Science and Technology Facilities Council

Conference Proceedings
STFC-CONF-2023-002

Simulation Meets Molecular Spectroscopy (SiMol) Meeting

Milton Hill House, Abingdon, UK
30th October - 1st November 2023

J Armstrong, M Sarter (editors)

November 2023

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DOI: 10.5286/stfcconf.2023002

ISSN 2753-5800

Book of abstract for the 2023 SiMolSpec meeting

Combining Quasielastic Neutron Scattering and Molecular Dynamics to Study Methane Motions in ZSM-5

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Quasi-elastic neutron scattering (QENS) and molecular dynamics (MD) simulations are applied in combination to investigate the dynamics of methane in H-ZSM-5 zeolite catalysts used for methanol-to-hydrocarbons (MTH) reactions. Methane is employed as an inert model for the methanol reaction feedstock and studies are made of the fresh catalyst and used catalysts with varying levels of coke build-up to investigate the effect of coking on reactant mobility. Measurements are made in the temperature range from 5 – 373 K. Methane mobility under these conditions is found to be extremely high in fresh ZSM-5, with the majority of movements occurring too fast to be resolved by the QENS instrument used. A small fraction of molecules undergoing jump diffusion on QENS timescales is identified and found to correspond with short-range jump diffusion within single zeolite pores as identified in MD simulations. Agreement between QENS and MD mobility measurements is found to be within 50%, validating the simulation approach employed. Methane diffusion is found to be minimally affected by moderate levels of coke build-up, while highly coked samples result in the confinement of methane to single pores within the zeolite with minimal long-range diffusion.

Reference

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Project Abstract

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Diffusion of hydrogen in steel pipelines results in embrittlement (HE). A variety of hydrogen sources like cathodic protection [CP] and corrosion results in H diffusion and embrittlement of steel. Detection of hydrogen in steels is the primary challenge. Conventional techniques for elemental identification and quantification such as mass spectrometry are unable to measure hydrogen directly, thereby limiting the understanding of how hydrogen leads to cracking. The possibility to visualize hydrogen distribution *in situ* would allow quantification and a clearer interpretation of the processes involved in HE-assisted fatigue crack growth. There is a pressing need to advance our mechanistic understanding of hydrogen transport, storage and embrittlement in structural materials.

The visualisation of hydrogen distribution via neutron imaging would allow quantification and a clearer interpretation of the processes involved in HE-assisted fatigue crack growth in steels. The corresponding transmission obtained from the evaluated intensities in both charged and uncharged samples is measured using IMAT. The results will determine the minimum measurable hydrogen concentration in steels, which will help determine the sensitivity of neutron imaging at IMAT and facilitate an understanding of the relationship between H concentration in steel and fatigue crack growth rates (FCGRs). Once the sensitivity is established, local hydrogen detection ahead of crack tips will be made and correlated to FCGRs.

Influence from polarised QENS for proteins in solution

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ISIS Neutron and Muon Source

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.

Understanding how inhaled particles impact the brain-blood-barrier

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Worldwide, 50 million people live with dementia and this number is predicted to triple over the next 30 years. It is estimated that the economic impact of dementia is greater than that attributed to cancer and heart disease combined. In the UK, the total cost of dementia, including costs to the NHS, paid social care and unpaid care, has risen to £34.7bn per year, and will rise to £94.1bn by 2040. In the 2020 Lancet Commission on Dementia Prevention, Intervention, and Care, air pollution was included as one of the three newly added modifiable life-course risk factors for dementia. Accumulating epidemiological evidence has shown that exposure to air pollution, especially ambient particulate matter (PM), is an important environmental risk factor for neurodegenerative and neurological disorders, such as Dementia. Given the significant impact of brain diseases on individuals, the population at large and the economy there is an urgent need to better understand the causal pathways underpinning these observed associations.
Some have suggested that inhaled particles could impact the brain directly, by entering the brain via the olfactory nerve, or across the blood-brain barrier (BBB). Other hypotheses suggest that effects on the brain are indirect and that chemical messengers released in the lung when exposed to air pollution may reach and travel across the BBB to promote neuroinflammation and damage the brain. A better understanding of the precise mode of action of inhaled PM, allied to a hazard ranking of the constituents within this complex mixture will help inform the design of effective policy and interventions to maintain optimal brain health in the population.

This research study is part of a scoping project “Impacts of inhaled particles on the brain”, which was awarded by the Science and Technology Facilities Council (STFC) Air Quality Network (SAQN) in 2021. This study will employ ambient PM, as well as source specific samples (diesel exhaust PM) combined with focused ion beam scanning electron microscopy (FIB-SEM) and Confocal microscopy at central laser facility (CLF) to explore if and how particles translocate across the blood-brain-barrier (BBB). We will also apply the ISIS Neutron and Muon Source facilities at STFC to investigate the effects of particulate exposure on lipid dynamics, which would provide insights into biological endpoints likely to be affected by exposure to air pollutants and contribute to the growing scientific understanding of the mechanisms by which inhaled particles reach and impact upon the brain.

Simulated INS experiments from phonon calculations

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Vibrational spectroscopy is sensitive to changes in chemical environments, and inelatic neutron scattering (INS) can provide access to vibrational spectra inside a solid lattice, away from the Gamma-point and without selection rule restrictions. The resulting spectra are difficult to interpret intuitively and so are routinely compared with frequencies from atomistic simulation methods such as density-functional theory.

However, the measured scattering function is not precisely equivalent to the phonon DOS or band structure obtained by such calculations. In order to reach more direct comparisons between theory and experiment we need to account for additional complications. Using software developed at STFC we can account for instrument geometry, resolution limits and multi-phonon effects. In the AbINS package, instrument parameters are selected from a user-friendly interface and phonon data is imported from popular atomistic codes. Fine numerical sampling with the Euphonic library allows subtle dispersion effects to be accounted for.

We examine some of the long-established assumptions in these software tools as well as recent developments and good practices.

Catalysis in Zeolites, Neutron Spectroscopy Meets Simulation

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Lignin, a major component of lignocellulosic biomass comprising 40% of its energy, holds great promise as a renewable alternative feedstock to crude oil for producing fuels and chemicals. Zeolites show potential as catalysts for lignin conversion, with their versatile framework topologies and acid site densities enabling precise control of the catalytic process. Cresol molecules, obtained from lignin pyrolysis, are model lignin monomers that can be upgraded into various chemicals including pharmaceuticals, polymers and fuels. Neutron spectroscopy techniques have been employed to study the rate-limiting steps involved in cresol conversion, namely diffusion throughout the zeolite framework and adsorption onto the acid sites.
In this study, we employed quasielastic neutron spectroscopy and molecular dynamics (MD) to investigate the mobility of cresol isomers within highly Brønsted acidic zeolites H-Y and H-beta, with pore sizes of 7.4 and 6.7 Å respectively. Our results showed that, while liquid cresol exhibits both long-range jump diffusion and local rotational motions, when confined within zeolites only a slower isotropic rotation motion was observed on the timescale of the instrument. More mobility was observed in H-Y compared to H-beta, corresponding with lower rates of rotation. By accessing longer timescales using MD, we observed translational diffusion from 3-7 x 10-11 m2s-1. The slow self-diffusion was attributed to significant hydrogen bonding interactions. Inelastic neutron spectroscopy was applied to further investigate the adsorption interactions, which indicated that cresols bonded to the zeolite acid sites through their hydroxy group. Our future work will involve performing density function theory calculations to gain insights into the strength and location of these interactions, thereby enhancing our understanding of the processes pertaining to biomass conversion.

Quasielastic Neutrons Scattering with polarisation analysis at LET

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Polarisation analysis (PA) plays an increasing role in the neutron scattering, because it allows to separate explicitly magnetic, nuclear coherent and nuclear spin-incoherent contributions to the scattering function. Although PA is explored for many years, due to severe limitations in the count rate as well as complicated technical realization it is still not very common for time-of-flight spectroscopy.
Recently implemented polarised option at LET offers an opportunity to apply the PA to the study of diffusion and molecular dynamics in such objects as aqueous systems, proteins, polymers, metal-organic frameworks, proton and alkali-ion conductors.
Herein we discuss the capabilities of the polarised LET and present recent user highlights on few the above-mentioned topics.

Cation Dynamics as a Structure Explorer in Hybrid Perovskites:a Neutron Scattering Perspective

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Hybrid Organic-Inorganic Perovskites (HOIPs) have emerged as a promising and very popular class of materials for efficient solar energy conversion and a growing range of optoelectronic appli- cations, including light-emitting diodes and photodetectors [1]. To date, a considerable number of studies aimed at determining the structural properties of HOIPs using diffraction and optical spectroscopy continues to reveal evident inconsistencies and challenges [2-3]. To a significant extent, these difficulties can be traced back to the extraordinary softness of the metal-halide framework [4], which results in pronounced deformations of the inorganic scaffold and the emergence of nano-size domains of different origins [5-6]. As a result, a consistent understanding of these materials at the atomic scale has not been reached to date. This situation is particularly severe for the case of the quintessential HOIP methylammonium lead iodide ($MAPbI_3$), where the ferroelastic relations between thermodynamically stable phases result in intrinsic crystal twinning, severely impedinga thorough understanding of its structure using diffraction techniques. In this contribution, we present our recent efforts to explore the local structure around the organic cations, exploiting the exceptional sensitivity of Inelastic Neutron Scattering (INS) to hydrogen motions [7]. We also provide an overview of our methodology, combining broadband high-resolution INS experiments with state-of-the-art *ab initio* molecular dynamics beyond the harmonic picture, to interrogate local structure in the archetypal case of $MAPbI_3$ across its pressure-temperature phase diagram and under chemical pressure induced by cation engineering [8-12].

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Abstract

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Computer simulations such as Molecular Dynamics (MD) are an effective tool in studying a broad range of structural and dynamical properties in atomic-scale systems, but the connection to X-ray or neutron scattering experiments is not always straightforward. In this work, we present a Python workflow that starts with the construction of machine learned potentials and ends with the prediction of experimental observables, such as the dynamical structure factor. Specifically, the Python package Calorine is used to train neuro-evolution potentials (NEP) with ab-initio level accuracy using the software package GPUMD. The trained potentials are then used to run MD within GPUMD, also done through Calorine. Finally, the resulting MD trajectories are analyzed with the Python package Dynasor in order to compute dynamical structure factors, which in turn may be convoluted with instrument parameters for a direct comparison to experiments. We apply our workflow to crystalline benzene as a prototype system, but the workflow may be readily extended to different systems.

Abstract

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In my group we are very interested in using molecular dynamics simulations to understand the molecular scale interactions that drive the self-assembly of soft matter. We have found that the results obtained from our simulations are often the perfect complement to the results that are obtained from neutron scattering experiments and therefore allow us to provide a truly multi-scale understanding of the structural properties of the resulting nanoparticles. In this talk, I will present two different recent studies where we have used molecular dynamics simulations in combination with machine learning and graph theoretical approaches to provide a unique understanding of the mechanisms that drive the self-assembly of polymers and lipids. In doing so, I will also highlight how we are able to combine our results with those of neutron experiments done by experimental collaborators to provide a complete understanding of each system.

Abstract

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Bacteria are now becoming more resistant to most conventional antibiotics. Extended spectrum beta-lactamase resistant (ESBL-E.coli), tetracycline resistant E.coli (TetR-E. coli), a complex of multidrug-resistant Gram-negative bacterial strains, has proven especially problematic in both hospital and community settings by deactivating conventional antibiotics.3 To address the challenging situation of potency, spectrum, toxicity and drug resistance in early stages of antimicrobial drug discovery, synergistic combination agents provide an important and largely unexploited strategy to 'repurpose' existing chemical space. Here, a simple strategy that by molecular combination of the tetracycline/minocycline (TC/MC) and a broad-spectrum antimicrobial lipopeptide (C8G(IIKK)2I and C8G(IIRR)2I were designed based on previously designed G3) with efficient bacterial cell disruption was proposed. This approach allows TC/MC to approach onto bacteria membrane as a combination with lipopeptide via complex hydrophobic interactions, that improves the efficiency of antibiotic's membrane targeting and its later intramembrane accumulated concentration. Also, such kind of combination kept the advantage of lipopeptide's fast time-killing and facilitated tetracycline uptake after lipopeptides' effects on outer membrane permeability and dissipation of inner membrane potential.

SIMULATION as a TOOL for PREDICTING STRUCTURAL and DYNAMIC PROPERTIES of SOLIDS and THEIR SURFACES

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We will provide a general overview of the role and capabilities of modelling techniques in solid state and surface science, and will then focus on three areas

(i) Modelling and prediction of the structures of crystals, surfaces and nano-particles,

(ii) Modelling of the dynamical properties of sorbed molecules,

(iii) Modelling of vibrational spectra of sorbed species.

We will discuss the complementarity of simulation tools to neutron based experiment, especially neutron spectroscopy.

Abstract

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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
• 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 biological materials
• Kinetic studies of hydrogen binding
• Aging effects in disordered media and low frequency dynamics in glasses

Here we will give an actual update about the status of the instrument and the available sample environments. Additionally we will show some ideas for an upcoming upgrade of the instrument.

New Strategies to Fight Cancer – The Role of Neutrons

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Normal-to-cancer transition (NTC) is still an ill-understood process, closely related to cellular biomechanical properties. These are strongly dependent on intracellular water´s structural and dynamical profiles, which play a fundamental role in cellular function. Improved chemotherapeutic strategies are an urgent clinical need, since cancer is still the second leading cause of death worldwide with an expected rising incidence. Metal-based drugs developed upon the discovery of cisplatin (*cis*-$(NH_3)_2PtCl_2$) have aimed at coupling an enhanced efficacy to decreased acquired resistance and deleterious side effects. These include Pt- and Pd-complexes with more than one metal centre, which trigger a selective DNA damage – through metal coordination to DNA bases or *via* electrostatic interaction with the phosphate groups.

The authors have applied inelastic and quasi-elastic neutron scattering (INS and QENS), complementary techniques to Raman and Fourier Transform Infrared (FTIR, including with synchrotron radiation), to provide a comprehensive set of data, at the conformational and dynamic levels, on: (i) NTC transformation; (ii) activity of newly developed Pt/Pd-anticancer agents (on DNA, glutathione, proteins, cellular metabolism and intracellular water). Variations in the dynamical profile of intracellular water were unveiled for malignant cells/tissues as compared to healthy ones. In addition, clearly distinct effects were revealed for Pt- *vs* Pd-agents regarding their impact on either the cellular cytoplasm or hydration water in cancer cells, as well as concerning their specific interactions with biomolecules.

This innovative assessment of the impact of a chemotherapeutic agent on the dynamics of vital cellular components (such as DNA) through an effect on their hydration layers, as well as on their native structure, is key for a thorough understanding of their molecular basis of cytotoxicity. This will contribute to the design of new anticancer agents with optimised efficacy, that may act simultaneously on more than one site (multitarget approach), intracellular water being suggested as a potential pharmacological target.

Abstract

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Inverse eigenstrain analysis methodologies have demonstrated their efficacy in the reconstruction of residual stresses within planar eigenstrain problems, commonly referred to as "continuously processed bodies." However, the reconstruction of residual stresses in bodies subject to discontinuous processing, or those exhibiting intricate shapes and/or complete eigenstrain distributions, presents significant challenges and necessitates the adoption of simplifying or regularization assumptions. In this study, a numerical experimentation approach is employed to propose a novel and comprehensive inverse eigenstrain method capable of accurately reconstructing eigenstrain and residual stress fields in discontinuously processed bodies. The method introduced in this paper leverages the superposition of eigenstrain radial basis functions, in conjunction with a limited set of experimental data, to achieve model-free (unconstrained) determination of unknown eigenstrain fields. A notable feature of this approach is its capacity to account for all six strain components within an isotropic body, without resorting to simplifying or regularization assumptions. By expressing the eigenstrain distribution as a weighted summation of radial basis functions, the limitations imposed by global basis functions such as polynomials are circumvented. Through the avoidance of simplifying assumptions and the utilization of intricate formulations, the accuracy of the 3D eigenstrain reconstruction method using radial basis functions becomes intrinsically tied to the quality and quantity of the experimental data employed for reconstruction, as well as the accurate definition of boundary conditions.

Role of hydration in the flexibility of mucins

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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. Hydration of these residues, and the subsequent excluded volume interactions are thought to modulate the flexibility of the chain. We have observed and quantified the number of water molecules strongly bound to the glycosylations 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.

Abstract

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Measurement of the dynamics of confined fluids is well established with QENS. One application is investigating diffusion of adsorbed species in catalytic zeolites. In these heterogenous systems it is difficult to separate the elastic and inelastic scattering contributions of the two materials and this must be treated in the data analysis. It is an indication of this difficulty that there is no commonly accepted method to separate the scattering contributions, although many treatments have been used.

Improvements in instrumentation now mean that polarization analysis (PA) is now practical with QENS to separate the coherent (non spin-flip) and incoherent (spin-flip) scattering components in an experimental measurement. This has shown that a more nuanced view of the components in the time dependent scattering function is needed.

For an absorbate in a zeolite crystal, both the coherent structure and dynamics will be dominated by the zeolite, with the incoherent signal mostly arising from the sorbate. This allows unprecedented clarity in separation of the elastic signal from the two materials, vastly improving determination of the EISF. For determination of peak widths, this allows removal of the Bragg peaks from the zeolite, which can overwhelm the inelastic signal when measured without polarization.

We will present a PA-QENS study of the diffusion of cyclohexane in ZSM-5 to show the effectiveness of this technique in a catalytic context. This powerful new method is currently limited by the instruments available and a proposed dedicated backscattering PA-QENS instrument (SHERPA) will be briefly presented.

Abstract

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The unique properties of Egyptian blue, a cuprorivaite pigment, were herein studied through a holistic computational and experimental approach. A reliable model of the crystal of cuprorivaite was obtained through periodic-DFT calculations, allowing for the elucidation of its lattice dynamics, including assessment of structural, electronic and vibrational properties.
From this model, a sound assignment of the inelastic neutron scattering spectrum was obtained, along with estimated values of heat capacity and Debye temperature, band-gap values and magnetic properties of the crystal. Inelastic neutron scattering and diffuse reflectance infrared Fourier transform spectroscopies provided enlightenment on the chemical surfaces of the pigment Egyptian blue – which was missing hitherto and is critical for potential applications of the pigment, such as those involving host-matrix interactions or requiring surface derivatization, – while confirming the simulation results. On that account, it was found that the intensity of the stretching mode of the dangling SiOd moieties is ca. 8%-13% of the total SiO stretching modes. Moreover, and regarding the electronic properties, the band structure confirmed that $CaCuSi_4O_{10}$ is a direct band gap semiconductor, with the Valence Band Maximum (VBM) and the Conduction Band Minimum (CBM) located at the gamma-point, in both the alpha (spin-up) and beta (spin-down) density bands.

Accurate Estimation of Diffusion Coefficients and their Uncertainties from Computer Simulation

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Self-diffusion coefficients, D*, are routinely estimated from molecular dynamics simulations by fitting a linear model to the observed mean-squared displacements (MSDs) of the mobile species. MSDs derived from simulation suffer from statistical noise, which introduces uncertainty in the resulting estimate of D*. An optimal scheme for estimating D* will minimise this uncertainty, i.e., will have high statistical efficiency, and will give an accurate estimate of the uncertainty itself. We will present a probabilistic scheme for estimating D* from a single simulation trajectory with high statistical efficiency and accurately estimating the uncertainty in the predicted value. The statistical distribution of MSDs observable from a given simulation is modelled as a multivariate normal distribution using an analytical covariance matrix for an equivalent system of freely diffusing particles, which we parameterise from the available simulation data. We then perform Bayesian regression to sample the distribution of linear models that are compatible with this model multivariate normal distribution, to obtain a statistically efficient estimate of D* and an accurate estimate of the associated statistical uncertainty. We will then go on to discuss how the probabilistic approach taken could be extended to allow for a more accurate estimation of the intermediate scattering function and, therefore the dynamic structure factor, from molecular dynamics simulation.

Abstract

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Prohibitins (PHB1 and PHB2) are highly conserved heterodimeric proteins arranging into a large multimeric ring at the inner mitochondrial membrane. They play a crucial role in premature cellular aging, tumor suppression, cell cycle regulation, apoptosis and mitochondrial homeostasis via their interaction with AAA-proteases. We set out to (i) characterize the interaction between the N-terminal helices of PHB (NT-PHB) with the membrane and establish a possible synergy of the two PHB homologues, and (ii) understand the role of cardiolipin in this interaction using interface and bulk neutron techniques.

NR and QCM-D experiments demonstrate that both peptides are able to remove lipid from the bilayer. In SANS, the addition of peptide disrupts the membrane, modifying its integrity by fusing membranes and removing lipids from the vesicles. The effect of the peptide on the membrane is concentration-dependent as well as dependent on the lipid membrane composition with CL and PE enhancing the effect of the peptides on the vesicles.

We propose that both PHB peptides act in synergism to remodel the inner mitochondrial membrane in order to create raft-like areas that allow the support of large protein complexes such as the hexameric AAA-proteases. Future studies will investigate how the interaction between PHB and AAA-proteases is realized at a molecular level in order to understand PHB's role in mitochondrial homeostasis.

In situ XANES study of electronic state evolution and bonding of gold atoms in glasses

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The history of developing gold (Au)-doped coloured glass could be traced back to the 4th century or even earlier. It has been established that precipitating gold nanoparticles (Au NPs) in glass represents an effective way to develop unique colours. One critical step in the process of forming Au NPs is to reduce gold ions (Au+) into gold atoms (Au0), which significantly affects the subsequent growth and distribution of Au NPs, alongside colour appearance. However, the understanding of the dynamics in this redox reaction and the correlations of processing-chemistry-structure-colour in colour glass is still incomplete. Here, by means of the real-time X-ray absorption spectroscopy (XAS) technology, we provided the first in-situ observation of a one-way progressive change of Au electronic state in glass under heating the samples at elevated temperatures for an extended duration. Our results, on the basis of a series of Linear Combination Fitting (LCF) analysis, can quantify the fraction of Au+ and Au0 throughout, thus for the first time revealing the kinetics of reduction of Au+ ions into Au0 atoms and the variation of Au bonding in glass matrix.

Structure and Dynamics of Imidazole based Organic Ferroelectrics using First Principles Simulations and Neutron Spectroscopies

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Flexible functional materials are important candidates of current research, because of their potential applications in plastic electronics [1]. Organic ferroelectrics are one of such materials having applications in data and charge storage devices as well as in medical electronics. Since flexibility and ferroelectricity are rare combinations, organic ferroelectrics are candidates of intense research promising of designing new ferroelectrics due to their low cost, free from non-toxic materials, flexibility and environment friendliness. Most of these organic ferroelectrics are hydrogen bonded and structure of those hydrogen bonds play an important role in their ferroelctric properties [2]. In these materials chemically distinct donor-acceptor pairs are connected through these hydrogen bonds, which act as a bridge for charge or proton transfer. In presence of an external field this transfer of protons changes the polarisation of these materials by changing geometry from centro symmetric to non-centrosymmetric. The microscopic knowledge of structure and dynamics of proton or the charge carriers are thus important to understand to design these materials fit for the purpose. As a continuation of our previous investigations [2-6] on hydrogen bonded ferroelectrics, in this poster we investigate one of double substituted haloimidazoles ferroelectrics [7], 4,5--dichloro--2--methy--imidazole (C4H4Cl2N2), called DCMeI hereafter, a low temperature ferroelectric, using neutron scattering and first principles simulations.

Using INS and QENS experiments complimented by first principles simulations we will present the investigation on dynamics of N-H—N bonds and their contributions to ferroelectricity along with their structure property correlations. This investigation provides an important insight in the understanding of molecular ferroelectric in organic materials.

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Abstract

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Following our continuing efforts, the structural dynamics of CCl4, CHCl3 and CH2Cl2 chloromethane derivatives was evaluated through our computational spectroscopy approach by comparing experimental INS spectra with the corresponding simulated spectra obtained from periodic DFT calculations. To the best of our knowledge, there are no reports on the Inelastic Neutron Scattering (INS) spectra of these compounds. INS spectroscopy provides a unique assessment of the structural dynamics that is not amenable from its optical counterparts, infrared and Raman spectroscopies. Large amplitude/low wavenumber modes, including lattice modes, usually problematic for optical spectroscopy, tend to yield intense bands in INS spectra. The excellent agreement between experimental and calculated spectra allowed a confident assignment of the vibrational features, including not only the molecular fundamental modes, but also lattice and combination modes. In particular, an impressive overtone sequence for CHCl3 is fully described by the simulated INS spectrum. In the CCl4 spectrum, the splitting of the stretching mode at ca. 765–790 wavenumbers is discussed on the basis of the Fermi resonance vs. crystal splitting controversy.

Abstract

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Proteins are complex molecular systems whose internal dynamics is characterized by a vast spectrum of time scales, ranging from a fraction of a picosecond for bond vibrations to seconds and beyond for large conformational rearrangements. Quasielastic neutron scattering (QENS) probes the self-relaxation of the hydrogen atoms in the ps to nanosecond region, which is characterized by strongly non-exponentially decaying intermediate scattering functions, as far as the intramolecular relaxation of single-domain proteins is considered. In this case the elastic scattering amplitude, the Elastic Incoherent Structure Factor (EISF), cannot be separately measured and must be estimated from an appropriate model. Using QENS spectra from Myelin Basic Protein and myoglobin in solution, we show that a consistent estimation can be achieved by integrating the measured elastic intensity into the fitting procedure, and we propose a generalization of the model to integrate also inter-domain motion of multi-domain proteins. We apply the model to the analysis of QENS data from Phosphoglycerate kinase (PGK), which reversibly catalyzes the production of ATP in many organisms, and we show in particular that our model captures the functional dynamics of this enzyme. To support our findings we present results from normal mode calculations which integrate the amplitude of inter-domain motions monitored by long-time MD simulations.

Hydrogen vibrational dynamics in the nitride-hydride Ca3CrN3H

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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 ions and hydride ions in the same substructure. Recently, the new nitride-hydride Ca3CrN3H was discovered and shown to be promising to be used as a 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 dynamics is crucial, but such an understanding is at present lacking.

In this work, we investigated on the nature of hydride-ion vibrational dynamics in Ca3CrN3H, using inelastic neutron scattering (INS) coupled to density functional theory (DFT) and ab-initio molecular dynamics (AIMD) calculations. The combined analysis of the INS, DFT, and AIMD data reveals vibrational modes of hydride-ions, polarized along the crystallographic c-axis (parallel modes) and in the ab-plane (perpendicular modes), decoupled from the host-lattice vibrations. Interestingly, the parallel modes are found strongly dispersive while the perpendicular modes are not, indicating a strong hydrogen-hydrogen interaction along the c-axis. Moreover, the DFT and AIMD calculations showed that the parallel modes frequency and associated mean square displacement are sensitive to the presence of neighbouring hydride-ions along the c-axis. These modes can therefore be used to probe the hydrogen content in Ca3CrN3H from its INS spectrum and understand the geometry of a potential hydride-ion diffusive motions in the material.

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Exploring asymmetry induced entropy in tetraalkylammonium-urea DES systems: Insights from Inelastic Neutron Scattering.

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In this work, the Inelastic Neutron Scattering (INS) spectroscopy is used to investigate the impact of entropic factors on the behaviour of deep eutectic solvents (DES).
Previous studies took advantage of the unique capabilities of INS to gain insights into the structure and dynamics of choline chloride-based DES [1,2]. It was demonstrated that the chemical surroundings of the cationic head of tetraalkylammonium ions may be probed by observing the methyl torsional modes, whose frequency increases as free space around methyl groups decreases. Methyl torsions are readily observable using INS, but weak and hard to resolve in optical spectra.
Up to date, most fundamental studies focused on enthalpic rather than entropic effects, and the influence of entropic changes remains unclear. This knowledge gap can be address by comparing systems with similar enthalpic profiles but contrasting degrees of tetraalkylammonium symmetry – e.g., substituting an ethyl by a methyl group in tetraethylammonium chloride.
Periodic-DFT calculations provide a reliable assignment of the vibrational modes of pure compounds. This assignment guides the analysis of INS spectra of eutectic mixtures – with particular attention to methyl torsional modes. Deviations from ideality in the mixtures of tetraalkylammonium chloride salts with urea are readily determined through a simplified thermodynamic approach. In conclusion, this study reports and discusses the relationship between the cation's asymmetry, the deviation from ideality, and the INS spectra of the eutectic mixture.

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Abstract

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MDMC: a new program to refine force field parameters against experimental data
We present a newly developed program that combines molecular dynamics (MD) simulations with an optimisation protocol. e.g. using Monte Carlo (MC) methods, in order to determine the force field parameters values that lead to the best agreement with experimental data. The program is currently focussed on classical MD simulations and quasi-elastic neutron scattering (QENS) data, such as dynamic structure factor measurements. However, the program is designed to be extensible to other simulation engines and measurable data types.

Fitting Molecular Dynamics simulations to neutron and X-ray diffraction and spectroscopy data using a Markov Chain Monte Carlo approach on the example of water

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Neutron and X-ray scattering experiments provide valuable insights into the nanoscopic properties of matter, a scale that is also accessible through Molecular Dynamics (MD) simulations. If the simulations repoduce the experiments, they can give greater insight into the material properties on the nanoscopic scale than traditional data analysis methods based on analytical expressions of idealized models.
However, existing MD forcefields are primarily optimized to reproduce macroscopic quantities, limiting their applicability to the study of nanoscopic structure and dynamics. By leveraging scattering data, it becomes possible to parameterize force fields in a manner that optimally captures the behavior of the system at the precise time and length scales they will be evaluated on.
We establish in our research a connection between published experimental data from neutron and X-ray experiments on liquid water, specifically focusing on diffuse scattering and quasielastic neutron scattering, and MD simulations. This connection is facilitated by a Bayesian fit algorithm, which enables us to obtain a set of parameters that simultaneously captures the nanoscopic structure and dynamics as described by neutron and X-ray experiments. To achieve this, we integrate state-of-the-art tools for MD simulation (LAMMPS) and scattering curve computation (Sassena) with a custom-built Bayesian framework that employs a Markov Chain Monte Carlo approach to sample the parameter space. Unlike classical fit algorithms, our approach explores a broader range within the parameter space, enhancing the likelihood of finding the global minimum of forcefield parameters.
It is important to note that this approach is highly versatile and can be adapted to different systems. Here, we utilize liquid water as a proof of concept for the entire workflow.

Calculating scattering patterns from atomistic simulations using the program

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Sassena is one of a few options to compute scattering patterns from atomistic simulations. It was developed originally in the group of Jeremy Smith at Oak Ridge National Laboratory [1]. For both, neutrons and X-rays it can calculate: diffractograms, coherent and incoherent intermediate scattering functions, and elastic coherent/incoherent structure factors. If requested, an orientational average is performed explicitly by computing the quantities for different orientations of the scattering vector. This leaves great flexibility to calculate single crystal, cylindrical or spherically orientational averaged quantities but comes of course with increased performance requirements when compared to analytic orientational averaging with the Debye formula.

Sassena is written in C++ and therefore very fast. It has been created with distributed and shared memory parallelization at heart and has been shown to work on up to 7000 processors and systems with nearly 4 million atoms or 5 million time frames. We have recently improved the single core performance further by introducing vectorization.

We have also implemented the removal of the spurious SANS signal caused by the finite extension of the simulation box. Further work is underway to modernize the program, with a focus on documentation, testing, and the introduction of a Python interface. [2]

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Shapespyer: a Python-driven toolchain for soft matter simulations

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The Shapespyer project is a thriving collaboration between SCD and ISIS under the overarching ALC scheme, which aims to support and facilitate frontier interdisciplinary research by bridging between theoretical analysis, computer simulation and experimental studies. In particular, the primary goal is to equip Small Angle Scattering (SANS/SAXS) experimentalists with seamless simulation and analysis protocols allowing for verification of theoretical conjectures about complex molecular nanoaggregates by direct comparison between SAS experiments and detailed computer simulations. To this end, Shapespyer software package has been developed as an experimentalist-oriented Python-driven framework providing toolchains and workflows to disencumber tedious computer simulations and analyses of semi-functional self-assembling nanostructures ubiquitous in soft matter and biomolecular applications.

Taking a number of template molecules (one per species, optionally as SMILES strings), Shapespyer automatically generates nanoscale aggregates that can be further used for molecular dynamics (MD) simulations carried out on any computing platform, including heterogeneous CPU/GPU HPC resources. The supported molecular arrangements include:

- flat disks or rings
- spheres, e.g. micelles or vesicles
- cylinders, e.g. rods or worm-like structures
- aggregates arranged on 3D lattice(s)
- bicontinuous 3D phases (Schwarz minimal surfaces)

The template molecule coordinates are taken from .gro, .xyz or .pdb files. Shapespyer automates the setup, including the initialisation steps for adding ionic species and solvent, and task-farming for MD simulations with force fields available in Gromacs package.

Specialised batch scripts are provided to perform equilibration and production simulations, with optional post-simulation analyses, which include: (i) cluster classification and size distribution analyses, (ii) radii of gyration and its principal components, (iii) hydration layer and cavity occupation analyses, (iv) molecular order analysis within a cluster.

Online resources:
https://www.scd.stfc.ac.uk/Pages/Shapespyer.aspx
https://gitlab.com/simnavi/shapespyer

Abstract

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Dynamics of proteins plays a central role for many biologically relevant processes on the molecular level. These include, for instance, protein folding, protein-ligand binding, enzymatic function or in general the properties of intrinsically disordered proteins. In my group at Forschungszentrum Jülich, we are using quasielastic neutron scattering (QENS) as well as high-resolution neutron spin-echo spectroscopy (NSE) to study the relevance of dynamics for the above mentioned scientific topics in selected model systems. One model system in the field of protein folding is apo-myoglobin that can be trapped in different folded conformations. Here, we used QENS and NSE to study the difference in molecular dynamics between the differently folded states. Another examples concerning protein-ligand binding were the cases of streptavidin and the ligand biotin as well as phosphoglycerate kinase (PGK) and the ligand bound PGK complex. Interestingly, ligand binding can either reduce the flexibility of the protein, which is the case for streptavidin, or increase it as we have observed it for PGK. From a thermodynamic perspective these changes in protein dynamics contribute substantially to the conformational stabilization of ligand binding. The relevance of conformational stabilization is pivotal for drug design on the molecular level but has been largely unexplored until now. The combination of QENS coupled with MD simulations could be a crucial development for the field of drug design in the coming future.

Computer Simulations for Muon Experiments: The Muon Spectroscopy Computational Project (MSCP)

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In this talk, I would like to present the Muon Spectroscopy Computational project to the wider simulation community working on simulations of neutron spectroscopy experiments The MSCP is a project that develops software and methods to help experimental muon scientists understand and better interpret their results with the help of state-of-the-art computational techniques.

The project is a joint collaboration of the UKRI Scientific Computing Department and the ISIS Muon Group, and we have a very close collaboration with other muon sources around the world.

A key point in a muon experiment is that, contrary to what happens in a neutron experiment, in a muon experiment the muon is implanted in the sample rather than diffracted by it. It is from this stopping site that the muon transmutes into a positron, which is emitted outside of the sample and detected.

We have, so far, developed three software tools:

1) pymuon-suite: which can be used to estimate the muon stopping site.
2) muspinsim: which used for studying the spin dynamics of a system of muon and other spins.
3) MuDirac: which can be used for modelling negative muon experiments. Specifically, the prediction of frequencies and probabilities of transition between energy levels of muonic atoms.

All of these tools are available as command-based tools, but we have also developed a web-based GUI for them called muongalaxy, which is an instance of the Galaxy platform: a web-based system for accessible, reproducible and transparent computational research.

Modelling structures of long RNA molecules by coarse-grained model comparing with scattering data.

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RNA molecules are not only messengers of genetic information from DNA to proteins, but are also actively involved in various intracellular regulations such as splicing, transcription, translation, heterochromatin formation and stress response, where the molecular tertiary structures play essential roles in the functions. Recent experimental studies have shown that long-stranded RNAs, such as mRNAs and lncRNAs, can form multiple patterns of secondary structures and transient tertiary interactions. For example, cryo-electron microscopy studies have directly observed mRNAs forming diverse secondary structure patterns. Various chemical probing techniques (e.g. DMS and SHAPE) have been developed, and in recent years the secondary structure of RNA in vivo has been detected at the single-molecule level. However, the identification of tertiary structures, which are not unambiguously defined, and higher order structures of RNA-RNA interactions are challenging to fully capture. We are developing a novel nucleotide-resolution coarse-grained model for RNA to simulate ensembles of RNA structures and discuss them in the context of cellular functions. In this presentation, I will show the current state of the model with a special focus on simulations of long (> 1000 nts) RNA molecules, where comparisons with scattering data (SAXS and SANS) are essential to validate and improve the model accuracy.

Structural ensembles of the 5'-UTR of hepatitis C virus RNA with and without microRNA

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The Hepatitis C Virus (HCV) genome accumulates in the liver due to a liver-specific microRNA called miR-122 [1]. miR-122 targets two sites in the 5'-untranslated region (UTR) of the HCV RNA to suppress an alternative secondary structure of the first 117 nucleotides, promoting the formation of a functional internal ribosome entry site that initiates cap-independent translation of the viral RNA [2]. However, structural characterisation of the HCV genome remains limited to short fragments or complexes bound to different cellular partners, from short fragment data [3]. Thus, details of how miR-122 interactions alter the structure of the viral 5'-UTR remain elusive. Here, we used molecular simulations to characterise the structures of different-sized fragments of the viral 5' terminus and their interactions with miR-122. The simulation data were compared with chemical probing and molecular scattering data for validation and further improvement of the model. Initial result verified the binding orders of miR-122 to the two predicted binding sites at the first 117 nucleotides of the 5'-UTR. We also observed alternative secondary structures [2]. Without other cellular partners, the free full-length HCV 5'-UTR has many different secondary structures, all different to the conventional secondary structure proposed based on available structural data [4]. Further investigations on miR-122 interactions with those alternative conformations are important to understand the roles of structure heterogeneity and additional binding sites on how microRNA modulate HCV RNA abundance in the liver.

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Thianthrene and nitrosonium tetrafluoroborate study

Rachel Rushworth

The reagents for the production of the thianthrene cation radical tetrafluoroborate were studied. These reagents were thianthrene and nitrosonium tetrafluoroborate. Thianthrene has undergone varies studies previously to study the geometric changes that accompany its redox activity. Various techniques were used to study both reagents, such as Renishaw-Raman spectroscopy, IR spectroscopy and inelastic neutron spectroscopy as well as DSC and high-resolution neutron diffraction.

Rationalising the vibrational spectra of pharmaceutical polymorphs of pyrimethanil as a possible in-line quality assurance tool using ab initio DFT calculations

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Pyrimethanil (PYL) has been identified as a suitable active pharmaceutical ingredient (API) for inhibiting methionine overproduction, a condition that can lead to degenerative neurological diseases. The suitability of PYL is hampered by its high vapour pressure and short effective lifetime.1 The pharmaceutical properties of PYL can be improved through the facilitation of polymorphic salts and cocrystals, maintained by energetically favourable supramolecular interactions, (i.e. hydrogen bonds and electrostatic interactions) with a non-toxic coformer in a new, synthon adduct. 2 Deciphering the nature of hierarchical supramolecular interactions between the API and coformer in the synthon is crucial for understanding how variations in important structural features can have a significant impact on understanding the properties of pharmaceutical solid forms.3 Herewith, a novel approach towards pharmaceutical polymorph analysis which marries X-ray crystallography, neutron scattering measurements, infrared (IR) and Raman vibrational spectroscopies and complementary discrete and periodic ab inito DFT calculations, is reported for a salt (PYL-LTA-MS) and cocrystal (PYL-LTA-CC) of PYL. X-ray diffraction revealed the overall crystal geometries within PYL-LTA-MS (monoclinic/C121) and PYL-LTA-CC (orthorhombic/P21212). A combination of X-ray and neutron diffraction studies also provided resolution towards synthon molecular recognition in the asymmetric unit of the two crystal systems, indicating a (i) N–H···O hydrogen bond and charge assisted N+–H···–O hydrogen bond, and a (ii) N–H···O and a single potential well N···H···O continuum for each material respectively. Using the unit cells obtained from diffraction studies, ab initio DFT calculations were performed in Gaussian and CASTEP to predict the infrared (IR) spectra,4 and produce phonon datasets for the materials, which were then validated using AbINS, and inelastic neutron scattering (INS).5,6 The complete vibrational assignments and the identification of hydrogen bonding modes for PYL-LTA-MS and PYL-LTA-CC were performed in terms of an “approximate description” of the normal modes using the phonon data to visualise the most relevant vibrational contribution to the spectral signature, using a combination of Jmol and Material studio visualising software.7,8 Ultimately, it is hoped that communication of this work will serve as a vehicle to demonstrates the potential of IR spectroscopy as a possible in-line quality assurance tool for implementing good manufacturing practise (GMP) routines to produce pharmaceutical materials.9

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Water and polymer side chain dynamics in hydrated PNIPAM microgels

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PNIPAM microgels are stimuli responsive materials, i.e. the particles undergo a sudden volume change at e.g. a critical temperature. This makes them highly interesting for various applications, such as drug delivery. This so called volume phase transition is ultimately linked to the atomic scale water-polymer interactions. Almost all previous neutron scattering experiments have been performed on samples in solution, which contains huge amount of bulk-like water. The few exceptions targeting samples with lower water content were carried out with backscattering- or spin-echo spectroscopy. Our Time-of-flight spectroscopy experiments (at Focus, PSI) on D_2O and H_2O hydrated samples, which access the fastest diffusional motions of polymer and water, complements those experiments very well. With the energy resolution of 50 microeV, we could observe two distinct polymer motions, namely methyl and side-chain rotations, from which only the later is influenced by the amount of water (at least for the low hydration cases). We started classical MD simulations in order to get more insights, especially on the side chain motion, because the experimental data is not sensitive enough to distinguish between different scenarios. The presence of protonated water results in two additional Lorentzians and suggest a dramatic suppression of long range diffusion, at least for the studied conditions of few water molecules per monomer (well below the percolation threshold). Definitely, samples with higher water content has to be investigated in the future, and simulations will be very valuable in order to study experimentally not available situations.

An experimental and theoretical study of the $^{3}He@C_{60}$ endofullerene.

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The $^{3}He@C_{60}$ endofullerene consists of a single ^{3}He atom entrapped inside a C_{60} fullerene cage. The confining potential, arising from the non-covalent interaction between the enclosed He atom and the C atoms of the cage, is investigated by inelastic neutron scattering. These measurements, performed on both PANTHER and TOSCA, allowed us to obtain information in both energy (ω) and momentum (Q) transfers in the form of the dynamical structure factor S (Q, ω). Simulations of the S (Q, ω) maps are performed for a spherical anharmonic oscillator model. Quantitative agreement between the experimental and simulated data sets is achieved. This talk aims to highlight the complementarity between the different types of instruments, direct and indirect geometry spectrometers in this case, in order to get a full picture and understanding of dynamics in confined systems.

Abstract

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MDANSE (Molecular Dynamics Analysis of Neutron Scattering Experiments) is a software package designed specifically to calculate neutron experiment observables from molecular dynamics trajectories. The main goal is to include instrument-specific parameters such as energy resolution and Q-coverage in the analysis, while using the relevant parameters such as neutron scattering lengths of chemical elements as the weighting factors. This way the simulation results can be compared directly with the experimental data sets.

Currently MDANSE is being re-written in order to cover the needs of a wider community of users. The changes are aimed at making the code run both in a modern graphical desktop and in purely text-console based environments. The new code structure allows MDANSE to be used as a library of calculations usable in Python scripts. At the same time, a graphical interface is provided for interactive sessions and visualisation of results.

User participation is particularly welcome at this stage of the software development, since the programming effort behind different features of MDANSE can still be relocated. This presentation is aimed at informing the user community about the current direction of the development, allowing the users to express their expectations and wishes for the new release of MDANSE.

Simulation of Fast Neutrons Thermalization in ^{11}B based Oxide Glasses

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Delivering energy from nuclear fusion may be the greatest technical challenge of the twenty-first century. Monitoring neutron emission profiles and fuel circulation requires advanced materials and sensors that can withstand the harsh environment of a fusion reactor. Glass scintillators may be suitable for these applications. They offer a range of advantages in comparison to other types of scintillators, such as a high stability, radiation hardness, ease of manufacture even at large sizes and unusual geometry. Commonly used glass scintillators are most suitable for use for thermal neutron detection and spectroscopy, rather than the fast neutrons typical of fusion. In response to this, this work presents a numerical study that investigates the efficiency of potential glass scintillators to be used in fusion reactor environments where 14.1 MeV neutron are prominent. The results of particle transport modelling are provided which considered a series glass compositions enriched in ^{11}B that were interrogated monoenergetic neutron beam. The measurement of the thermalization distance of neutrons in each sample is reported and compared to find the optimal glass composition.

DANAI - A novel syntax to quantify atomic interactions.

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The DL_ANALYSER Notation for Atomic Interactions**,** DANAI, is a notation syntax to describe interactions between atoms and molecules [1]. This notation can annotate precisely the detailed atomistic interactions without having to resolve to diagrammatic illustrations, and yet can be interpreted easily by both human users and computational means. The Notation directly encodes a universal chemical-sensitive information [2], thus making atomic interactions to become data discoverable and searchable and is useful for cheminformatic applications and comparative studies with other molecular systems. By making a reference to molecular simulations of HFA134a, a liquid propellant, as an example case [3], DANAI provides localized interactions, 'at a glance', of the liquid model on otherwise a typically disordered system consists of complex network of intermolecular interactions. From such, statistical analysis such as the correlation coefficients can be carried out, to assess the behaviour of various modes of interactions identified within the system.

DANAI is implemented in DL_ANALYSER [4], a general analysis software program for DL_POLY molecular dynamics simulation software.

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