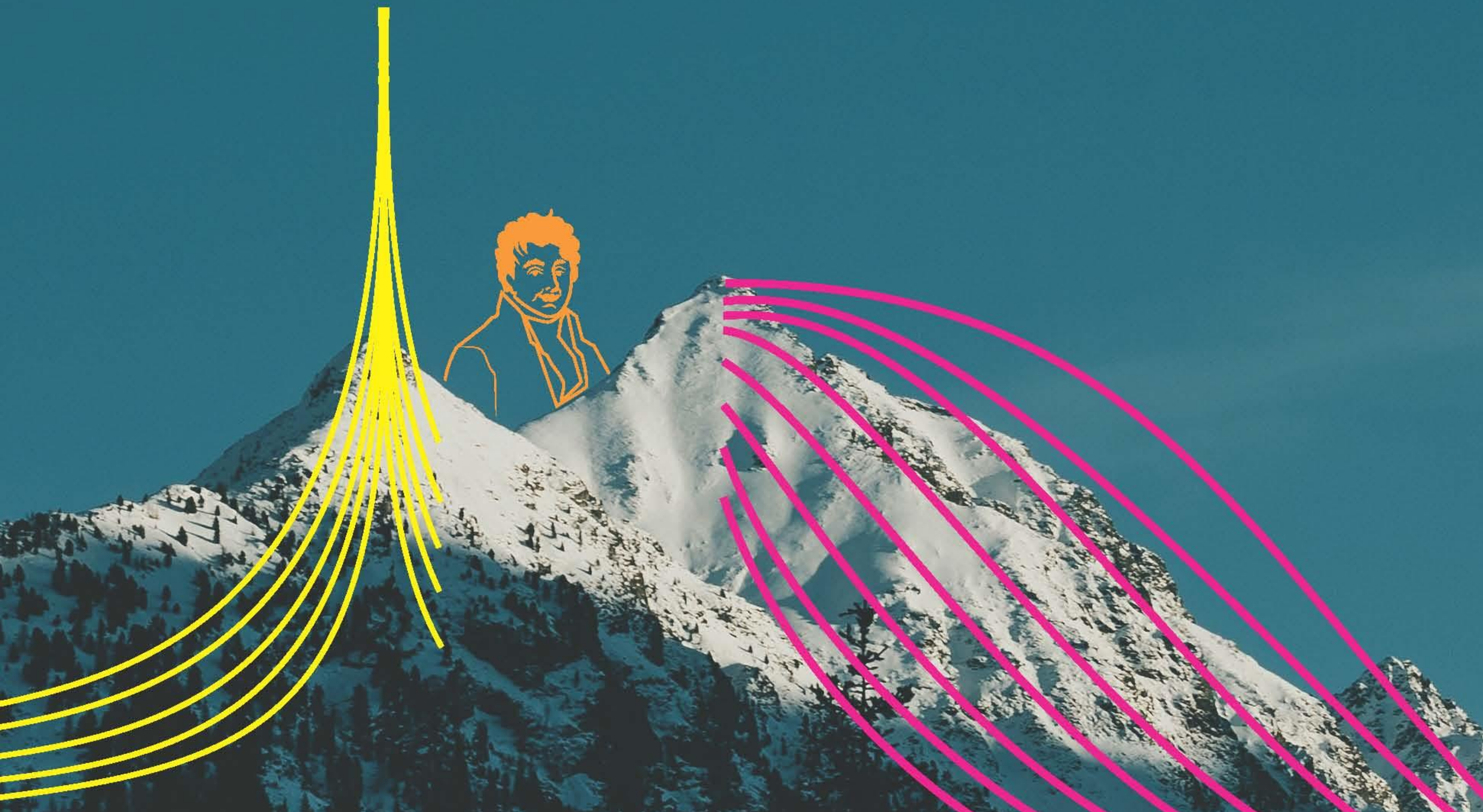

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HIRES 2023
Synergies in HIgh RESolution Spectroscopy
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Programme

HIRES 2023

Synergies in High RESolution Spectroscopy

December 2023

	12 Tuesday	**13** Wednesday	**14** Thursday	**15** Friday
8:45-9:00	**Opening – Ken Andersen**			
	Session Chair: NN	**Session Chair: F. Schreiber**	**Session Chair: T. Keller**	**Session Chair: F. Juranyi**
9:00-9:20	**Bernhard Frick** *(invited)* Introduction to Neutron Backscattering Spectroscopy and overview over instrument developments and applications	**Anita Girelli** *(invited)* Diffusion during protein phase separation probed with x-ray and neutron scattering	**Romain Sibille** *(invited)* High-resolution neutron spectroscopy of low-energy excitations in quantum spin ice materials	**Juan Colmenero** *(invited)* Neutron spectroscopy with polarisation analysis: the key to unveil coherent scattering of liquids and glass-forming systems at the mesoscale
9:20-9:40				
9:40-10:00	**Goran Nilsen** *(invited)* Recent developments in polarized neutron spectroscopy at ISIS	**Wolfgang Doster** High resolution problems with biological samples	**Emma Lenander** Magnetically frustrated dynamics on the Cairo pent agonal lattice	**Dimitrios Bessas** Phonon spectroscopy using X-rays and a nuclear resonance
10:00-10:20		**Masato Matsuura** High-resolution measurements on inelastic and quasielastic neutron scattering at TOF near backscattering spectrometer DNA for hardmatters	**Danny Mannix** Exploiting polarised neutron and ultrafast X-ray pump-probe synergies to reveal the spin and heat dynamics in spin caloritronics	**Y Z** Unusual dynamics of tetrahedral liquids caused by the competition between dynamic and chemical heterogeneity
10:20-10:40	**COFFEE**			
	Session Chair: F. Mezei	**Session Chair: P. Deen**	**Session Chair: R. Biehl**	**Session Chair: J.M. Zanotti**
10:40-11:00	**Peter Falus** *(invited)* Neutron Spin Echo on WASP	**Lukas Beddrich** Comparison of time-of-flight with MIEZE spectroscopy of H_2O: Necessity to go beyond the spin-echo approximation	**Michihiro Nagao** Collective lipid acyl tail dynamics and membrane viscosity	**Antonio Faraone** Q-dependent collective relaxation dynamics of glass-forming liquid $Ca_{0.4}K_{0.6}(NO_3)_{1.4}$
11:00-11:20		**Olaf Holderer** SNS-NSE: First decade of operations	**Antonino Calio** Unravelling the mechanisms of adaptation to high pressure in proteins	**Henrich Frielinghaus** The dynamics of ionic liquids in mild confinement
11:20-11:40	**Bela Farago** *(invited)* Matching NSE and TOF together	**Nicolas R de Souza** Emu µeV-resolution neutron spectrometer at ANSTO: Capabilities and complementarities	**Felix Roosen-Runge** Structure and dynamics during hydrophobic collapse of elastin-like polypeptides	**Jon Maiz** Exploring the impact of composition and cross-linking degree on single-chain nanoparticles derived from
11:40-12:00		**Philipp Gutfreund** Rheo-Neutron Spin Echo spectroscopy	**Alessandro Paciaroni** Diffusive dynamics of bacterial proteome as a proxy of cell death	**Maksim Plekhanov** QENS study of citrate dynamics on the surface of iron oxide nanoparticles
12:00-14:00	**LUNCH**			
	Session Chair: B. Frick	**Session Chair: V. Garcia Sakai**	**Session Chair: A. Arbe**	**End of meeting**
14:00-14:20	**Fankang Li** *(invited)* The Larmor labeling program at ORNL for high resolution neutron diffraction and spectroscopy.	**Rony Granek** *(invited)* Dynamic structure factor of undulating vesicles: Finite-size, spherical geometry, and membrane viscosity effects with application to neutron spin echo experiments	**Margarita Kruteva** *(invited)* Different aspects of polymer dynamics probed by neutron spin-echo and PFG NMR	
14:20-14:40				
14:40-15:00	**Pascale Deen** *(invited)* The spectroscopy suite of the ESS	**Virginie Marry** *(invited)* Water dynamics in clays: coupling molecular dynamics and NSE experiments	**Quentin Berrod** *(invited)* Energy related materials probed with neutrons: electrolytes, nanoconfinement and multiscale dynamics	
15:00-15:20				
15:20-15:40	**F. Schreiber, F. Roosen-Runge** Round Table: Future of NSE at ESS?	**COFFEE**		
		Session Chair: H. Frielinghaus	**Session Chair: A. Faraone**	
15:40-16:00	**COFFEE**	**Arantxa Arbe** Collective dynamics and self-motions in the van der waals liquid tetrahydrofuran from meso- to inter-molecular scales disentangled by neutron spectroscopy with polarization analysis	**Fanni Juranyi** TOF and BS synergies to deal with increasing complexity in materials research	
16:00-16:20	**Guidehall visits**	**Ralf Biehl** Polyelectrolytes: Interchain hydrodynamic interaction and internal friction	**Peter Fouquet** Hydrogen mobility and reactivity in MoS_2 catalyst	
16:20-16:40		**Laura-Roxana Stingaciu** Probing the secrets of life with neutron scattering: high resolution spectroscopic techniques to unveil dynamics insights into proteins, living cells, and plants	**Kanming Shi** Molecular dynamics in the metal halide perovskites $FASnX_3$ (X = Cl, Br, I) investigated using quasi-elastic neutron scattering	
16:40-17:00			**Lukas Fine** Hydrogen dynamics in the nitride-hydride Ca_3CrN_3H	
17:00-17:20		**Poster session** **with “aperitive dinatoire”**		
19:30-22:30			**Workshop dinner** **in the restaurant Epicurien in the city center**	

Tuesday 12 December 2023

Introduction to Neutron Backscattering Spectroscopy and Overview over Instrument Developments and Applications

Bernhard Frick
Institut Laue-Langevin, Grenoble

Session: Instrumentation/Soft Matter and Liquids/Biology/Materials/Magnetism

We briefly introduce the basics of neutron backscattering spectroscopy and its history. We address instrumental progress which was achieved during more than 55 years of its existence, give an overview over the existing instrument suite on reactors and spallation sources, mention its complementarity with other spectroscopic techniques and refer to new backscattering instrument projects.

The range of scientific applications of neutron backscattering spectroscopy is today very broad. Based on some typical examples, selected from a large number of published backscattering experiments, we try to review science and trends.

"Recent developments in polarized neutron spectroscopy at ISIS"

Goran Nilsen
ISIS Neutron and Muon Source, goran.nilsen@stfc.ac.uk

Wide-angle neutron spectroscopy provides unprecedented insight into the structural and magnetic dynamics of materials over broad ranges in space and time. In some cases, however, it is difficult to identify the dynamics of interest in the presence of other scattering contributions: to give one example, the incoherent and coherent quasi-elastic components in the spectra of deuterated molecular liquids can be similar in magnitude, making either component difficult to distinguish. Polarization analysis provides a solution to this problem by permitting the separation of the neutron cross section into its components - nuclear coherent and isotope incoherent, spin incoherent, and magnetic – and it has therefore been a long-standing goal in the community to add polarized capabilities to wide-angle spectrometers. Since the final technical obstacles to this were overcome in recent years, polarized capabilities have been installed on several spectrometers, including LET at ISIS. Early experiments have demonstrated the potential of the technique for a range of applications across physics, chemistry, and biology.

In this talk, I will briefly introduce the concepts behind polarized wide-angle spectroscopy, using its implementation on the LET as an example [1]. This will be followed by a review of several recent scientific highlights from the same instrument, including work on collective dynamics in molecular liquids and mixtures [2-4], ionic transport in a battery electrolyte material, and magnetic diffuse scattering in a frustrated magnet [5]. I will conclude by discussing some future perspectives for polarized wide-angle spectroscopy, focusing mainly on a new concept indirect spectrometer, SHERPA [6].

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Neutron Spin Echo on WASP

Peter Falus
Institut Laue-Langevin, Grenoble, France

Neutron Spin Echo (NSE) instruments are the highest energy resolution neutron spectroscopy instruments. In neutron spectroscopy the difference between the incoming and outgoing energy of a scattered neutron is to be measured. Other spectroscopy techniques need to monochromatize either the incoming or the outgoing neutron beam or both, losing neutron flux. Neutron spin echo encodes the energy difference in the neutron spin using magnetic fields. As a result the intensity is preserved even when using very high energy resolutions. WASP the Wide Angle Spin echo is the youngest member of the NSE family. WASP's magnetic configuration is unique, it is the single functioning instrument of its kind. Covering a similar wavevector range to time of flight or backscattering instruments yet having higher resolution, it is ideally suited to be combined with other spectroscopy techniques. After an introduction to Neutron Spin Echo and the WASP instrument specifically, it will be shown on practical examples how the very wide angle coverage and high intensity of this flagship instrument opens new science areas for exploration.

Matching NSE and TOF together

Bela Farago
Institute Laue Langevin, Grenoble

Session: Instrumentation

NSE and TOF are great complementary techniques. TOF mesures in omega space while NSE measures the fourier transform, thus the time dependence of the intermediate correlation function. While NSE extends the energy resolution by up to three orders of magnitude, there is a quite substantial overlap between the two. In addition as NSE always uses polarised beam, beside forurier transform, other subtle differences need to be taken into account to find full agreement in the complementary measurments.

These aspects will be discussed through some new experimental results

The lamor labelling program at ORNL for high resolution neutron diffraction and spectroscopy

Fankang Li

Neutron Technologies Division, Oak Ridge National Laboratory, Oak Ridge, TN, 37830, USA
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Conventional neutron scattering techniques encounter limitations when measuring scattering events characterized by small momentum or energy transfers due to the inherent trade-off between resolution and useable flux. To address this challenge, we explore the innovative utilization of neutron spin Larmor precession. By harnessing this phenomenon, it becomes possible to encode neutron momentum or energy changes as substantial shifts in the total Larmor phase, thereby circumventing the resolution-useable flux trade-off. This approach holds the potential to enhance the resolution of established techniques such as small angle neutron scattering, neutron diffraction, and inelastic neutron scattering. A pivotal element for effectively encoding neutron beams within the momentum-energy phase space is the magnetic Wollaston prism (MWP). In this presentation, I will introduce the advancement of superconducting MWPs and showcase their application in Larmor labeling of neutron spin in the space of both momentum and energy transfer. Through compelling demonstrations across various neutron scattering methodologies, we illustrate the transformative impact of this technique, opening new avenues for high-resolution investigations in neutron research.

The Spectroscopy suite of the ESS

Felix Villacorta[1,2], **Pascale Deen[2]**, Daria Noferini[2], Rasmus Toft Peterson[3,2],
Christian Franz[4,2] and Monica Hartl[2]

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Session: Instrumentation

Uniquely, the ESS is a pulsed spallation source with a peak brightness that matches the most intense neutron sources worldwide and a long pulse that provides great possibilities to trade neutron flux for resolution in energy and momentum transfer. The enhanced fluxes will provide new opportunities for neutron scattering with a particular focus on time dependent phenomena and novel states of matter under extreme conditions.

The ESS are constructing 16 instruments for day 1 with a current beam on target date of May 2025. The spectroscopy instruments combined cover nearly 6 orders of magnitude in energy transfer and three orders of magnitude in wave vector transfer. The spectroscopy instruments are, in order of accessible energy transfer, MIRACLES, a back scattering spectrometer, CSPEC, the cold chopper spectrometer, BIFROST, an extreme environment indirect geometry spectrometer, TREX, a thermal chopper spectrometer, and VESPA, a vibrational spectrometer.

I will provide an overview of the day 1 spectroscopy suite of the ESS with particular focus on their high energy resolution capabilities, novel capabilities and updates on their construction. A consideration of possible future instruments will also be presented.

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Wednesday 13 December 2023

Diffusion during protein phase separation probed with x-ray and neutron scattering

Anita Girelli

Department of Physics, Stockholm University, Sweden

Biological systems are often hierarchical, and require to investigate solutions from a molecular level to macroscopic length scales to have a complete understanding. As a consequence, accessing the diffusion of their components requires to probe timescales from picoseconds to hundreds of seconds. This broad range of time and length scale cannot be accessed with a single technique, but with combination of neutron, light and x-ray scattering. We investigated the diffusion of a model system for liquid-liquid phase separation (LLPS). The solution of γ-globulin and PEG spontaneously separates and reaches a gel state due to the high concentration of the more concentrated of the two phases. At LLPS domains length scale, x-ray photon correlation spectroscopy (XPCS) reveals correlation functions which show typical characteristics of ballistic motion as seen for colloidal gels [1]. The experimental results are compared with simulated data based on the Cahn-Hilliard equation. Qualitative comparability is obtained by adding a strong dependence of the protein mobility on protein concentration. The validity of this assumption is confirmed by probing the diffusion on the protein length scale and below with neutron quasi-elastic scattering and XPCS [2,3]. The comparison of dynamic light scattering and XPCS shows the feasibility of measuring on molecular length scale and time as short as 220 ns.

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High resolution problems with biological samples

Wolfgang Doster
Technical University Munich, Physics Department and FRM 2, Garching, Germany

Session: Biology

Instead of “high resolution” I prefer “appropriate resolution” in a relevant time window. Biological samples are mostly aqueous solutions. Even with D_2O, there is a dominant water background scattering to be subtracted. The fraction of coherent to incoherent scattering varies with Q [1]. The resolution of protein internal dynamics is further limited by translational and rotational diffusion and the small accessible temperature range. These restrictions can be overcome, using hydrated samples: no diffusion, small water signals and a wide temperature range due to non-freezable water with stable structures and bio-activity [2]. Biological samples often come in small concentrations, mg/ml range, while reasonable inelastic signals and beam time require some 50 mg/ml. This problem can be partially overcome by focussing on the strong elastic component at variable resolution [3]. Merging data of several spectrometers in the time domain, reveals two dynamic components [4,5] as demonstrated with casein solutions [6,7]. The natively-disordered protein shows a third slow dynamic component, in excess to those of hydrated globular proteins.

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High-resolution measurements on inelastic and quasielastic neutron scattering at TOF near backscattering spectrometer DNA for hardmatters

M. Matsuura[1], T. Yamada[1], T. Tominaga[1], H. Tamatsukuri[2], K. Ohuchi[1], H. Nakagawa[2,3] and Y. Kawakita[2]
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Session: Instrumentation

DNA is a TOF type Si crystal analyzer backscattering spectrometer installed at the Japan Proton Accelerator Research Complex (J-PARC) spallation source. DNA uses a pulse-shaping chopper to extract sharp pulsed neutron from strong but broad spectrum of coupled moderator, which achieves good energy resolution (a few μeV) and wide dynamic range up to 1.5meV. Furthermore, the signal-to-noise ratio at DNA achieves ~10^5, which enables to detect small scattering amplitudes or allow small quantities of samples. Using these characteristics features, DNA is used for precise measurements of QENS as well as INS in a wide range of research fields from biomaterials, ionic conductors, soft materials, magnetism and functional materials. Although the energy resolution near $E=0$ is not as good as that of backscattering spectrometers at reactors, DNA can offer the highest energy resolution data for inelastic scattering due to the good energy resolution in wide energy range. We will show typical examples of QENS and INS measurements utilizing these characteristic features of DNA with focus on physics of condensed matters.

Comparison of time-of-flight with MIEZE spectroscopy of H2O: Necessity to go beyond the spin-echo approximation

L. Beddrich[1], J. K. Jochum[1], P. Bender[1], L. Spitz1,2, A. Wendl[1,3], C. Franz[4],
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We report a comparison of high-resolution neutron spectroscopy of pure water at 300 K as performed at the ToF spectrometer TOFTOF (FRM II) with MIEZE spin-echo spectroscopy as performed at RESEDA (FRM II). We introduce a theoretical framework beyond the spin-echo approximation to transform arbitrary dd from the energy to the time domain. Using this framework, we analyze the inelastic neutron scattering data in two ways: (1) The ToF spectra were transformed directly into the time domain. (2) Describing dd in terms of several Lorentzian distributions, both data sets were described in terms of an individual and a combined function, taking the instrumental resolution into account.

Within both approaches good agreement is achieved, where discrepancies suggest the need for an improved understanding of instrumental resolution, neutron detection efficiency over a broad range of energy transfers, and background scattering. As instrument performance proves to be key for a rigorous comparison of various neutron spectroscopic techniques, advanced computational modelling of real systems, such as molecular dynamics simulation, in conjunction with instrument modelling, such as Monte-Carlo simulations of neutron propagation, promise to disentangle instrument peculiarities from sample-specific interactions.

SNS-NSE: First Decade of Operations

Piotr Zolnierczuk[1], Laura Stingaciu[1], **Olaf Holderer**[2],
Tadeusz Kozielewski[3], Dieter Richter[3] and Michael $^{\text{Monkenbusch3}}$

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Session: Instrumentation

High resolution neutron spin echo (NSE) spectroscopy is the highest energy resolution technique involving cold neutrons [1]. The SNS-NSE spectrometer [2] located at the BL-15 of the Spallation Neutron Source, Oak Ridge National Laboratory is the first, and to date, the only one “classic” NSE spectrometer installed at a pulsed neutron source. It is also the first successful, user serving, high resolution NSE instrument to employ superconducting coils.

The SNS-NSE entered the SNS user program 2010 and is currently one of the two NSE instruments available in Americas. We will present the history and the parameters of the spectrometer along with examples of science results. The operation of a NSE spectrometer at a pulsed source presents additional challenges and opportunities to the data acquisition and reduction [3] that will be addressed as well.

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A portion of this research used resources at the Spallation Neutron Source, a DOE Office of Science User Facility operated by the Oak Ridge National Laboratory. Parts of the research were supported by Forschungszentrum Jülich GmbH, Germany.

Emu μeV-resolution neutron spectrometer at ANSTO: Capabilities and complementarities

Nicolas R de Souza
ACNS, Australian Nuclear Science and Technology Organisation, Australia

The Australian Centre for Neutron Scattering, ANSTO, operates the Emu backscattering spectrometer installed at the OPAL reactor since 2016. The spectrometer is set up with Si (111) analyzers in near-perfect backscattering on the primary and secondary neutron flight paths so as to deliver a 1 μeV FWHM energy-transfer resolution, and with a Doppler drive offsetting the incident energy by up ±31 μeV [1]. Emu is well-suited for quasielastic neutron scattering (QENS) and low-lying excitation spectroscopy, with good sensitivity to hydrogenous species. In favorable cases, sensitivity to weaker scattering elements such as e.g. D, Na, O, … is also achieved [2,3]. The most-requested sample environment is a 1.5…750K cryofurnace in optional combination with other sample conditioning schemes such as gas delivery, pressure, light irradiation, electrical field, … Other sample environment equipment may be requested or jointly developed. Beamtime allocation is merit-based.

Applications to various areas will be presented to highlight the versatility of the spectrometer. At present studies addressed solid-state dynamic disorder to liquid-state dynamics [2-7], including a variety of soft-matter and composite phases from polymer and the bio- sciences, from the fundamental to the applied domains.

Emu data are typically combined with data from other neutron spectrometers and other techniques, soft-matter studies accounting for the bulk of experiments to date. Recent works highlight the complementarity with time-of-flight spectroscopy [6,4,3] and neutron polarization analysis [5] in the Asia-Pacific region. In light of the recent scarcity of neutrons for quasi-elastic scattering, complementarity with spectrometers in the Europe and United States is also addressed.

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Rheo-Neutron spin echo spectroscopy

Philipp Gutfreund[1], Manuchar Gvaramia[2], Peter Falus[1] , Tim Gunter[2] and Max Wolff[2]

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Session: Instrumentation-Soft Matter and Liquids

Rheology serves as a valuable method for investigating the viscoelastic properties of polymer materials. By conducting oscillatory shear experiments and measuring the complex modulus as a function of deformation and frequency, we can gain insights into how a sample responds to shear at the macroscopic level. Theoretical models can be applied to establish connections between these properties and microscopic time and length scales, such as the longest relaxation time or the tube diameter in the reptation model. Neutron spin echo (NSE) spectroscopy measurements provide a means to access the microscopic quantities related to viscoelastic properties and have been used in verifying the reptation model and its extensions, including contour length fluctuations and convective constraint release. Viscoelastic properties of polymers are divided into three regimes based on time scales: linear, nonlinear, and intermediate. In the linear regime, polymers follow the reputation model at low shear rates. At high shear rates, the Weissenberg effect occurs, causing conformational changes. Small angle neutron scattering (SANS) can probe these changes. The intermediate regime exhibits strong non-linearities, requiring models like convective constraint release and chain disentanglement. NSE experiments combined with rheology are needed to study these effects. To facilitate these combined measurements, we have developed a shear cell specifically designed for NSE and small-angle neutron scattering (SANS) experiments. This shear cell is capable of loading and shearing high-viscosity polymer melts and has been optimized for NSE and SANS measurements. By rotating the sample between a single crystalline silicon wafer and a ceramic rotor, we can prevent SANS signals originating from the shear cell itself. Additionally, all components of the cell are non-magnetic to prevent depolarization of the neutron beam during the NSE experiments. With this setup, shear rates of up to 200, 1/s can be achieved, which surpasses the longest relaxation time for many entangled polymer blends. However, it is worth noting that at higher shear rates, the velocity of the rotating cone and the sample reaches values that may introduce inelastic scattering, potentially affecting the NSE measurements. In our presentation, we demonstrate that this Doppler scattering can be accurately calculated for realistic experimental conditions, and can be accounted for during data reduction. We will showcase a combination of calibration measurements, simulations, and theoretical calculations as well as first measurements on polymer melts, blends, and wormlike micelles to illustrate these concepts.

Dynamic structure factor of undulating vesicles: Finite-size, spherical geometry, and membrane viscosity effects with application to neutron spin echo experiments

Rony Granek[1], Ingo Hoffmann[2], Elizabeth G. Kelley[3], Michihiro Nagao[3], Anton Zilman[4], Hammad A. Faizi[5], Petia M. Vlahovska[6]

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Session: Soft Matter and Liquids

We consider the dynamic structure factor of quasi-spherical vesicles, and present a generalization of an expression that was originally formulated by Zilman and Granek (ZG) for scattering from isotopically oriented quasi-flat membrane plaquettes [1,2]. The expression is obtained in the form of a multi-dimensional integral over the undulating membrane surface. It reduces to the original ZG formula in the limit of large wavenumbers q, provided that the vesicles are sufficiently large – in the micron range or larger – known as giant unilamellar vesicles. For much smaller unilamellar vesicles, deviations from the original ZG expression are noticeable even if one assumes that the Seifert-Langer (SL) monolayer density mode is completely relaxed and membrane viscosity is neglected. To avoid the need for an exhaustive numerical integration while fitting to neutron spin echo (NSE) data, we provide a useful approximation that tests well against numerical integration of the complete formula. Applying the approximation to NSE data sets recently obtained at ILL demonstrates an advantage of the present approach over other approximations that have been developed and deployed over the years to fit the NSE data.

We also consider theoretically the effect of membrane viscosity on the undulation relaxation frequencies, and moreover generalize the SL model for quasi-spherical vesicles [3]. Membrane viscosity is shown to be important for the small vesicle sizes studied by NSE and affects in general the longer wavelength undulation modes. We show that for typical parameter values the SL density mode may be slowly relaxing during the NSE time window, which may further complicate the NSE data interpretation.

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Water dynamics in clays: coupling molecular dynamics and NSE experiments

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Session: Soft Matter and Liquids

Clays are porous lamellar materials with remarkable adsorption and retention properties, making them good candidates for environmental applications such as water pollution control or waste storage (radioactive waste, CO_2). In geological environments, the retention properties of mobile species are strongly influenced by the behavior of the water present in the medium.

Classical molecular simulations make it possible to describe and quantify diffusion processes in water/clay systems at the molecular level, as long as the interactions between atoms are well defined. At the same scale of observation, QENS experiments provide valuable information on water dynamics in these systems, provided a reliable analytical model can be found to analyze the data. I will show how the NSE model/experiment coupling helps to choose the right analytical model, to better interpret the experiments and their evolution with temperature, but also to show the limits of molecular simulations.

Collective Dynamics and Self-Motions in the Van Der Waals Liquid Tetrahydrofuran from Meso- to Inter-Molecular Scales Disentangled by Neutron Spectroscopy with Polarization Analysis

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Session: Soft Matter and Liquids

By using time-of-flight neutron spectroscopy with polarization analysis, we have separated coherent and incoherent contributions to the scattering of deuterated tetrahydrofuran in a wide scattering vector (Q)-range from meso- to inter-molecular length scales [1]. The results are compared with those recently reported for water [2] to address the influence of the nature of inter-molecular interactions (van der Waals vs hydrogen bond) on the dynamics. The phenomenology found is qualitatively similar in both systems. Both collective and self-scattering functions are satisfactorily described in terms of a convolution model that considers vibrations, diffusion, and a Q-independent mode. We observe a crossover in the structural relaxation from being dominated by the Q-independent mode at the mesoscale to being dominated by diffusion at inter-molecular length scales. The characteristic time of the Q-independent mode is the same for collective and self-motions and, contrary to water, faster and with a lower activation energy (≈1.4 Kcal/mol) than the structural relaxation time at intermolecular length scales. This follows the macroscopic viscosity behavior. The collective diffusive time is well described by the de Gennes narrowing relation proposed for simple monoatomic liquids in a wide Q-range entering the intermediate length scales, in contraposition to the case of water.

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Polyelectrolytes: Interchain hydrodynamic interaction and internal friction

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Session: Soft Matter

Polyelectrolytes are widespread in nature and technical applications. Their properties are mainly determined by their intrinsic charge and the surrounding screening solvent including salt ions. Polyelectrolytes are of general interest resulting in a lot of publications mainly describing macroscopic properties. Theoretical models are often based on simplifications like full hydrodynamic screening to deal with problems like hydrodynamics or counterion condensation. A connection of macroscopic properties to molecular structure and dynamics is missing.

Neutron spinecho spectroscopy (NSE) is a unique technique to access the chain dynamics on a molecular level which we use in combination with dynamic light scattering (DLS) and PFG-NMR. We connect macroscopic diffusion to molecular chain dynamics revealing a decoupling of local chain dynamics from interparticle interactions. Collective diffusion is described within a colloidal picture including electrostatic and hydrodynamic interactions. Chain dynamics is characterized by the classical Zimm model of a neutral chain retarded by internal friction due to condensed counterions.

We think that this new approach opens new possibilities to found the theory of polyelectrolytes from a molecular level based on experimental data from neutron spinecho spectroscopy that validate the approach.

Probing the Secrets of Life with Neutron Scattering: high resolution spectroscopic techniques to unveil dynamics insights into proteins, living cells, and plants

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The studies presented here delve into the fascinating realm of dynamic molecular processes within biological entities, utilizing cutting-edge neutron spin echo and backscattering techniques. With a focus on proteins, living cells, and plants, will explore the intricate motions and interactions that are fundamental to the very essence of life. Through these innovative methods where neutrons illuminate the secrets of motion at the atomic level, will take the biophysics approach to unravel the hidden internal dynamics of biomolecules and bio-membranes, shedding light on essential biological functions and potential applications in fields ranging from medicine to agriculture.

Thursday 14 December 2023

High-resolution neutron spectroscopy of quantum spin ice materials

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Magnetic systems with competing interactions often adopt exotic ground states, which can be relevant to study new physics in quantum matter [1]. A recurrent ingredient to stabilize such phases is frustration, such as in pyrochlore oxides where rare-earth magnetic moments decorate a lattice of corner-sharing tetrahedra. In the pyrochlore Ho2Ti2O7 for instance, a local constraint – the 2-in-2-out 'ice rule' acting on each tetrahedron [2,3] – leads to a manifold of degenerate ground states in which the spin correlations give rise to emergent magnetostatics [4]. Spin flips violating the ice rule generate magnetic monopole excitations [5], a mobile magnetic charge regarded as a quasiparticle carrying half of the dipole moment. A quantum analogue of the spin ice state is predicted to be a special type of quantum spin liquid formed through the coherent superposition of spin ice configurations [6,7]. Remarkably, the low-energy physics of this quantum spin ice state is predicted to be a lattice analogue of quantum electrodynamics. We review recent results that aim at tracking the experimental signatures of quantum mechanical ground states in rare-earth pyrochlores based on Pr3+ [8-9] or Ce3+ [10-13]. We discuss the importance of high-resolution neutron spectroscopy to unveil and distinguish the signatures of the different types of excitations expected in quantum spin ice materials: gapless 'magnetic photons' and gapped electric/magnetic monopoles. Using neutron backscattering spectroscopy, we show how the density of states of the gapped excitations can be understood in the light of recent theoretical studies [14-16].

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Magnetically frustrated dynamics on the Cairo pentagonal lattice

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The octahedrally and tetrahedrally coordinated Fe^{3+} (S=5/2) ions in $Bi_2Fe_4O_9$ form a quasi two-dimensional Cairo pentagonal lattice (Pbam). Combined with predominantly antiferromagnetic interactions, this leads to a strong frustration with T_n=245 K while θ_{CW}=1670 K in a fairly unexplored geometry. The magnetic structure for $T<T_n$ can be indexed with k=(½, ½, ½) with a noncollinear magnetic structure of Fe_1 and Fe_2 moments, 2 different sites, and an interpenetrating pattern of fourfold spin rotations, a very novel magnetic state. Previous measurements studying the low-lying excitations, 0–30 meV, in the ordered phase were measured on small single crystals (< 0.6 g). Beauvois et al. (PRL2020) argues that no anisotropy is required while Duc Le et al. (PRL2021) indicates an easy-plane single-ion anisotropy. Accordingly, Beauvois find the acoustic AF mode is not gapped while it is for Duc Le. We have synthesized a 2.35 g, high quality single crystal of $Bi_2Fe_4O_9$ and are re-examining the magnetic excitations. At low energy transfers we see distinct discrepancies between our data and previous data in the ordered phase using CAMEA (PSI). Our data show a clear double spin gap, that allows us to differentiate axial and planar anisotropy scenarios. We have also taken data at higher energies at EIGER (PSI) and IN20 (ILL). This has enabled us to establish the microscopic model at low temperatures and we will later perform quantitative analysis of the frustrated state above T_n.

Exploiting polarised neutron and ultrafast X-ray pump-probe synergies to reveal the spin and heat dynamics in spin caloritronics

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Session: Magnetism

Spin caloritronics are currently a science highight due to their potential exploitation in the next generation of spintronics applications. Composite systems, consisting of magnetic-insulator (MI) and heavy-metal (HM) bilayers, they combine both spintronic and thermoelectric functionalities by interconversion of charge, spin, and heat currents. A prominent example are devices exploiting the spin Seebeck effect (SSE), where thermoelectric generation is achieved by a thermally induced spin-current, which is then converted into an electric charge current by the inverse spin Hall effect within the HM layer. At low temperatures the generation of a net spin current in the MI can be understood in terms of thermal excitation of chiral magnons [1]. Therefore, the development of emerging technologies based on spin caloritronic physics, requires a high resolution microscopic understanding of their magnetic and heat dynamics. We present new insights into the dynamics of spin caloritronic materials, by exploiting the synergies of polarised neutron inelastic scattering at IN20 of the ILL and ultrafast X-ray pump-probe techniques using the LCLS XFEL. These studies were applied to prototype spin caloritronic materials, based on the rare-earth iron garnets of $Tb_3Fe_5O_{12}$ and $Gd_3Fe_5O_{12}$ (TbIG and GdIG). Polarised neutron inelastic scattering studies of single crystal TbIG can be correlated with the low energy chiral magnon modes responsible for the SSE in this material. In addition, our theoretical calculations highlight the influence of the crystal field anisotropy on the energy regime of these chiral magnon modes [2]. Femtosecond X-ray pump-probe techniques were used to study the thermal [3] and magnetic dynamics induced in GdIG thin epitaxial films, using resonant X-ray magnetic scattering techniques. These results open new routes to investigate acoustic wave induced magnetic excitations with photon energy resolution beyond the current limits of RIXS. This work has motivated a new proposal accepted on IN20, to study a 160Gd isotope sample of GdIG.

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Collective lipid acyl tail dynamics and membrane viscosity

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Session: Soft Matter and Liquids or Biology

Lipid molecular motions determine the membrane properties on local and global scales. Here we aim to link the molecular and macroscopic motions by employing neutron spin echo and quasielastic neutron and gamma-ray scattering techniques to measure the collective lipid acyl tail dynamics on length scales of approximately 0.5 nm and timescales ranging from ps to hundreds of ns. There are two relaxation processes in the fluid phase with relaxation times of the order of 10 ps and 100 ps. The fast mode originates from density fluctuations of lipid acyl tails and are about an order of magnitude slower than those of the bulk liquid alkanes with the corresponding number of carbons. This slowdown is attributed to the effects of two-dimensional confinement and associated restrictions of the molecular orientations. The slower relaxations are related to the diffusive motions of the lipid molecules. Meanwhile, in the gel phase, the lipid acyl tail dynamics are quite heterogeneous, and the relaxation processes span 4 to 5 orders of magnitude in time. The average relaxation time changes from 10s ns to 100s ns with decreasing temperature from the lipid main transition temperature. More recently we have also begun to combine neutron time-of-flight and backscattering measurements to also study the fast dynamics in the lipid fluid phase. These efforts include separating the coherent and incoherent contributions to the measured dynamics using polarized QENS. We will showcase results for saturated phosphatidylcholine lipids with different tail lengths and compare the measured relaxation dynamics with reported values for the membrane viscosity in literature.

Unravelling the mechanisms of adaptation to high pressure in proteins

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Session: Biology

The adaptation of proteins to high pressure is still an open debate, but understanding it could shed light on the origins of life [1], lead to a better knowledge of protein dynamics, and deliver new tools to engineer pressure-resistant enzymes for biotechnological purposes. While the thermodynamic and dynamical properties of model proteins under pressure have been extensively studied [2], the evolutionary aspects of their adaptation are still unclear.

Disentangling the contributions of pressure adaptation from those of another adaptation, such as high or low temperature, is a difficult task. In fact, genomic studies could not determine a clear pattern among the order of Thermococcales.

Recent experiments by our group focused on whole cells of two closely related species (*Thermococcus barophilus*, Tba, and *Thermococcus kodakarensis*, Tko) that grow at the same optimal temperature (85°C) but differ only for the optimum pressure (400 bar for Tba, 1 bar for Tko), and they highlighted the differences in the dynamics of the two organisms' proteomes [3]. To take this investigation to the molecular level, we studied the *Phosphomannose Isomerase* and the *Ribosomal protein S24e* from the two organisms with Elastic and Quasi-elastic Incoherent Neutron Scattering, 2-D NMR Spectroscopy, X-ray crystallography and Molecular Dynamics Simulations. Our results evidence that the substitutions of amino acids enhancing pressure stability are those in the hydrophobic core, which eliminate cavities, and those on the surface, which modulate the interaction of the proteins with the surrounding water layer and give them the right flexibility to perform their function under high pressure. Therefore, the study of the dynamics of these proteins enabled us to gain detailed structural information, to describe their behaviour under extreme conditions and characterize their adaptation.

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Structure and dynamics during hydrophobic collapse of elastin-like polypeptides

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Session: Soft Matter and Liquids/Biology

Elastin-like polypeptides (ELPs) are versatile responsive biopolymers intensively used in biomedical applications such as drug delivery, protein purification and tissue engineering in the last decade. ELPs mimic a hydrophobic amino acid repeat segments of elastin which is a key protein of the extracellular matrix, and provides elastic properties to biological tissues – such as lung and ligaments – with extraordinary long-term stability and resilience. ELPs undergo a hydrophobic collapse upon crossing a lower critical solution temperature, which can cause compaction of individual chains, and the formation of ELP condensates. Although key to the elasticity of elastin and the stimulus-response of ELPs, a comprehensive mechanistic characterization of the collapse in terms of dynamical and structural evolution is so far missing.

We report on an integrative research program combining dynamic and static scattering techniques with computer simulations. To study the interaction between peptide segments, we first study a short ELP chain of 18 amino acids. From dynamic light scattering (DLS), we obtain a clear signature of condensation, which is also supported by an increasing size using small-angle neutron scattering (SANS). Computer simulations evidence that these short ELPs show a temperature-induced attraction and form transient complexes [1]. Interestingly, the chains remain extended in simulations, while at the same time the complexes appear more compact after collapse based on the scattering signature. Finally, the chain dynamics explored by quasi-elastic neutron scattering (QENS) shows no strong dynamic transition, evidencing very dynamic, fluid-like assemblies.

Experimental signatures for longer ELP chains show a more diverse behavior with significant effects both on structure and dynamics as seen from QENS, SANS and DLS. Despite stronger chain collapse, we still obtain a high dynamic flexibility, rejecting earlier explanations for the hydrophobic collapse based on the formation of specific secondary structures.

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Diffusive Dynamics of Bacterial Proteome as a Proxy of Cell Death

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Session: Biology

Temperature variations have a big impact on the bacterial metabolism and death, yet an exhaustive molecular picture of these processes is still missing. For instance, whether the thermal death is determined by the deterioration of the whole or a specific part of the proteome is hotly debated. In our work [1], by monitoring the proteome dynamics of E. coli we show that only a minor fraction of the proteome unfolds at the cell death. First, we prove that the dynamical state of the E. coli proteome is an excellent proxy for the temperature dependent bacterial metabolism and death. The proteome diffusive dynamics peaks at about the bacterial optimal growth temperature, then a dramatic dynamical slowdown is observed which starts just below the cell's death temperature. Next, we show that this slowdown is caused by the unfolding of just a small fraction of proteins which establish an entangling inter-protein network—dominated by hydrophobic interactions— across the cytoplasm. Finally, we prove that the deduced progress of the proteome unfolding and its diffusive dynamics are both key to correctly reproduce the E. coli growth rate.

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Different aspects of polymer dynamics probed by neutron spin-echo and PFG NMR

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Session: Soft Matter

The high resolution quasielastic neutron scattering (QENS) is a relevant method to investigate the dynamics of polymer over broad time and length scale typically covering very local segmental motions and chain dynamics. Pulsed field gradient (PFG) NMR complements QENS at longer time and length scale. In my talk I will present a few examples of synergy of QENS and PFG NMR to study the polymeric systems. Different aspects of polymer dynamics in the melt state will be addressed. For relatively short chains the Rouse model provides a good description with physically realistic parameters of the chain dynamics. Dynamics of long or strongly entangled chains can be successfully described by the reptation model. The intermediate range of polymer chain lengths is less understood. By neutron spin echo (NSE) and PFG NMR we study the dynamics of a polymers with molecular weight in the transition regime between Rouse and reptation dynamics [1,2].
The relaxation dynamics of ring polymers is fundamentally different to linear analogous, where chain relaxation takes place via the chain ends leading to reptation, contour length fluctuation (CLF) and constraint release (CR). Small Angle Neutron Scattering (SANS) and NSE/PFG NMR results on very large polymer rings in the melt will be presented [3]. The ring conformation demonstrate a clear signature of the theoretically predicted elementary loops. Their size is in the range of an entanglement strand for a linear polymer melt and they are characterized by Gaussian statistics. Other than extracted from numerous simulations that are interpreted in terms of a cross over to mass fractal, we do not observe such a cross over.

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Energy related materials probed with neutrons: electrolytes, nanoconfinement and multiscale dynamics

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We investigate the physics of Ionic Liquids (ILs) doped with lithium salts in both bulk and when confined within vertically aligned Carbon NanoTubes (CNT) composite membranes. Recognized for their exceptional chemical and electrochemical stability, ILs have emerged as promising electrolytes for the development of secure and sustainable energy storage systems. When confined in a macroscopic 1D CNT scenario, we demonstrate a significant one-order-of-magnitude enhancement in the ionic conductivity of IL-based electrolytes. This suggests that CNT membranes offer a potential avenue to amplify Li^+ transport properties and consequently increase the specific power of solid-state batteries. In this context, we scrutinize the multiscale dynamics of IL-based electrolytes in both bulk and 1D CNT confinement, employing neutron scattering techniques (QENS, NSE, and backscattering at the molecular scale), PFG-NMR (at the micrometer scale), and electrochemical measurements (at the macroscopic scale). At the molecular scale, under confinement, the dynamics are activated at lower temperatures (between 10 and 20°C) compared to the bulk, accompanied by an increase in the long-range translational diffusion coefficient. Molecular dynamics simulations results allow us to attribute the conductivity improvement to a reorganization of the electrolyte's nanostructure under confinement.

TOF and BS synergies to deal with increasing complexity in materials research

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Session: Materials

As science moves towards more complex materials, the need for complementary information is increasing. I will present here two examples from different areas of materials science to showcase opportunities, and challenges, when combining time-of-flight and backscattering spectroscopy.

The first example is about ligand shell dynamics and elasticity of colloidal nanocrystals [1]. The combination of the two types of spectrometers allowed us to identify and quantify rotational ligand motions and extract the true, (temperature dependent) vibrational mean squared displacement, which, at the end reflects the mechanical coupling of the nanocrystals.

I will also present our work on proton dynamics in the hureaulite-type phosphatic oxyhydroxide $Mn_5[(PO_4)_2(PO_3(OH))_2](HOH)_4$ [2]. Here, we combined not only the two neutron spectroscopy measurements, but also dielectric spectroscopy, simulations and diffraction in order to understand the dynamics of the five distinct proton sites.

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Hydrogen mobility and reactivity in MoS2 catalyst

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Session: Materials

Catalyst materials are an essential component in hydrogen production and for the operation of fuel cells. Today, however, they are mostly based on platinum, which is expensive and rare. Molybdenum sulfide, MoS_2, which is more abundant and significantly cheaper, has shown interesting catalytic activity and has been a focus point of research in recent years [1]. MoS_2 is a van-der-Waals bonded 2d material that, theoretically, provides a high density of active sites, but little is known about the mobility and the reaction steps of hydrogen and water in MoS_2.

In this presentation we will discuss our recent studies on hydrogen mobility and reactivity in MoS_2 [2]. To shed light on the possible pathways for improving the performance of MoS_2 and other 2d catalyst materials, we have studied the diffusion of hydrogen and water by means of neutron scattering and X-ray photoelectron spectroscopy combined with nuclear reaction analysis and molecular dynamics simulations.

We observed a very fast hydrogen diffusion parallel to the basal planes and a very slow diffusion perpendicular to the MoS_2 basal planes. Water, on the other hand, cannot penetrate the perfect crystal, but can intercalate within volume defects, where it can access edge sites of the material. MD simulations were essential for the identification of the contributions of the different hydrogen species (H(+), H2, H2O) to the chemistry in MoS_2.

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Molecular dynamics in the metal halide perovskites FASnX$_3$ (X = Cl, Br, I) investigated using quasi-elastic neutron scattering

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Session: Materials

Metal halide perovskites (MHPs) are currently attracting considerable attention because of their promise for application in future photoelectric conversion devices. Formamidinium lead Iodide (CH(NH2)2PbI3) is one of the promising materials in this respect, but its instability to heat and humidity as well as the environmentally unfriendly nature of lead, makes it dubious for industrial commercialization. Increasingly evidence shows A-site cation dynamics relate to perovskite structure, electrical properties, stability, etc. Here we report on a study of the rotational dynamics of formamidinium cations in the lead free MHP FASnX3 with X = Cl, Br, or I, by using quasi-elastic neutron scattering . By monitoring the intensity of the quasi-elastically scattering upon heating the sample from 44 K to 350 K, we can find different kind of cation motions including 2-site jump diffusion and isotropic rotational diffusion at different temperature with picosecond scale residence/relaxation time. Relating to the phase transition temperature, FASnX3 has been observed that the QENS intensity undergoes the rapid changes in the temperature range of 200-250 K, where perovskite tetragonal *P4/mbm* phase (150 – 250 K) changed to cubic *Pm3m* phase (> 250 K). Meanwhile, under 150 K when perovskites exhibit the orthorhombic *Pnma* phase, we did not observe any significant QENS signals. Among this series of perovskites, FASnCl3 shows less QENS intensity than the others at the same temperature (< 350 K), especially under 200 K, it does not show a clear QENS signal. This might relate to its samllest unit cell and Cl- smallest ionic radius, which generate stronger dipole interaction to the FA+ limit the organic cation’s dynamics.

Hydrogen dynamics in the nitride-hydride Ca_3CrN_3H

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Session: Materials

Hydride-ion (H-) conductors attract more and more interest for their potential application as solid electrolytes in, e.g., batteries, fuel cells, and for catalysis. A particularly promising class of hydride-ion conductors are nitride-hydrides, which accommodate both nitride and hydride ions in the same substructure. Recently, a new nitride-hydride Ca3CrN3H was discovered and shown to be a promising catalyst for ammonia synthesis.1 For the rational development of Ca3CrN3H, or related materials, towards such an application, a fundamental understanding of its hydride-ion mobility is crucial, but such an understanding is at present missing.
In this work, we report on the nature of hydride-ion mobility Ca3CrN3H, using quasielastic neutron scattering (QENS). The measurements reveal hydride-ion dynamics on the timescale of ≈ 10 picoseconds. Interestingly, the timescale of the dynamics seems almost temperature independent, and the dynamics are observed all the way down to 100 K. Our more detailed analysis shows that the dynamics are likely to reflect localized motions of the hydride-ions, possibly related to hydride-ion jumps along vacant hydride-ion sites in the channel-like structure of Ca3CrN3H.

1Cao, Y. et al. Topochemical Synthesis of Ca3CrN3H Involving a Rotational Structural

Friday 15 December 2023

Unveiling the Collective Dynamics of Liquids and Glass-Forming systems at the Mesoscale by Neutron Spectroscopy with Polarization Analysis and MD-Simulations

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Session: Soft Matter and Liquids

The collective dynamics of liquids and glass-forming systems at the ‘intermediate scales’ (or mesoscales) is of paramount importance to understand, for instance, the role of specific interactions –as hydrogen bonds— and the formation of dynamic aggregates of supramolecular nature. However, due to inherent difficulties, the mesoscopic range is still a rather ‘white area’ in the relaxation map of these systems. In principle, quasi-elastic neutron scattering (QENS) is the ideal technique with which to directly observe the collective relaxation of liquids and glass-forming systems in a wide Q-range (Q: momentum transfer) due to the high energy resolution currently available. The problem is that the measured intensity always contains a combination of coherent and incoherent contributions, which can be very relevant at the mesoscopic range. The only way to clearly separate coherent and incoherent contributions is to apply polarization analysis techniques, which, for spectroscopy, still are rather scarce. By means of the recently implemented neutron polarization analysis on a wide time-of-flight spectrometer (LET @ ISIS), we have recently measured coherent and incoherent dynamic structure factor of heavy water with sub-meV resolution in a wide Q-range [A. Arbe et al., Phys. Rev. Research **2**, 022015(R) (2020)]. The results obtained were interpreted with the help of massive MD-simulations [F. Alvarez et al., J. Chem. Phys. **155**, 244509 (2021)]. This work has convincingly proved the power of neutron spectroscopy with polarization analysis that can hugely impact the progress of microscopic dynamic investigations in different fields. We are extending this kind of studies to other liquids as, for instance tetrahydrofuran [A. Arbe et al. **158**, 184502 (2023)]. We are also exploring the possibilities of using polarization analysis methods based on wide angle spin echo techniques (WASP). In this talk I will give an overview of the current status of these investigations.

Phonon spectroscopy using X-rays and a nuclear resonance

D. Bessas and A. I. Chumakov
ESRF – The European Synchrotron, Grenoble, France

Session: Instrumentation

In this contribution I am going to present some features of nuclear inelastic scattering of synchrotron radiation for carrying out vibrational spectroscopy in condensed matter systems.

Nuclear inelastic scattering of synchrotron radiation is particularly useful when is of interest:
- the *true* (in absolute units) partial or total density of vibrational states,
- the element specific inter-atomic mean force constant,
- the vibrational entropy,
- the speed of sound,

or when:
- sub-meV energy resolution (down to 50-100 μeV) for any energy transfer along the inelastic spectrum is required,
- simultaneous measurements of different samples are foreseen.

The conceptual background and the experimental setup of nuclear inelastic scattering will be explained, state of the art results relevant to spectrographic measurements will be presented, complementarity to other high-resolution spectroscopies will be given, and hints on future studies and collaborations will be initiated.

Unusual Dynamics of Tetrahedral Liquids Caused by the Competition between Dynamic and Chemical Heterogeneity

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Session: Soft Matter and Liquids

Tetrahedral liquids are intriguing: they don’t pack the entire space, and they form networks of a variety of structures. As a result, tetrahedral liquids often exhibit fascinating phase behaviors like water. In this talk, I will discuss our recent neutron spin echo measurements of the collective dynamics as well as molecular dynamics (MD) simulations, using a neural network forcefield (NNFF), of another prototypical AX_2-type tetrahedral network liquid, $ZnCl_2$. We observed an unusual non-monotonic temperature-dependence of the stretching exponent β as the liquid is supercooled. Further simulations revealed that this unusual dynamic behavior is due to the competition between dynamic and chemical heterogeneity. This discovery may provide new insight into the unusual thermodynamic properties of tetrahedral liquids.

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Q-dependent collective relaxation dynamics of glass-forming liquid $Ca_{0.4}K_{0.6}(NO_3)_{1.4}$

Peng Luo[1], Yanqin Zhai[1,2], Peter Falus[3], Victoria García Sakai[4], Monika Hartl[5], Maiko Kofu[6], Kenji Nakajima[6], **Antonio Faraone**[7], Y Z[1,2,8]

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Session: Soft Matter and Liquids

The relaxation behavior of glass formers exhibits spatial heterogeneity and dramatically changes upon cooling towards the glass transition. However, the underlying mechanisms of the dynamics at different microscopic length scales are not fully understood. Employing the recently developed wide-angle neutron spin-echo WASP (ILL, France) in conjunction with the Time-of-Flight spectrometer Amateras (MLF, J-PARC, Japan), we measured the Q-dependent coherent intermediate scattering function of a prototypical ionic glass former $Ca_{0.4}K_{0.6}(NO_3)_{1.4}$, in the highly viscous liquid state. In contrast to the structure modulated dynamics for $Q < 2.4$ Å^{-1}, i.e., at and below the structure factor main peak, for $Q > 2.4$ Å^{-1}, beyond the first minimum above the structure factor main peak, the stretching exponent exhibits no temperature dependence and concomitantly the relaxation time shows smaller deviations from Arrhenius behavior. This finding indicates a change in the dominant relaxation mechanisms around a characteristic length of $2\pi/(2.4$ Å$^{-1}) \approx 2.6$ Å, below which the relaxation process exhibits a temperature independent distribution and more Arrhenius like behavior.[1]

P. Luo, et al., *Nature Comm.*, **13**, 2092 (2022).

The dynamics of ionic liquids in mild confinement

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Session: Soft Matter and Liquids

The structure and dynamics of the ionic liquid 1-ethyl-3-methylimidazolium acetate was studied using x-ray diffraction, neutron backscattering and neutron spin echo spectroscopy. The bulk liquid was compared to the confined liquid by nanopores of porous glasses. In the bulk, the liquid did not display long-range order while in the pores onion-shaped lamellar ordering was observed. This also had impact on the dynamics: The ions could propagate along the random domains in the bulk sample, while they needed to overcome a barrier of the lamellar arrangement. The second topic was the addition of small amounts of water, studied under different contrasts with neutron spin echo spectroscopy (WASP). Here, we discovered a strong binding of the water to the acetate ion.

Exploring the Impact of Composition and Cross-Linking Degree on Single-Chain Nanoparticles Derived from Poly(tetrahydrofuran-co-epichlorohydrin) copolymers: An Investigation using Quasielastic Neutron Scattering, Calorimetric Analysis, and Broadband Dielectric Spectroscopy

Jon Maiz[1,2], Ester Verde-Sesto[1,2], Isabel Asenjo-Sanz[1], Fanny Juranyi[3], José A. Pomposo[1,2,4], Arantxa Arbe[1] and Juan Colmenero[1,4,5]

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Session: Soft Matter and Liquids

In recent years, the industrial sector has witnessed significant advancements in multiphasic polymer materials or complex systems. These include polymer blends, polymer composites (including nanocomposites) and different types of copolymers [1].

Over the past few years, our research group has been actively involved in a project centered around poly(tetrahydrofuran-*co*-epichlorohydrin) P(THF-*co*-ECH) based single chain nanoparticles (SCNPs). Briefly, SCNPs are nano-sized structures formed by crosslinking functionalized linear polymers, referred to as "precursors" [2]. This area of study remains complex and multifaceted, leaving many questions unanswered, particularly regarding how the type and extent of internal crosslinking influence the system's structure and dynamics.

Up to now, our specific focus has been on functionalizing the ECH comonomer, and the studies have primarily concentrated on a molar ECH content of 27% in the precursor sample. At this composition, the precursor chains do not exhibit crystallization. As a showcase of complex material based on these macromolecules, we have investigated an all-polymer nanocomposite consisting of SCNPs immersed in a matrix of linear chains of their precursors. Our study demonstrates the power of QENS combined with isotopic labeling, allowing us to discern details on the dynamics that are not well resolvable by other techniques [3].

In this study, we present a comprehensive investigation utilizing quasielastic neutron scattering (QENS), DSC and broadband dielectric spectroscopy (BDS) techniques on poly(THF-*co*-ECH) copolymers at two different compositions and different crosslinking degrees. Our aim is to explore how the copolymer composition can be manipulated to control their ability to crystallize and understand the influence of crystallizability on their dynamic properties. Furthermore, our objective is to investigate the effects of the internal degree of crosslinking within a bulk of SCNPs and assess how these internal crosslinks influence the overall structure and, more importantly, the dynamics for each composition.

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QENS study of citrate dynamics on the surface of iron oxide nanoparticles

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Session: Soft Matter and Liquids

Surface functionalization is needed for the synthesis and the control of properties of iron oxide nanoparticles (IONPs) in various applications for biomedicine, ferrofluids, or heterogeneous catalysis[1-2]. Yet, experimental investigations of interfacial properties such as the dynamics of ligand and water molecules near the nanoparticle surface have been scarce. Previously, quasielastic neutron scattering (QENS) was successfully used to access the dynamics of water molecules on surfaces of TiO_2 and SnO_2 NPs[3]. QENS studies of ligand dynamics on the surface of NPs have also emerged recently. Dodecanethiol on PbS nanoparticles and oleates on IONPs were shown to exhibit rotational motion[4-5]. However, the dynamics of citrate ligands on the surface of magnetic IONPs are largely unknown. We report on QENS experiments on 6 nm IONPs synthesized according to ref 6 and stabilized at 8 % relative humidity in H_2O (measured at IN16B, ILL) and D_2O (measured at Emu, ANSTO). Elastic and inelastic fixed window scans in the temperature range of 2 – 334 K and covering a Q-range of 0.19 – 1.83 Å^{-1} were used to distinguish contributions from phonons, magnetic relaxations, and ligand motion. We stress the broad understanding which can be gained just from fixed window scans by comparing scaled changes of elastic and quasielastic intensities at different Q values in- and outside of magnetic Bragg peaks. Further, the Q-dependence of energy-resolved spectra shows, that citrate molecules only exhibit a rotational motion on the IONP surface.

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HiRES 2023
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Posters

High resolution spectroscopy on IN16B

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Session: Instrumentation

Neutron spectroscopy gives unique insight into microscopic dynamics and excitations in matter. Crystal spectrometers such as IN16B at the Institut Laue-Langevin in Grenoble (France) operating in backscattering geometry provide high energy resolution down to sub-micro-eV.

IN16B serves its international user community for nearly a decade by now. We will highlight some examples of the various experimental possibilities on IN16B, such as the high resolution Doppler monochromator mode including elastic/inelastic fixed energy window scans, the extended transfer range of the ToF mode 'BATS', and the flexibility to accommodate standard and non-standard sample environments including the option for in-situ dielectric spectroscopy.

The science fields covered by the instrument so far encompass e.g. dynamics in biological and soft matter systems, magnetic excitations, energy materials, glass forming liquids and complex liquids both in bulk and in confinement, as well as diffusion in the solid state. We are looking forward to discuss further needs, opportunities and possible synergies of different high-resolution techniques with the user community.

Effects of Polydispersity on Diffusion

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Session: Soft Matter and Liquids/Biology

The diffusive properties of colloid suspension depend on several parameters such as temperature [1], additives [2,3] or crowding [4]. However, in biological environments, the colloids are exposed to a polydisperse environment. To investigate the effect of polydispersity on the diffusive processes, we have performed different studies. Applying both, advanced modelling as well as partial deuteration, we have been able to investigate diffusive properties of the tracer particles in different polydisperse environments. While the monodisperse volume fraction dependence can often be described with colloid physics, the description breaks down for polydisperse environments.

In the systems investigated, the particles smaller than the averaged particle radius are additionally accelerated compared to the diffusion of the same particles in a monodisperse solution at the same overall volume fraction. Larger particles are slowed down in respect to monodisperse case.

We observed this general finding in a bidisperse solution, were we able to separate simultaeously the diffusion coefficients of the differently sized tracer particles [5] as well as in several studies, where a polydisperse deuterated environment, mimiking a cellular environment, was used. Here, we investigated the influence at different volume fractions with one fixed tracer size [6] as well as the dependence on the tracer size [7].

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The onset of molecule-spanning dynamics in heat shock protein Hsp90

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Session: Biology

Protein dynamics have been investigated on a wide range of time scales. Picosecond dynamics and millisecond-dynamics have been assigned to local fluctuations and large conformational changes, respectively. For the multi-domain protein heat shock protein 90 (Hsp90), we uncover dynamic modes on the 100 ns to 200 ns time scale which we attribute to molecule-spanning real-space movements . A comprehensive understanding of these molecule-spanning dynamics, became possible by combining nanosecond fluorescence correlation spectroscopy (ns-FCS), time-resolved anisotropy (TRA), neutron spin echo spectroscopy (NSE), neutron backscattering spectroscopy (NBS), small angle neutron scattering (SANS) and molecular dynamic (MD) simulations [1].
Interestingly, we find that the 100 ns-dynamics are affected by the co-chaperone Sba1 and that the time scale depends on the conformational state of Hsp90. Altogether, our data is best described by fast molecule-spanning dynamics, which likely precede larger conformational rearrangements in Hsp90 and thus could provide the molecular basis for allostery.

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New Analysis Frameworks for FWS-QENS of Colloid Suspensions

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Session: Instrumentation

High-resolution neutron backscattering spectroscopy offers access to diffusive properties of colloid suspensions such as the center of mass diffusion coefficients as well as to internal diffusive properties. The backscattering spectrometer IN16b offers the possibility of acquiring quasi-elastic neutron spectra (QENS) at specified energy transfers, so called fixed Window Scans (FWS). By reducing the amount of energy transfers investigated, the acquisition time can be reduced by two orders of magnitudes. Based on systematic measurements on different colloid suspensions, we have established a framework to extract from these FWS the apparent global diffusion of the colloids with quantitative agreement compared to the analysis of the QENS with quasi-continuous energy axes [1].

By acquiring additional energy transfers, contributions of different particles in solution, such as diffusing and aggregated colloids, can be separated.

Given the short acquisition time for concentrated solutions, FWS allow to investigate time dependent processes with a minute resolution and also to investigate lower concentrated solutions.
FWS-QENS can therefore now be applied to samples which were not suitable before for neutron backscattering due to long acquisition times. This includes low concentrated samples as well as samples changing with time [2] or as a function of a control parameter [3].

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Comparison of time-of-flight with MIEZE spectroscopy of H2O: Necessity to go beyond the spin-echo approximation

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We report a comparison of high-resolution neutron spectroscopy of pure water at 300 K as performed at the ToF spectrometer TOFTOF (FRM II) with MIEZE spin-echo spectroscopy as performed at RESEDA (FRM II). We introduce a theoretical framework beyond the spin-echo approximation to transform arbitrary $\frac{\mathrm{d}\sigma 2}{\mathrm{d}\Omega \mathrm{d}E\mathrm{f}}$ from the energy to the time domain. Using this framework, we analyze the inelastic neutron scattering data in two ways: (1) The ToF spectra were transformed directly into the time domain. (2) Describing S(2$S(2\vartheta, E)$ in terms of several Lorentzian distributions, both data sets were described in terms of an individual and a combined function, taking the instrumental resolution into account.

Within both approaches good agreement is achieved, where discrepancies suggest the need for an improved understanding of instrumental resolution, neutron detection efficiency over a broad range of energy transfers, and background scattering. As instrument performance proves to be key for a rigorous comparison of various neutron spectroscopic techniques, advanced computational modelling of real systems, such as molecular dynamics simulation, in conjunction with instrument modelling, such as Monte-Carlo simulations of neutron propagation, promise to disentangle instrument peculiarities from sample-specific interactions.

Phonon spectroscopy using X-rays and a nuclear resonance

D. Bessas, A. I. Chumakov

ESRF – The European Synchrotron, Grenoble, France

Session: Instrumentation

In this contribution I am going to present some features of nuclear inelastic scattering of synchrotron radiation for carrying out vibrational spectroscopy in condensed matter systems.

Nuclear inelastic scattering of synchrotron radiation is particularly useful when is of interest:
- the *true* (in absolute units) partial or total density of vibrational states,
- the element specific inter-atomic mean force constant,
- the vibrational entropy,
- the speed of sound,

or when:
- sub-meV energy resolution (down to 50-100 μeV) for any energy transfer along the inelastic spectrum is required,
- simultaneous measurements of different samples are foreseen.

The conceptual background and the experimental setup of nuclear inelastic scattering will be explained, state of the art results relevant to spectrographic measurements will be presented, complementarity to other high-resolution spectroscopies will be given, and hints on future studies and collaborations will be initiated.

New ZETA+ on IN22

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Session: Instrumentation

The first neutron-resonance spin echo option on IN22 (ZETA) was commissioned 15 years ago. The objectives of this option were, on the one hand, to measure the lifetime of dispersive excitations, and on the other hand, to characterize the crystallographic quality of a crystal (its mosaisity, but also the distribution of its lattice constants) and to track its volumetric thermal expansion as a function of temperature. In both cases, the aim was to improve the resolution by 2 orders of magnitude with respect to conventional methods. ZETA@IN22 gave its first promising results in the 2010s.

However, this option was rather complicated to use and set-up. Moreover, the signal was degraded by RF amplifiers which were not class A, leading to a sub-optimal efficiency of the resonant spin flippers (RSFs). Finally, even if it was built with independent coupled RSFs, the minimum accessible `Fourier' time was around 10 ps thereby lowering the effective accuracy of the measurements.

In 2016, after the failure of several RF coils, an upgrade was decided, including the procurement of fully class A amplifiers. Taking advantage of the independent RSFs, we proposed a solution to create a virtual zero field integral, giving a Fourier time ≈ 0 ps. This possibility increases largely the capabilities of ZETA and hence, the precision of the measurements. The first tests performed at the end of 2021 and this year show that we are able to reach a zero Fourier time and to obtain an accuracy on the distribution of lattice constants of a standard crystal lower than 10-5. Taken together, these improvements allow us offering for the end of this year a user-friendly upgrade of the ZETA option, that will allow measuring reliably very small lattice transition as well lifetime of different kind of excitations.

Dynamics of subdomains in polyelectrolyte complexes

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Session: Soft Matter and Liquids

The formation of interpolyelectrolyte complexes (IPECs)[1] by mixing oppositely charged polyelectrolytes has been studied extensively over the last years. Depending on the mixing ratio and the solvent properties, they may form soluble complexes or biphasic systems with either a solid precipitate or a second liquid phase (coacervate).

Soluble IPECs are interesting as they are colloidal particles that can, for instance, be applied as carrier systems for active agents, e.g. in drug delivery [2], where the polyelectrolytes can be hydrophobically modified and so they can form hydrophobic domains. These domains allow incorporating hydrophobic molecules that are otherwise not soluble in water. Despite the investigations that have been performed so far, a detailed picture of the stability of such hydrophobic pockets is still missing. This competition between hydrophobic and hydrophilic domains has been also observed in polyelectrolyte surfactant complexes (PESCs) [3]: surfactants are entropically favoured to form hydrophobic micelles exposing the charged head, which interacts with the PEs, forming similar structure to IPECs.

An experimental investigation conducted at IN15 is aiming to offer a clear description of the stability of the hydrophobic domains inside IPECs: mixing hydrophobically modified quaternized Poly(2-(dimethylamino)ethyl methacrylate) PDMAEMA as polycation and a double hydrophilic block-copolymer given by polyethylene glycol as nonionic tail and polymethacrylic acid as polyanion block, IPECs formation is induced and we can characterize the dynamics of the subdomains inside the IPECs via neutron spin-echo (NSE).

In this talk we will discuss the results obtained from NSE, along with SANS and molecular dynamics simulations.

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Are Proteins Dynamically Heterogeneous?

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Session: Soft Matter and Liquids/Biology

We address the question, whether proteins as ‘complex systems’ can be characterized dynamically by specific spatial motions, instead of resorting to heterogeneous diffusion in energy space (1,2). The neutron scattering spectra of dry and hydrated proteins are investigated in the time domain based on a generic dynamic model with two major components, rotational transitions of side chains, mainly methyl groups and local translational diffusion of non-methyl side chains (3). The significance of the fits is based on data covering a wide range in time, momentum exchange and temperature. The translation-rotation model applies the established theory of space time correlation functions successfully, which is compatible with the idea of homogeneous quasi-elastic spectra (4). Dynamical heterogeneity on a small scale can be deduced from the rate distributions of the motional components. The temperature dependent elastic intensity is shown to support the dynamic analysis. The puzzling change of the intensity at zero momentum exchange is easily explained by multiple scattering effects, without resorting to energy landscape models (1,5).

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α-relaxation dynamics of thin films of glass-forming liquid crystal

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Session: Soft Matter and Liquids

Glasses are formed by rapidly cooling substances, which is accompanied by a slowing down (or freezing) of the stochastic movements around the glass transition temperature, T_g. Due to their universal properties, the glasses have a variety of applications, e.g. in amorphous pharmaceuticals, material engineering and the production of optical fibers. In applications where performance is essential, the physical aging of the glasses can be a problem, resulting in structural and property changes. Glasses produced by Organic Molecular Beam Deposition (OMBD) below the glass transition temperature ($0.85T_g$) can be a solution. OMBD is a technique that allows the formation of organic thin films in ultra-high vacuum by evaporating the material whose the molecules impinge on a cold substrate, creating layers. By carefully controlling the deposition rate and substrate temperatures, the stable glasses can be created. However, the OMBD method has so far not been used for liquid crystals, but for other organic compounds, such as glycerol.

In this work, we have verified the possibility to create thin films of liquid crystal below the glass transition temperature by the OMBD method. We have also investigated the effect of OMBD measurement parameters on the layer thickness and the α-relaxation dynamics of compound under study. Thin films represent an alternative geometry to investigate molecular dynamics. Some striking differences in relaxation dynamics related with glassy state of thin films compared to the sample in the bulk form have been observed.

Influence of Amyloid β42 Peptide and NSAID’s on the Dynamics of Brain Phospholipid Unilamellar Vesicles

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Session: Soft Matter and Liquids

Amyloid β42 (Aβ42) peptide is predominantly found in the extracellular spaces of the brain cells in the form of plaques. Aβ42 peptide is mainly responsible for the neurodegenerative disease Alzheimer's disease. Aβ42 peptide depending upon aggregation states monomer (M)/oligomer (O)/proto-fibril (pf)/fibrils (F), and amino acid length can strongly interact with the model membrane mimetic systems. Furthermore, nonsteroidal anti-inflammatory drugs (NSAID’s) are most widely used drugs for their anti-inflammatory, analgesic, antiplatelet, and antipyretic characteristics. The primary target of NSAID’s is the inhibition of cyclooxygenase (COX) activity. Studies have shown that alternate mechanism of action of NSAID’s through plasma membrane interaction. NSAID's has great impact on the human health and Aβ42 peptide, a principle cause of cognitive dysfunctionality, has vital role in the pathogenesis of Alzheimer disease. Both NSAID’s and Aβ42 have propensity to alter the structure and the dynamics of the membrane mimetic systems. Therefore, investigating the influence of NSAID’s and different aggregation states of Aβ42 peptide on the dynamics of physiologically relevant membrane system will add to our understanding of NSAID’s membrane interaction and the Aβ42 peptide induced perturbation in the membrane mimetic system and Alzheimer pathogenesis, respectively. We have prepared unilamellar vesicles, a model membrane system, from phospholipids extracted from porcine brain tissues. These unilamellar vesicles, diameter of ~100nm, are mainly composed of phosphatidylcholine (PC), phosphatidylethanolamine (PE) phospholipids, sphingomyelin (SM), free fatty acids (FFA) and hexosylcermides (HexCER). We have investigated the influence of NSAID’s, aspirin, ibuprofen, and diclofenac and Aβ42 – monomer and proto-fibrils, individually, on the dynamics of the brain phospholipids unilamellar vesicles using quasielasitc neutron scattering (QENS) IN5, a disk chopper time of flight (TOF), and neutron spin echo (NSE) IN15 spectrometers suitable to probe the motions on the picosecond and nanosecond time scale, respectively.

Quantification of the hydration of mucins

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Session: Soft Matter and Liquids/Biology

Mucins represent an important class of glycoprotein with a distinct structural motif - a peptide linked backbone is surrounded by a bottle brush of charged sugar residues. These materials function as soft wet barriers in vertebrate physiology in a range of solvent conditions: pH, salt and water content. Hydration of these residues, and the subsequent excluded volume interactions along the peptide linked backbone are thought to modulate the flexibility of the chain. We have observed and quantified the number of water molecules strongly bound to the glycosylations in simple mixtures of water and mucins with a combination of QENS measurements and THz frequency dielectric spectroscopy. Our eventual aim is understand the role of solvent conditions on the flexibility and the extensional rheology of mucin solutions.

Anomalous Protein Diffusion and Solvent-mediated interactions in Crowded Solutions with Coherent X-Ray Scattering Using XFELs

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Session: Biology

Proteins play essential roles in life, for instance serving as carriers, participants in the immune response or for their structural role. In vivo, they exist within crowded environments with protein volume fractions typically ranging as high as 30%. When the environment becomes highly concentrated, the dynamics of proteins deviate significantly from those observed in a dilute system. However, the precise mechanisms influencing these dynamics across different time scales are not yet fully understood. Here we present our recent results [1], where we investigated the effect of self-crowding on protein diffusion in a ferritin solution with varying concentrations using X-ray Photon Correlation Spectroscopy. This technique allows simultaneous monitoring of both the structure, through small angle scattering, and the diffusion, through intensity-autocorrelation functions, of the protein solution, as demonstrated in our previous study [2]. By analyzing the scattering intensity, we observed that the ferritin particles become more densely packed with increasing protein concentration, indicated by a pronounced peak in the structure factor that shifts towards lower momentum transfer values. The protein diffusion, measured at all concentrations, follows a Brownian type of motion, but exhibits deviations from the expected quadratic dependence of the decorrelation time at the peak position. This deviation can be attributed to the crowding effect caused by neighboring proteins, which act through hydrodynamic interactions. The hydrodynamic functions, which reflects these interactions, exhibit a peak which coincides with that of the structure factor indicating the connection of the crowding and the hydrodynamic interactions. To elucidate the underlying mechanism, we compare the hydrodynamic functions with estimations based on the δγ-theory, which considers the non-trivial interactions between particles. The model indicates that the protein diffusion is slower than that of non-interacting hard spheres due to the presence of solvent-mediated interactions and effective local friction between the particles.

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Investigating Innovative All-Polymer Nanocomposites: Blending Ferroelectric PVDF with Single-Chain PMMA-Based Nanoparticles

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Session: Soft Matter and Liquids

Due to their inherent bistable nature and the ability to be repeatedly switched by an electric field, ferroelectric polymers are highly valuable [1]. Poly(vinylidene fluoride) (PVDF) and its copolymers are widely recognized for their exceptional polarization response when subjected to an electric field [2,3]. PVDF exhibits four distinct crystalline phases, namely the β-, γ-, and δ-phases, which are ferroelectric, while the α-phase is paraelectric. Pure PVDF films typically crystallize in the non-ferroelectric α-phase. However, studies have indicated that PVDF can predominantly crystallize in the ferroelectric β-phase when blended with the miscible poly(methyl methacrylate) (PMMA) [4,5].

Although previous research has explored the ferroelectric phase diagram of the PVDF/PMMA system [4], limited information is available regarding the connections between processing, structure, dynamics, and functionality in the amorphous portions of such nanocomposites. Additionally, the impact of the PMMA component's topology on the PVDF crystal structure and the dynamics of the remaining amorphous fraction remains unclear. We have initiated a project utilizing PMMA-based Single Chain Nanoparticles (SCNPs) as fillers for PVDF. SCNPs are soft polymeric nanostructures formed by controlled folding and intrachain cross-linking of individual polymer chains [6].

Our objective is to examine the impact of PMMA-based SCNPs on the behavior of PVDF in nanocomposites at various compositions and analyze the properties of the resultant materials. We will include control samples comprising PVDF blended with linear precursor chains. To investigate these materials, we will employ techniques including Broadband Dielectric Spectroscopy (BDS), Differential Scanning Calorimetry (DSC), and Small- and Wide- Angle X-Ray Scattering (SAXS and WAXS). Quasielastic Neutron Scattering (QENS) data have H RES 2023 Synergies in HIgh RESolution Spectroscopy been collected; however, the analysis is still in progress. Our primary goal is to integrate and evaluate the complementarity of these techniques, with a specific focus on BDS, in order to conduct a comprehensive study of the dynamics in these ferroelectric materials.

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Dynamic structure factor of undulating vesicles: Finite-size, spherical geometry, and membrane viscosity effects with application to neutron spin echo experiments

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Session: Soft Matter and Liquids

We consider the dynamic structure factor of quasi-spherical vesicles, and present a generalization of an expression that was originally formulated by Zilman and Granek (ZG) for scattering from isotopically oriented quasi-flat membrane plaquettes [1,2]. The expression is obtained in the form of a multi-dimensional integral over the undulating membrane surface. It reduces to the original ZG formula in the limit of large wavenumbers q, provided that the vesicles are sufficiently large – in the micron range or larger – known as giant unilamellar vesicles. For much smaller unilamellar vesicles, deviations from the original ZG expression are noticeable even if one assumes that the Seifert-Langer (SL) monolayer density mode is completely relaxed and membrane viscosity is neglected. To avoid the need for an exhaustive numerical integration while fitting to neutron spin echo (NSE) data, we provide a useful approximation that tests well against numerical integration of the complete formula. Applying the approximation to NSE data sets recently obtained at ILL demonstrates an advantage of the present approach over other approximations that have been developed and deployed over the years to fit the NSE data.

We also consider theoretically the effect of membrane viscosity on the undulation relaxation frequencies, and moreover generalize the SL model for quasi-spherical vesicles [3]. Membrane viscosity is shown to be important for the small vesicle sizes studied by NSE and affects in general the longer wavelength undulation modes. We show that for typical parameter values the SL density mode may be slowly relaxing during the NSE time window, which may further complicate the NSE data interpretation.

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Towards an improved understanding of sustainable food emulsions via scattering and neutron spectroscopy

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Session: Soft Matter and Liquids

The stability of food emulsions depends -beside other effects- on a complex interplay between proteins, phospholipids, oil and water. Preparing milk-based and sustainable plant-based emulsions requires good knowledge in interfacial and emulsion stabilization mechanisms, affected by the emulsion composition. To understand these mechanisms in detail different length scales from interatomic to macroscopic distances need to be investigated.

Neutron scattering techniques provide insight into such emulsions on these length scales depending on the technique used. Combining structural information on molecular length scales from small angle x-ray and neutron scattering (SAXS and SANS) with time dependent neutron spin echo spectroscopy (NSE) allows to expand our understanding towards intermolecular interactions within the interface. These interactions are linked to the emulsion stability – the elastic properties of the protein or protein/phospholipid stabilized oil/water interface on molecular length scales. NSE provides in this combination the time dependent correlation function in reciprocal space, S(q,t), on molecular length scales and time scales in the nanosecond range relevant for thermally driven motion of mesoscopic systems such as the emulsion interfaces.

This presentation introduces the neutron and x-ray scattering techniques which broadens the classical characterization of food emulsions. Results from emulsions stabilized with β-lactoglobulin as a representative milk protein, and different plant-based proteins, are presented and discussed. Contrast variation by deuteration of some components of the emulsions is applied to focus on the interfacial region, relying on the uniqueness of neutrons.

Connecting these emerging results with classical characterizations such as interfacial tension or viscoelasticity helps understanding the complex mechanisms of interfacial stability and may contribute to a knowledge driven development of sustainable food emulsions.

Complexes of Oppositely Charged Microemulsions and Polyelectrolytes are Highly Dynamic

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Session: Soft Matter and Liquids

Both Microemulsions and Polyelectrolytes are part of formulations in detergency and personal care products. Therefore, it is rather surprising that only little is known about their interactions. Here, we investigate the interactions of slightly positively charged oil in water droplet microemulsions consisting of the surfactants tetrradecyldimethylamine oxide and tetradecyltrimethylammonium bromide (TDMAO, TTAB 95:5 mol:mol), the cosurfactant 1-hexanol and the oil decance with different anionic polyelectrolytes.

Small angle neutron scattering (SANS) measurements show the formation of elongated aggregates. Measurements of the dynamics on different time and length scales through neutron spin-echo spectroscopy (NSE, up to hundreds of nanoseconds and tenths of nanometres) and dynamic light scattering (DLS, milliseconds and hundreds of nanometres) show qualitatively different behaviour. While NSE measurements show a bimodal relaxation with a fast mode corresponding to the diffusion of free ME droplets and a slower mode corresponding to the diffusion of the aggregates, DLS measurements show a monomodal decay with a diffusion coefficient between the two values obtained from NSE. This finding suggests that the aggregates observed by SANS are highly transient, with a life time between the nanoseconds time scale of NSE and the milliseconds timescale of DLS [1].

Simplistic random walk simulations show that the observed SANS patterns are compatible with free microemulsion droplets which are slowed down by a fixed factor in the immediate vicinity of the PE chain. This means that the formation of aggregates can be observed in SANS without any attractive interactions between the microemulsion droplets.

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RESEDA – a spin-echo spectrometer for magnetism

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Session: Instrumentation

The MIEZE (Modulation of Intensity with Zero Effort) technique is a high-resolution spin-echo time-of-flight technique, for which all spin manipulations are carried out upstream of the sample, in contrast to classical neutron spin-echo spectroscopy. Perhaps most intriguingly, this technique is robust against depolarizing conditions at the sample position.
Therefore, magnetic, or strongly incoherently scattering samples can easily be measured without loss of signal.
The spectrometer RESEDA is being further optimized for measurements in a small angle geometry. We present the current status of the newly installed superconducting solenoids as part of the RF flippers to significantly extend the dynamic range as well as the development and installation of a new detector on a translation stage within a new larger SANS-type vacuum vessel for flexibility with angular coverage and resolution.

Nanoscale dynamics in chain asymmetric lipid membranes

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Session: Soft Matter and Liquids or Biology

While most lipids found in cell membranes have hydrophobic tails that are similar in length, an often-overlooked class of lipids are those that have two tails with different numbers of carbons. Sphingolipids that are enriched in the central nervous system and play an important role in cell recognition have asymmetric tails that can vary in length from 2 to 28 carbons, and some yeast strains produce saturated chain asymmetric lipids in abundance. What is even more fascinating is recent lipidomic studies have shown that yeast cells compensate for a lack of unsaturated fatty acids by synthesizing chain asymmetric lipids. There are very few studies of model chain asymmetric lipid membranes in literature, but the limited data have led to the speculation that chain length asymmetry is an important regulator of membrane fluidity. However, the phrase membrane fluidity is a general term for membrane dynamics that belies the complexity of dynamic processes that span several orders of magnitude in length and time scales from motions of the individual lipid tails to larger scale membrane remodeling. Here we use a combination of neutron scattering methods to probe the nanoscale dynamics in model chain-asymmetric lipid bilayers. We find that despite the partial interdigitation that is known to persist in the fluid phase of these membranes, the collective fluctuations are enhanced on timescales of tens of nanoseconds, and the chain-asymmetric lipid bilayers are softer than an analogous chain-symmetric lipid with the same average number of carbons in the acyl tails. These studies add to growing evidence that the partial interdigitation in chain-asymmetric lipid membranes in the fluid phase is highly dynamic and influences membrane dynamic processes from the molecular to mesoscopic length scales without significantly changing the bilayer thickness or area per lipid.

BIFROST - An indirect geometry cold neutron spectrometer at the European Spallation Source

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Session: Instrumentation

Inelastic neutron scattering has been instrumental in making key discoveries in a broad range of materials, such as superconductors, multiferroic materials and quantum magnets.
However, the applicability of the technique is limited by the inherently weak signals. Since mapping of significant portions of (Q, ω)-space can take days, parametric studies as a function of temperature, pressure and magnetic field is essentially impossible. The triple axis community has improved the efficiency of triple-axis spectrometers by constructing multiplexing analyser arrays covering large intervals of scattering angles in the horizontal plane. Exemplified by CAMEA (PSI) and MACS (NIST).

BIFROST, currently under construction at ESS, employs a CAMEA-like multiplexing backend on an indirect geometry time-of-flight (ToF) front end. The primary spectrometer enables an unprecedented polychromatic sample flux exceeding 1010 n/s/cm2 at 5 MW accelerator power, with a bandwidth of 1.7 Å, whilst retaining a primary spectrometer resolution ΔEi/Ei of 4 %, common in cold neutron spectroscopy. The multiplexing backend consists of 9 Q-channels, each containing 5 fixed analyzers probing a scattered neutron energy range of 2.7 to 5.0 meV. The analyzers utilize the graphite crystal mosaicity combined with position sensitive neutron detectors to gain continuous energy sensitivity, resulting in a back-end resolution considerably better than on a classical TAS setup.

We here present extensive McStas simulations of BIFROST and its energy resolution in different operation modes. We show that it is possible to obtain excellent energy resolution even in deep inelastic scattering where we at 10 meV energy transfer find a resolution of approx. 30 μeV.

Gradual melting of magnetic stripes in superconducting and non-superconducting cuprates

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Session: Magnetism

In cuprate superconductivity (SC), there is a subtle connection between SC and antiferromagnetism. The magnetism in cuprate $La_{2-x}Sr_xCuO_4$ (LSCO), and similar cuprate SC, is found as static or dynamic stripes, i.e., small patches of antiferromagnetism, separated by 'rivers' of charge, giving rise to magnetic signals at incommensurate positions in Q. However, it remains important to clarify the details of these stripes to pinpoint their relation to SC.

Dynamic stripes are present at all temperatures, while static stripes appear only below the SC critical temperature, $T_N = T_C$, and only for low-doped samples $0.05 < x < 0.13$, measured by cold neutron triple axis spectrometers with a resolution of 150 μeV. However, μSR measurements curiously find a much lower ordering temperature, $T_N < T_C$. While this discrepancy has been attributed to the different timescales between the methods, this was never proven directly.

Here we present a direct comparison between data from LSCO samples with $x = 0.05$ and $x = 0.07$, measured on backscattering spectrometers DNA (JPARC) with a 15 μeV resolution, IN16 (ILL) with a 1 μeV resolution and the μSR instrument GPS (PSI) with corresponding resolution of 0.015 μeV. The static stripe signal, seen with the better resolution of backscattering spectrometers, shows a T_N that agrees with the muon data, proving that the timescale hypothesis was correct. In addition, we observe a quasielastic broadening of the stripes at temperatures close to T_N.

Thus, we will for the first time have revealed how static stripes convert over quasielastic to dynamic stripes in a cuprate SC as temperature changes. This elucidates the correlation between stripe pinning and the onset of 2D / 3D SC.

Magnetically frustrated dynamics on the Cairo pentagonal lattice

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Session: Magnetism

The octahedrally and tetrahedrally coordinated Fe3+ (S=5/2) ions in Bi2Fe4O9 form a quasi two-dimensional Cairo pentagonal lattice (Pbam). Combined with predominantly antiferromagnetic interactions, this leads to a strong frustration with Tn=245 K while θCW=1670 K in a fairly unexplored geometry. The magnetic structure for T<Tn can be indexed with k=(½, ½, ½) with a noncollinear magnetic structure of Fe1 and Fe2 moments, 2 different sites, and an interpenetrating pattern of fourfold spin rotations, a very novel magnetic state.

Previous measurements studying the low-lying excitations, 0–30 meV, in the ordered phase were measured on small single crystals (< 0.6 g). Beauvois et al. (PRL2020) argues that no anisotropy is required while Duc Le et al. (PRL2021) indicates an easy-plane single-ion anisotropy. Accordingly, Beauvois find the acoustic AF mode is not gapped while it is for Duc Le.

We have synthesized a 2.35 g, high quality single crystal of Bi2Fe4O9 and are re-examining the magnetic excitations. At low energy transfers we see distinct discrepancies between our data and previous data in the ordered phase using CAMEA (PSI). Our data show a clear double spin gap, that allows us to differentiate axial and planar anisotropy scenarios. We have also taken data at higher energies at EIGER (PSI) and IN20 (ILL). This has enabled us to establish the microscopic model at low temperatures and we will later perform quantitative analysis of the frustrated state above Tn.

Metabolic effect of Pd(II) Trinuclear Chelate with spermidine on TNBC-Sensitive and -Resistant to cisplatin

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Triple-negative breast cancer (TNBC) is the worst prognosis subtype of breast cancer. TNBC accounts for *ca.* 15% of all breast cancers, being more prevalent in premenopausal young women. Due to its aggressiveness and metastatic potential, it has a high risk of recurrence leading to mortality in the first 5 years after diagnosis. The overall survival for women with TNBC is 13.3 months, and less than 30% of the cases survive longer than 5 years. While the average time relapse for non-TNBC is 35–67 months, in TNBC patients it is only 19–40 months [1-3]. The development of novel chemotherapeutic strategies against TNBC is therefore an urgent clinical need, specifically aiming at a higher efficacy regarding cell growth inhibition and decreased angiogenesis and invasiveness, coupled to lower acquired resistance and deleterious side effects.

Raman and Fourier transform infrared (FTIR) vibrational spectroscopies are promising tools in drug development, since they are highly accurate analytical techniques that enable rapid and non-invasive *in vitro* and *in vivo* measurements (without the need for dyes or probes) and deliver unique molecular level signatures of biospecimens with high accuracy, sensitivity and specificity [3-5]. The present work aimed to assess the metabolic impact of a trinuclear palladium complex with biogenic polyamine, Pd_3Spd_2 (Spd, $H_2N(CH_2)_4NH(CH_2)_3NH_2$) on cisplatin-sensitive and cisplatin-resistant TNBC human cells (MDA-MB-231 and MDA-MB-231/R, respectively). Spectral biomarkers of cytotoxicity were identified: for MDA-MB-231 a major impact of Pd_3Spd_2 was found on DNA, cellular proteins and lipids; for MDA-MB-231/R, in turn, the main effect was on proteins.

This improved knowledge of the main cellular targets of this kind of metallodrugs constitutes a paramount advance in cancer therapy, particularly regarding a type of malignancy with such a poor prognosis and limited chemotherapeutic success. In addition, these findings are expected to contribute to the development of new and optimized metal-based agents for specific breast cancer types and subtypes (apart from TNBC).

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Portable devices for adding Spatial-Intensity-Modulation-mode capabilities to polarized neutron beams at FRM-II

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Session: Instrumentation

The Modulated IntEnsity with Zero Effort (MIEZE) resonant spin-echo technique implemented at the RESEDA instrument at the FRM II has its optimum resolution at small scattering angles, i.e. SANS geometries. To extend the application of MIEZE to larger scattering angles, the incorporation of magnetic Wollaston prisms (MWPs) into the MIEZE beamline has been suggested [1]. MWPs can produce controlled spatial modulations of intensity, which in addition to the intensity modulations in time inherent to MIEZE would allow correction to the neutron time of flight differences and extend the MIEZE resolution to any desired scattering angle. Additionally, MWPs will be useful in the context of intra-particle mode-entangled neutron beams for potential use in probing many-body quantum entanglement in materials. Finally, the compact and modular nature of MWPs will allow them to be used to measure diffraction peaks with enhanced resolution at several polarized beam instruments such as KOMPASS, LaDiff, and small-angle neutron scattering instruments. In this contribution, we provide an update on the construction progress of superconducting MWPs intended for use at FRM II, describe the details of their operation, and discuss the various possibilities they offer.

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Diffusion in supramolecular hydrogel for drug delivery

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Session: Soft Matter and Liquids

The use of biologics as therapeutic agents has gained significant popularity due to their advantages in terms of selectivity and reduced side effects. However, their administration is limited to liquid formulations that require frequent injections due to rapid uptake. To address this issue, there is growing interest in using gels as pharmaceutical delivery vehicles, as their higher viscosity compared to liquid formulations allows for delayed drug release.

Supramolecular gels, a subclass of gels formed through non-covalent self-assembly of low molecular weight gelator molecules, offer easier tunability of viscosity and can be triggered by external factors such as temperature or pH changes. This project aims to investigate the relationship between macroscopic drug release from gel-based pharmaceutical formulations, which is influenced by factors like macroscopic viscosity, and microscale diffusion, affected by hydrodynamic and direct interactions. Neutron spectroscopy was employed to analyze the subtle effects occurring at the nanosecond-to-picosecond timescale. Time-of-flight Quasi-Elastic Neutron Scattering (QENS) on the BATS instrument was utilized to examine surface chemistry-dependent drug diffusion, revealing slower or faster dynamics in gels with respect to the bulk solution depending on the specific gel surface chemistry. Additionally, the Wide Angle Spin-Echo spectrometer (WASP) was employed to study small drug molecule dynamics at the nanosecond timescale, accounting for the confinement effect of the gel network on diffusion behavior. Lastly, the lower q range and longer timescale (100 ns) available on IN15 allowed to investigate the fibre network diffusion whose dynamics cannot be resolved by instrument resolution on ToF. The results demonstrate that the network exhibits weak movement at the timescale accessed by Spin-Echo, with varying mobility depending on the specific supramolecular gels.

Multi-scale diffusion in therapeutic monoclonal antibody solutions

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Session: Biology

Monoclonal antibodies (mAbs) are relevant for therapeutic applications due to their specificity and versatility. One of the current pharmaceutical challenges concerns mAb formulation for subcutaneous injection, which is becoming the preferred delivery route due to its improved convenience compared to other parenteral administration methods.

Being volumes <2mL better tolerated in the subcutaneous space, highly concentrated mAb formulations are needed to achieve significant therapeutic effects, potentially leading to high solution viscosities and altered drug injectability. The main challenge is therefore to keep their viscosity below 15-20 mPa·s [1] without altering the solution stability.

Since the understanding of macroscopic viscosity requires an in-depth knowledge on protein multi-scale diffusion, mutual interactions and aggregation [2,3], we employ high-resolution neutron spectroscopy techniques to investigate nine different mAbs of IgG1/IgG4 subtypes in aqueous solution as a function of protein concentration and temperature.

The synergy between quasi-elastic neutron scattering (QENS) and neutron spinecho (NSE) spectroscopy allows us to probe short-time self- and collective diffusion of the different mAbs, observe their different clustering behaviours and access their internal dynamics [4,5,6,7]. QENS data are treated using established analysis frameworks and interpreted using colloid physics models [4,7]. Polyclonal antibody solutions are measured for comparison [8,9].

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Collective lipid acyl tail dynamics and membrane viscosity

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Session: Soft Matter and Liquids or Biology

Lipid molecular motions determine the membrane properties on local and global scales. Here we aim to link the molecular and macroscopic motions by employing neutron spin echo and quasielastic neutron and gamma-ray scattering techniques to measure the collective lipid acyl tail dynamics on length scales of approximately 0.5 nm and timescales ranging from ps to hundreds of ns. There are two relaxation processes in the fluid phase with relaxation times of the order of 10 ps and 100 ps. The fast mode originates from density fluctuations of lipid acyl tails and are about an order of magnitude slower than those of the bulk liquid alkanes with the corresponding number of carbons. This slowdown is attributed to the effects of twodimensional confinement and associated restrictions of the molecular orientations. The slower relaxations are related to the diffusive motions of the lipid molecules. Meanwhile, in the gel phase, the lipid acyl tail dynamics are quite heterogeneous, and the relaxation processes span 4 to 5 orders of magnitude in time. The average relaxation time changes from 10s ns to 100s ns with decreasing temperature from the lipid main transition temperature.
More recently we have also begun to combine neutron time-of-flight and backscattering measurements to also study the fast dynamics in the lipid fluid phase. These efforts include separating the coherent and incoherent contributions to the measured dynamics using polarized QENS. We will showcase results for saturated phosphatidylcholine lipids with different tail lengths and compare the measured relaxation dynamics with reported values for the membrane viscosity in literature.

Current status of the neutron spin echo spectrometer, iNSE at Japan Research Reactor-3

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After a prolonged shutdown since 2011, the Japan Research Reactor 3 (JRR-3) in Tokai, Japan resumed operation in 2021. The neutron spin echo spectrometer at JRR-3, named iNSE has been under commissioning by a new instrument team. We completed repairs or maintenance of essential apparatus such as precession coil cooling system and two-dimensional detector in 2022 and confirms that the spin control devices, including the spin polarizing bender, spin flippers, and spin analyzer are working without major problems. For FY2023, iNSE accepted six user experiment proposals in the field of soft matter and the experiments will start soon.

The current status of iNSE and future plan will be presented.

Diffusive Dynamics of Bacterial Proteome as a Proxy of Cell Death

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Session: Liquids/**Biology**/Materials/Magnetism

Temperature variations have a big impact on the bacterial metabolism and death, yet an exhaustive molecular picture of these processes is still missing. For instance, whether the thermal death is determined by the deterioration of the whole or a specific part of the proteome is hotly debated. In our work [1], by monitoring the proteome dynamics of E. coli we show that only a minor fraction of the proteome unfolds at the cell death. First, we prove that the dynamical state of the E. coli proteome is an excellent proxy for the temperature dependent bacterial metabolism and death. The proteome diffusive dynamics peaks at about the bacterial optimal growth temperature, then a dramatic dynamical slowdown is observed which starts just below the cell's death temperature. Next, we show that this slowdown is caused by the unfolding of just a small fraction of proteins which establish an entangling inter-protein network—dominated by hydrophobic interactions— across the cytoplasm. Finally, we prove that the deduced progress of the proteome unfolding and its diffusive dynamics are both key to correctly reproduce the E. coli growth rate.

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SHERPA: A Polarized QENS Instrument for ISIS

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Session: Instrumentation

Quasielastic neutron scattering with polarisation analysis (PA-QENS) has the potential to revolutionise the field of QENS. This arises from the ability to unambiguously separate the coherent and incoherent contributions to the scattering. Particular areas of application are where there is a significant coherent contribution to the scattering, such as the use of buffers for biological materials or diffusion in a zeolite. In this poster we will describe SHERPA [1], which is a PA-QENS instrument proposed for ISIS. Based on ray-tracing simulations, we expect a greatly increased count rate compared to the present QENS spectrometer IRIS at ISIS. This is expected to be between 49 and 660 × IRIS in the conventional (i.e. unpolarised) mode and ~5-70 × IRIS in the PA-QENS configuration. This huge gain in the count rate would be achieved from the combination of three factors: a modern neutron guide with high *m* coating, the prismatic effect and a larger solid angle coverage at the energy analyser. We will illustrate the possibilities of the instrument with some current examples of PA-QENS studies.

QENS study of citrate dynamics on the surface of iron oxide nanoparticles

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Session: Soft Matter and Liquids

Surface functionalization is needed for the synthesis and the control of properties of iron oxide nanoparticles (IONPs) in various applications for biomedicine, ferrofluids, or heterogeneous catalysis[1-2]. Yet, experimental investigations of interfacial properties such as the dynamics of ligand and water molecules near the nanoparticle surface have been scarce. Previously, quasielastic neutron scattering (QENS) was successfully used to access the dynamics of water molecules on surfaces of TiO_2 and SnO_2 NPs[3]. QENS studies of ligand dynamics on the surface of NPs have also emerged recently. Dodecanethiol on PbS nanoparticles and oleates on IONPs were shown to exhibit rotational motion[4-5]. However, the dynamics of citrate ligands on the surface of magnetic IONPs are largely unknown. We report on QENS experiments on 6 nm IONPs synthesized according to ref 6 and stabilized at 8 % relative humidity in H_2O (measured at IN16B, ILL) and D_2O (measured at Emu, ANSTO). Elastic and inelastic fixed window scans in the temperature range of 2 – 334 K and covering a Q-range of 0.19 – 1.83 Å^{-1} were used to distinguish contributions from phonons, magnetic relaxations, and ligand motion. We stress the broad understanding which can be gained just from fixed window scans by comparing scaled changes of elastic and quasielastic intensities at different Q values in- and outside of magnetic Bragg peaks. Further, the Q-dependence of energy-resolved spectra shows, that citrate molecules only exhibit a rotational motion on the IONP surface.

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Structure and dynamics in dense suspensions of thermoresponsive polymer micelles

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Session: Soft Matter and Liquids

“Classical” block copolymers consisting of hydrophobic A and hydrophilic B blocks self-assemble into micelles when dissolved in water. Diblock (AB) copolymers form star-like micelles, whereas in triblock (ABA) copolymers the B block can either form a loop (flower-like micelle, both A blocks in the core) or bridge two different micelles (outer A blocks placed into two different cores). At high polymer concentration, di- and triblock copolymer micelles are densely packed and can form highly viscous, jammed suspensions. In the case of triblocks, the possibility of micellar bridging can in addition lead to the formation of clusters and ultimately to a (transient) network.
A special case is that of block copolymers formed by polystyrene (PS) and poly-N-isopropylacrylamide (PNIPAM) units. While PS is persistently hydrophobic, PNIPAM switches from hydrophilic to hydrophobic behaviour when the temperature is increased above a lower critical solution temperature of around 32 °C. The availability of switching “smart” micellar gels finds potential for applications ranging from drug delivery to sensors. From a fundamental point of view, the behaviour of dense solutions of PS-PNIPAM and PS-PNIPAM-PS block copolymers is however rather complex. Due to the high glass transition temperature of PS, the micellar cores are “frozen” and the kinetics of unimers exchange is quite slow. This makes the properties of PS-PNIPAM-PS suspensions somehow intermediate between those of “classical” triblock micelles and permanently cross-linked networks.

For this study, PS-PNIPAM and PS-PNIPAM-PS block copolymers were synthesised with narrow molecular weight distribution. Concentrated suspensions of block copolymer micelles were investigated with small angle neutron scattering (SANS), rheology, dynamic light scattering (DLS), pulsed field gradient (PFG)-NMR, and neutron spin echo (NSE). From the combination of structural and dynamical information, new insights into the role of polymer architecture and bridge formation in dense “smart” micellar suspensions are obtained.

Dynamics in systems with supramolecular structures of different topologies

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Session: Soft Matter and Liquids

We have recently started a project whose general objective is the characterization by neutron scattering of the coherent and incoherent scattering functions (structural relaxation and H-self-dynamics) of systems with different specific interactions and/or kinds of nano-structuration, from mesoscales to intermolecular length scales. Systems with hydrogen bonds (alcohols) leading to supramolecular aggregates with different topologies, as for instance chain-like or ring-like, and highly polar liquids are investigated. Combining different spectrometers including neutron spin echo, backscattering and time-of-flight instruments, the relaxation of the emerging nano-structures is accessed. One of the main interests of this study is to check in these systems the universality of a Q-independent mode recently found in water and tetrahydrofuran at mesoscales [1,2]. Another focus lies on the direct comparison of the microscopic information obtained from the different correlators accessed by neutron scattering with that compiled from other spectroscopic methods (dielectric, mechanical), that shall shed light on the puzzling situation currently present in the community when interpreting results from different macroscopic techniques.

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Influence from polarised QENS for proteins in solution

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Session: Soft Matter and Liquids/Biology

Protein dynamics play a vital role in biology. Quasi elastic neutron scattering is an ideal method to access these dynamics. Normally data analysis is performed based on the assumption that the scattering spectrum is incoherent. In order to be observe the full range of protein dynamics it is necessary to perform the experiments in solution. This solution is usually a fully deuterated buffer, while the protein remains protonated. It is generally assumed that while the buffer leads to a coherent contribution, this can be taken into account during data analysis by subtracting the buffer contribution from the sample spectrum. Up until recently there was no way to experimentally verify this assumption. Polarised QENS experiments on LET allow for the coherent and incoherent contributions to be separated. By comparing the results from the polarised QENS experiment and the standard analysis method from unpolarised QENS it was possible to experimentally check this assumption. This has shown that the pure incoherent spectrum obtained from polarisation analysis does not match the results for normal QENS. The implications of this will be discussed.

Bridging MD simulations with mutual scattering methods

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Session: Instrumentation/Soft Matter

As central quality the (dynamic) structure factor S (q,ω) links molecular dynamics (MD) simulations and scattering experiments with each other, either directly via the Van-Hove Formalism or by employing ray tracing techniques. Both probing the same length (0.1- 100 nm) and time scales (from sub ps to several 100 ns), that are at the very core of hard and soft condensed matter physics.
Here we review the theoretical relationship between the trajectories obtained from (MD) simulations and commonly used scattering techniques, based on the central role of S (q,ω) that

(i) links MD-simulations and all scattering techniques with each other - making them comparable,
(ii) experimentally obtained scattering data can serve to scale the force fields (interatomic potentials) employed in MD simulations recursively and
(iii) allows to potentially overcome the inherent interpretation-issues occurring if (quasi) elastic and inelastic scattering can't be neglected, but cannot be separated in an experiment.

With the contribution, we emphasize the need for tools that are easily handleable and can precisely translate between simulations and experiments, taking into account individual instrumental characteristics.

Concentration dependent dynamical investigation of hydrogen bonds in propanol-glycerol mixtures and evidence of slower relaxation process

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Session: Soft Matter and Liquids

Given the pervasiveness of hydrogen bonds in nature and their critical role in technological growth, it is imperative to comprehend their dynamics. Of course, water is the most significant hydrogen-bonded liquid, yet studying it can be difficult due to its very anomalous behaviour and difficulty in supercooling. In other hydrogen bonded systems, e.g., in poly-alcohol and biomolecules the dynamics of the hydrogen bonds merge with other dynamics. Mono-hydroxy alcohols have proven to be an interesting case for the study of hydrogen bonds, because the dynamics connected to the hydrogen bonds appears well separated in these systems. The mono-hydroxy alcohols have been studied extensively over the last decades due to their interesting relaxation pattern. Specifically, by dielectric spectroscopy it has been demonstrated that the hydrogen bonds of the mono-hydroxy alcohols give rise to a dominant relaxation process, the so-called Debye process, which is slower than the α-relaxation[1-3]. In the past years there is evidence building up that the relaxation of poly-alcohols can in fact also be separated into a Debye like process and an α-relaxation[4], though the processes are more merged in the case of poly-alcohols compared to mono alcohols. In this work, we study the dynamics of the hydrogen bonds in the mono-alcohol propanol with quasi-elastic neutron scattering. We compare this to a sample where the concentration of hydrogen bonds is increased by adding the poly-alcohol glycerol to the propanol. The addition of glycerol yields a stronger signal in the slowest part of the signal.

We study the mixtures both protonated and in a partly deuterated sample where the dynamics of the OH-groups are invisible to the neutrons. The deuteration strongly suppresses the slow part of the signal, which clearly demonstrates that this slow part of the dynamics is due to the hydrogen bonds. Model-fitting shows that the slow dynamics can be considered as a mode separated from the overall relaxation in the liquids.

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Data Collection for Dilute Aqueous Solutions via a Neutron Backscattering Spectrometer

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Session: Instrumentation

Incoherent quasi-elastic neutron scattering (QENS), which utilizes the highly incoherent scattering ability of hydrogen, is a powerful technique for revealing the dynamics of polymers or proteins in deuterium oxide (D_2O) solutions or moistures. The background scattering of sample cells suitable for aqueous solution samples, conducted with a neutron backscattering spectrometer, is evaluating[1–2]. Recently an aluminium sample cell coated with boehmite using D_2O was used for the QENS measurement at BL02 (DNA) in MLF, J-PARC, Japan for a 0.8 wt.% aqueous solution of a protein[3]. The cell was inert against aqueous samples at 283–363 K. In addition, meticulous attention to cells with small individual weight differences and the being great positional reproducibility of the sample cell relative to the spectrometer neutron beam position enabled the better subtraction of the scattering profiles of the D_2O buffer and the sample container. Consequently, high-quality information on protein dynamics could be extracted from dilute solutions. Some studies will be presented.

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The direct geometry cold chopper spectrometer TOFTOF

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Session: Instrumentation

TOFTOF is a direct geometry disc-chopper time-of-flight spectrometer. A cascade of seven fast rotating disc choppers is used to prepare a monochromatic pulsed beam which is focussed onto the sample by a converging super-mirror section. The scattered neutrons are detected by 1000 3He detector tubes with a time resolution up to 50 ns. The detectors are mounted at a distance of 4 m and cover 12 m2 (or 0.75 sr). The high rotation speed of the chopper system together with a high neutron flux in the wavelength range of 1.4 -14 Å allows free tuning of the energy resolution between 3 meV and 2 μeV.
The fast neutron background is suppressed by the s-shaped primary neutron guide. This enables the investigation of weak signals. The existing linearly tapered neutron guide yields a beam spot size of 23x47 mm2. As alternative option a focussing guide can be used. This leads to an intensity gain up to a factor of 3 (wavelength dependent) on a sample area of 10 x 10 mm2.

TOFTOF represents a versatile instrument combining high energy resolution, high neutron flux (also at short wavelengths), and an excellent signal-to-background ratio. It is perfectly suited for inelastic and quasielastic neutron scattering and scientific topics include e.g.:

- Diffusion in liquid metals and alloys
- Hydrogen dynamics in soft matter systems such as molecular liquids, polymer melts or colloids
- Molecular magnetism, quantum criticality in heavy fermion compounds, low energy excitations
- in multiferroic materials and novel magnetic phases
- Dynamic properties of energy storage materials, such as solid state hydrogen storage
- materials, electrolytes, or gas storage materials
- Energy-resolved quasi-elastic neutron scattering on proteins, vesicles, and biological materials
- Kinetic studies of hydrogen binding
- Aging effects in disordered media and low frequency dynamics in glasses
- Biological activity and functionality of proteins and cells under pressure

Pressure effect on protein cluster formation induced by multivalent ions

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Session: Soft Matter and Liquids

A thorough understanding of protein interactions in aqueous solutions is crucial for many areas of research in soft matter and biology. For example, a strong interprotein attraction can lead to protein aggregation, which is observed in several pathologies such as cataract and neurodegenerative diseases.

We have shown that a patchy particle model can describe the phase behavior of a system of acidic globular proteins such as bovine serum albumin in the presence of multivalent salts such as yttrium chloride. The phase diagram of the studied system as a function of salt concentration and temperature is quite complex, showing reentrant condensation, metastable liquid-liquid phase separation, cluster formation and crystallization. In particular, a lower critical solution temperature is observed which suggests that hydration plays an essential role in the ionmediated protein interactions.

This is also visible by changing the solvent from H2O to D2O. It leads to an increasing attraction potential between the proteins and the formation of clusters is observed. By neutron spectroscopy a slowing down of the protein short-time self-diffusion as a function of available yttrium ions per protein is observed. The effect is enhanced by increasing the temperature of the sample.

Using temperature as a control parameter has some disadvantages because temperature influences both the thermal energy and the density of the system. Furthermore, only a small temperature range is available for studying proteins since high temperatures lead to denaturation. As opposed to temperature, pressure influences only the density and can be considered to have milder effects.

Here we will present results from pressure dependent neutron spectroscopy experiments. In contrast to the previous studies at and above room temperature we found, that the slowing down of the short-time self-diffusion is less pronounced. This behavior of the short-time selfdiffusion will be discussed with the help of pressure dependant SAXS measurements.

The high resolution neutron backscattering spectrometer SPHERES

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Session: Instrumentation

The neutron backscattering spectrometer SPHERES (SPectrometer for High Energy RESolution) at MLZ is a third generation backscattering spectrometer with focusing optics and phase-space transform (PST) chopper. It covers a dynamic range of ± 31µeV with a high resolution of about 0.66µeV and a good signal-to-noise ratio. The instrument performance has been improved over the recent years by different measures. The intensity has been more than doubled by the upgrade of the PST chopper and the focusing guide. The signal-to-noise ratio can be significantly increased by employing the new background chopper.
SPHERES enables investigations on a broad range of scientific topics from the classical applications of backscattering like hyperfine splitting to investigations on new materials like high temperature polymer electrolyte fuel cells. It is in particular sensitive to the incoherent scattering from hydrogen and allows to access dynamic processes up to a timescale of a few ns. Therefore it is well suited to study the dynamics in soft matter systems like polymers and nanocomposites or in proteins. Other typical applications include molecular reorientations or motion of viscous liquids and water in confined geometry.

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