



# Book of abstracts

Erquy, 9-12 oct. 2023 JOURNÉES DE LA DIFFUSION NEUTRONIQUE Multidisciplinary science with neutrons

This **multidisciplinary scientific conference** JDN2023 will present new results, obtained by neutron scattering, on topics of interest to the entire scientific community. Thematic sessions will be organised in the respective fields of **Condensed Matter**, **Biology**, **Instrumentation**, **Magnetism**, and **Soft Matter**.

Each of these sessions will begin with an invited oral plenary talk (40 min) and a keynote talk (30 min). It will be followed by several oral communications.

In addition, sessions will be dedicated to **News from the Facilities**, **Neutron Scattering Associations**, and the **SFN PhD award ceremony**.

A special half-day session will be dedicated to **Compact Sources** including oral presentations and a round-table on Thursday morning. Two poster sessions covering all the scientific themes will also be organised on Monday and Tuesday afternoon.

Plenary sessions will be held in the main conference room (*La Rigadelle*). An additional room (*Le Grand Bleu*) will be used for parallel sessions.

After some free time, a musical session accompanied by an aperitif will be organized on Tuesday afternoon.



The organizing committee of JDN2023 is pleased to welcome you at Roz Armor in Erquy, and wish you a pleasant stay and very fruitful scientific discussions.

# Monday 9 October

12:30 – Registration & Buffet

14:30 - 14:50 Opening

# **Condensed Matter**

Chair: Carmelo Prestipino

14:50 - 15:30 Plenary - Jacky Even (Institut FOTON, Rennes)

Complex lattice dynamics in 3D perovskites for optoelectronic applications

15:30 – 16 :00 Keynote - Claire Villevieille (LEPMI, Grenoble)

Neutrons as key technique for a better understanding of lithium and postlithium batteries

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16:00 – 16 :30 Coffee break

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#### Chair: Stéphane Rols

16:30 – 16:45 Peter Fouquet (ILL, Grenoble)

Hydrogen mobility and reactivity in MoS<sub>2</sub> catalyst

16:45 –17:00 Mohammad Abdelsater (LPS, Orsay)

Hydrogen Storage and Dynamics in Clay Materials

# Biology p 11 Chair: Francesca Natali

16:30 - 16:45 Christian Beck (ILL, Grenoble)

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

16:45 –17:00 Christelle Saade (LLB, Saclay)

Structure of mTSPO membrane protein in different amphiphilic environments

# Session News from Facilities

#### Chair: Denis Morineau

17:00 – 17:20 European Spallation Source – Helmut Schober (ESS, Lund, Sweden)

- 17:20 17:40 Institut Laue Langevin Jacques Jestin (ILL, Grenoble)
- 17:40 –18:00 Laboratoire Léon Brillouin Arnaud Desmedt (LLB, Saclay)
- 18:00 18:20 Fédération Française de la Neutronique Fabrice Cousin (2FDN)

#### 18:30 – 19:30 Cocktail – Posters

19:30 – Dinner

# **Tuesday 10 October**

# Biology

## Chair: Sylvain Prévost

9:00 – 9:40 Plenary – Philippe M. Oger (INSA de Lyon)

Adaptation to the extreme in polyextremophilic archaea

9:40 – 10:10 Keynote - Martin Weik (IBS, Grenoble)

Dynamics of amyloid fibers and their hydration water as studied by neutron spectroscopy

# Instrumentation

Chair: Emmanuelle Suard

10:10 - 10:30 Alessandro Tengattini (ILL, Grenoble)

NeXT 2.0, the Neutron and X-Ray Tomograph at ILL

10:10 - 10:50 Stanislav Savvin (Instituto de Nanociencia y Materiales de Aragon, Zaragoza)

XtremeD: new single crystal and powder diffractometer for studies under extreme conditions

10:50 – 11:20 Coffee break

#### Chair: Frédéric Ott

11:20 - 11:40 Peter Falus (ILL, Grenoble)

Latest results from WASP the widest angle neutron spin echo spectrometer

11:40 – 12:00 Frédéric Bourdarot (CEA-Grenoble)

New ZETA+ on IN22

12:00 - 12:15 Martin Boehm (ILL, Grenoble)

Physics Informed Steering of Triple-Axis Spectrometers

12:15 - 12:30 Francesca Natali (CNR-IOM & ILL, Grenoble)

IN13+: more flux, more science

12:30 – 14:00 Lunch

# Posters

14:00 – 15:30 Posters

16:00 – 18:30 Free time

18:30 – 19:30 Aperitif – Musical performance

19:30 – Dinner

# p 11

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# Wednesday 11 October

# Magnetism

## Chair: Virginie Simonet

9:00 – 9:40 Plenary – Françoise Damay (LLB, Saclay)

Competing interactions in Dysprosium garnets

9:40 – 10:10 Keynote - Victor Balédent (LPS, Orsay)

Tuning properties in multiferroic with pressure and magnetic field

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# Magnetism

#### Chair: Dalila Bounoua

10:15 – 10:30 Ketty Beauvois (CEA, Grenoble)

Multi-k or single-k magnetic order in the frustrated spinel compounds GeNi<sub>2</sub>O<sub>4</sub> and GeCo<sub>2</sub>O<sub>4</sub>?

#### 10:30-10:45 Isabelle Mirebeau (LLB, Saclay)

*High Spin-Low spin transition inducing phase separation in a helical magnet* 

10:45 –11:00 **Manila Songvilay** (Institut Néel, Grenoble)

Unusual magnetic ground state of the cobalt honeycomb magnet BaCo<sub>2</sub>(AsO<sub>4</sub>)<sub>2</sub>

# Soft Matter p 31

#### **Chair: Fabrice Cousin**

10:15 – 10:30 Olivier Diat (ICSM, Marcoule)

Stabilization of a liquid foam by ionic charges: simultaneous multiscale study in Fourier space and real space

10:30 –10:45 Marie Plazanet (LIPhy, Grenoble)

Insights into the microscopic mechanisms responsible for phase separation in a biphasic acidic solution for metallic ion extraction

10:45 –11:00 **Swen Helstroffer** (ICS, Strasbourg)

Probing the emergence of electrostatic attraction between like-charged phospholipid membranes

#### 11:00 – 11:30 Coffee break

# SFN PhD award ceremony

- 11:30 11:35 Denis Morineau (IPR, Rennes) Introduction
- 11:35 12:00 Virginie Simonet (Institut Néel, Grenoble)

Spin texture in a chiral magnet: experiment and theory

- 12:00 12:05 President of Jury Presentation of the 2023 SFN PhD award
- 12:05 12:30 SFN PhD Award Laureate

12:30 - 14:00 Lunch

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# Soft Matter

## Chair: Christiane Alba-Simionesco

#### 14:00 – 14:40 Plenary – Natalie Malikova (PHENIX, Paris)

*Polyelectrolyte-based hydrogels: tuning structure and properties by ion specific effects and addition of charged anisotropic nanoparticles* 

#### 14:40 – 15:10 Keynote – Diane Rebiscoul (ICSM, Marcoule)

From nanoporous material evolution in solution to water behavior in confinement

Soft Matter	p 31	Magnetism	p 21		
Chair: Marie Plazanet		Chair: Ketty Beauvois			
15:15 – 15:30 <b>Jérôme Combet</b> (I Strasbourg) Light responsive wormlike micelle	CS, es	15:15 – 15:30 <b>Olivier Demortier</b> (LLB, Saclay) Spin dynamics and Thermal conductivity in KTb <sub>3</sub> F <sub>10</sub>			
15:30–15:45 Manon Pépin (LLB,	Saclay) nd rheology of	15:30 –15:45 <b>Soufiane El Bidaoui</b> (Institut Néel, Grenoble)			
a fluid allowing self-consistent subduction during convection in the laboratory 15:45 –16:00 <b>Coralie Pasquier</b> (ICSM, Marcoule)		Investigation of the remarkable magnetic behavior of itinerant electrons in (Sc,Nb)Fe2			
		15:45 –16:00 <b>Antoine Roll</b> (LPS & LLB, Saclay)			
COSAN nano-ion as novel solubilization agent of hydrophobic compounds		Multipolar interactions and spin dynamics in Tb <sub>2</sub> Ti <sub>2</sub> O <sub>7</sub>			
16:00 – 16:30 Coffee break					

# Neutron Scattering Associations

16:30 – 16:45 European Neutron Scattering Association - Natalie Malikova (ENSA)

16:40 – 18:30 General assembly of the French Neutron Scattering Association – **Pascale Launois** (SFN) Annual report and financial statement, election of the new board members

19:30 – Dinner

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# Thursday 12 October

# **Compact sources**

9:00 – 9:20 Frédéric Ott (LLB, Saclay)

High Current Accelerator-driven Neutron Sources - State of the art

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9:20 - 9:40 Mariano Paulin (LLB, Saclay)

The HERMES - LLB outstation at the JULIC Neutron Platform

9:40 – 10:00 Xavier Fabrèges (LLB, Saclay)

Diffraction around HiCANS : example of the PRESTO instrument

10:00 - 10:20 Quentin Faure (LLB, Saclay)

Spectroscopie : Etat de l'art et nouvelles opportunités

10:20 - 10:40 Marie Plazanet (LIPhy, Grenoble)

The ICONE Conceptual Design Report

10:40 - 11:00 Félix Fernandez-Alonso (UPV/EHU, Donostia - San Sebastian)

The IKUR-FUN initiative in the Basque Country – Raison d'être and first steps

11:00 – 11:30 Coffee break

11:30 – 12:30 Round table

12:30 - 14:00 Lunch

# End of JDN2023

## **Complex lattice dynamics in 3D perovskites for optoelectronic applications**

<u>J. Even</u><sup>1</sup> <sup>1</sup> Institut FOTON CNRS - INSA Rennes (France)

3D halide perovskites used for low-cost solar cells or light emitters, exhibit a complex lattice dynamics, which is not accounted for in nowadays descriptions of phonons and electron-phonon coupling. We combined several experimental spectroscopy techniques, including inelastic neutron scattering, to investigate low frequency lattice vibrations and relaxations in 3D hybrid halide perovskites. The extensive disorder of the lattice at normal optoelectronic device operation temperatures is similar to pre-melting where only long-range acoustic normal modes survive, while diffraction patterns exhibit extensive diffuse scattering contributions. Until now, electronic structure calculations for cubic halide perovskites are performed by assuming that the potential energy felt by electrons is described with the nuclei clamped at high symmetry lattice sites. This assumption inevitably misses the effect of polymorphism and anharmonicity which are ubiquitous in this class of materials. To address this issue, we developed recently a new computational approach for first-principles lattice dynamics, electron dispersions and electron-lattice coupling in such strongly anharmonic materials at finite temperatures.

Acknowledgments The research leading to these results has received funding from the European Union's Horizon 2020 program, through a FET Open research and innovation action under the grant agreement No 899141. (POLLOC), and under the Marie Sklodowska-Curie grant agreement No. 899546.

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# Neutrons as key technique for a better understanding of lithium and postlithium batteries

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Li-ion batteries are key players for the energy transition and help decrease greenhouse emission owing to their storage ability. But batteries are also very complex electrochemical systems that deserve an in depth characterisations to understand their limitation during cycling. Indeed, batteries are constantly under further development and employing new type of chemistry and materials that needs to be investigated during operation i.e., in operando mode.

Neutron powder diffraction (NPD) is a technique of choice to investigate structural changes, especially for light elements like lithium. Developing an electrochemical cell dedicated to operando neutron diffraction measurements is challenging due to the large amount of electroactive materials needed and due to the incoherent neutron scattering with hydrogen, highly contributing to the background.

We report here an optimal cylindrical cell based on 18650 design<sub>1-3</sub>: i) the casing is made of transparent TiZr alloy<sub>4</sub> and ii) several grams of active materials per electrodes. Additionally, we modified several set-up on D19 neutron diffraction beamline at ILL enabling the investigation of the electrochemical cell during relaxation processes giving information about diffusion. NCM811 vs. graphite was selected as promising full-cell battery due to the high energy density expected with this electrode couple. Figure 1 represents the neutron diffraction patterns evolution of both phases along the first (de)lithiation. As can be seen both the stages in graphite lithiation (peak (002)) and the insertion reaction of NCM811 (peak (003)) can be easily followed and analysed by mean of Rietveld refinement.



Figure 1. Contour plot representation of the neutron patterns collected during the operando experiment.

In another example, we used neutron imaging to follow the Li diffusion processes by using Li isotopes <sup>6</sup>Li and <sup>7</sup>Li. By monitoring fast charge behaviour in Li-rich cathode materials vs. Li metal, we demonstrated that the electrode engineering is a key parameter to optimise to allow fast charging protocol. Coupling experimental data to modelling reveals the hindrance of several Li transport pathway for electrocoupling fast charge and high-power features. Similar examples will be presented on solid state batteries.

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## Hydrogen mobility and reactivity in MoS<sub>2</sub> catalyst

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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<sup>(+)</sup>, H<sub>2</sub>, H<sub>2</sub>O) to the chemistry in MoS<sub>2</sub>.



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# Hydrogen Storage and Dynamics in Clay Materials

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Clay minerals, with their exceptional properties such as low cost, non-toxicity, and stability, have been extensively studied for their applications in various domains, including industrial sectors. However, their potential as hydrogen storage materials remains largely unexplored.

In this context, the present work aims to investigate hydrogen adsorption in pre-laponite, which is A material obtained at intermediate stage during the synthesis of laponite, a synthetic clay mineral, Indeed, we also show that pre-laponite has a significantly higher hydrogen sorption capacity compared to laponite.

We analyzed its structure using X-ray scattering technique and infra-red spectroscopy. Our results show that the local structure of pre-laponite is similar to that of laponite, although the long-distance order is significantly reduced.

Inelastic neutron scattering (INS) experiment was performed on pre-laponite using the spectrometer PANTHER at Institut Laue Langevin (ILL), injecting 1 bar of H2 on a pre-laponite sample at 77K and cooling down the system. The J01 peak, corresponding the transition of H2 from a singlet para-(molecular spin S = 0, angular momentum J = 0) state to a triplet ortho- (S = 1, J = 1) state exhibits a fine structure at 41K. It is fitted by two gauassian lines with intensity ratio of 2:1. Detailed analysis shows that the molecules are parallel to the adsorption wall and that the barrier height of the potential associated with their rotation is 52K. At lower temperatures, a third -non splitted- Gaussian component appears, that may be ascribed to hydrogen adsorption on a second layer (fig. 1a)

A diffraction study was also performed using the spectrometer D16 at ILL. Strong structuration peaks appears for  $D_2$  in a Q range compatible with intermolecular distances (blue arrows in fig. 1b), the fine structure of which is observed to be dependent on the morphology of the host (pre-laponite/laponite). The diagram associated to H<sub>2</sub> does not exhibits the same features. This difference suggests that quantum effects may play a role in the adsorption of the molecules between pre-laponite layers. Finally, we will discuss in a complementary manner the elastic and inelastic neutron scattering results.



Figure 1. (a) H<sub>2</sub> adsorption in pre-laponite: INS spectrum at 21K fitted by two Gaussian 1 and 1' characteristic of the adsorption site evidenced at 41K and by a Gaussian 2 which may correspond to a second adsorbed layer of H<sub>2</sub>. (b) Elastic scattering spectra of 1 bar gas (H2 / D2) injected in the host matrix (pre-laponite / laponite), after subtraction of the contributions from the (pre-laponite/laponite), from the cell and from the cryostat.

#### Adaptation to the extreme in polyextremophilic archaea

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Archaea are known to inhabit some of the most extreme environments on Earth. We are interesting to decipher the adaptation of archaea from deep-sea hydrothermal vents, which experience multiple environmental stresses, such as high hydrostatic pressure (HHP), high temperature, high salinity, high reduced metals... HHP has numerous deleterious effects on cellular components. HHP is however required for optimal activity of deep environment adapted microbes (piezophiles). At the same time, HHP is inhibiting cellular functions in piezosensitive organisms. We are interested in deciphering the structural differences between the two types of organisms. Our work is focused on the two cellular compartments which are most impacted by HHP and HT, the cell membrane, and the proteome. We use tools from biology, chemistry, and physics to explore the physics of the adaptation.

Adaptation of the Membrane: The ability of archaea to thrive at HT is proposed to be linked to the ability of lipids to form monolayer membranes. We have studied how archaea which have a membrane bilayer can grow at temperatures above 100°C. We have proposed that the stability of the membrane could be due to the presence of apolar polyisoprenoids at the midplane of the bilayer [1]. The presence of apolar lipids is thought to reduce the membrane permeability to ions and to increase membrane rigidity, which therefore could explain the extreme temperature and pressure resistance of the membrane of these extremophiles. Using neutron diffraction, spin echo, HP NMR and FT-IR, have demonstrated the validity of this novel membrane architecture to both HT and HHP, and several archaeal lipid mixtures [2-5]. Interestingly, diffraction data indicate the stable coexistence of different fluid lamellar and non lamellar phases at HT and HHP. Increasing the amount of squalane in the membrane was found to regulate the phase diagram and lead to an increase in the lamellar repeat spacing. The ability of squalane to regulate the ultrastructure of an archaea-like membrane at high pressure and temperature supports the hypothesis that archaea can use apolar lipids as an adaptive mechanism to extreme conditions. The wide range of P/T conditions at which phases coexist confirms that apolar lipids could be important for domain formation in archaea living under extreme conditions.

Adaptation of the proteome: Proteome adaptation to high pressure in Archaea is still an open debate. Genomic studies were not able to determine a clear adaptation pattern and HP adaptation is often considered as a crossover adaptation, that is, a concomitant process with the adaptation to temperature. Our studies on whole cells highlighted the differences in proteome dynamics between *T. barophilus* (Tba) and *T. kodakarensis* (Tko), two closely related species which grow at the same temperature (85°C) but differ in optimum pressure (400 bar for Tba, 1 bar for Tko) [6,7]. To refine our molecular model of adaptation, we have performed EINS and QENS in conjunction with High resolution NMR to unravel the dynamics of 2 proteins from the two organisms. This novel approach permits to characterize in detail the dynamics of the two proteins without the complications of a whole-cell environment. Our results indicate that the piezophilic protein's dynamics are more stable on a time scale of 10-100 ps, and that large side-chain motions are even favored by HP [8]. In contrast, the piezosensitive protein is destabilized by HP, becomes more rigid and its structure is likely affected. The detailed NMR atomic maps allowed to draw a very detailed model of the adaptation and the underlying physico-chemical drivers of evolution.

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# Dynamics of amyloid fibers and their hydration water as studied by neutron spectroscopy

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Protein amyloid fiber formation is the pathological hallmark in various neurodegenerative diseases such as Parkinson's and Alzheimer's. The physico-chemical origin of protein fibrilation, as well as the role that hydration-water might play remain elusive. We combine elastic and quasi-elastic neutron spectroscopy and molecular dynamics simulations on the intrinsically disordered proteins  $\alpha$ -synuclein (involved in Parkinson disease) and tau (involved in Alzheimer disease) to investigate both structural and dynamical properties of the protein-hydration water system. One of our findings is an increased water translational diffusion on fiber surfaces, suggesting that hydration-water entropy might be one of the driving forces for amyloid fiber formation.

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

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Protein dynamics have been investigated on a wide range of time scales. Nano- and picosecond dynamics have been assigned to local fluctuations, while slower dynamics have been attributed to larger conformational changes. By investigating the heat shock protein 90 (Hsp90), we can assign molecule-spanning dynamics on the 100 ns to 200 ns time scale to dynamic modes on this time scale. To obtain a comprehensive picture of these global real-space movements (Figure 1), we apply a combination of 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].

The dynamic modes of Hsp90, affected to various degrees by a co-chaperone Sba1, and its time scales depend on the conformational state of the Hsp90 dimer. Altogether, our data is best described by fast molecule-spanning dynamics, which precede larger conformational changes in Hsp90 and might be the molecular basis for allostery. Our integrative approach provides comprehensive insights into molecule-spanning dynamics on the nanosecond time scale for a multi-domain protein.



<u>Figure 1</u>: Representation of the molecule spanning dynamics. The different time-scales as well as the corresponding techniques used for their investigation are displayed in the different boxes [1].

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## Structure of mTSPO membrane protein in different amphiphilic environments

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TSPO (TranSlocator PrOtein) is a functionally important and ubiquitous transmembrane protein, mainly localized in the outer mitochondrial membrane. TSPO is of very strong pharmacological interest, thanks to its ability to bind ligands, used as markers for imaging and therapeutics in many inflammatory, cancerous, and neurodegenerative diseases. However, no crystals of mammalian TSPO have been obtained so far, and therefore a high-resolution structure remains challenging especially in absence of ligands [1].

mTSPO in SDS

mTSPO in DPC



**Figure 1.** SANS ab initio envelopes of mTSPO (without ligand) in SDS and DPC detergent (blue and green envelopes, respectively), using the DENFERT software [5] compared to the 20 ligand-binding conformers obtained by liquid NMR (2MGY.PDB).

We are presently studying mTSPO in more biomimetic environments to optimize a "native" refolding of the protein: in DPC/DMPC mixtures, where mTSPO recovers nM affinity [6], and in phospholipid nanodiscs. We aim to get structural information and to determine the optimal conditions where the protein is stable and functional for further high-resolution studies by the classical structural methods (radiocrystallography, NMR, cryo-EM). Understanding the structure/function of mTSPO is indeed crucial to develop new pharmacological molecules for diagnostics and therapeutics.

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# NeXT 2.0, the Neutron and X-Ray Tomograph at ILL

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NeXT-Grenoble is the Neutron and x-ray tomograph born in 2015 from a collaboration between the Institut Laue-Langevin (ILL) and University Grenoble Alpes (UGA). This instrument has undergone a major upgrade to further expand the portfolio of contrast options. This is the result of a collaboration between the initial partners (UGA and ILL) plus the Helmholtz-Zentrum Berlin (HZB), also via the newly founded international mixed research unit NI-Matters. This upgrade also adds MoTo, a Monochromatic Tomograph designed with grating interferometry and polarized neutron imaging in mind.

The upgrade has improved the highest attainable spatio-temporal resolutions by increasing the maximum flux (expanding the accessible collimation ratios L/D) as well as by upgrading the range of detectors. The simultaneous x-ray imaging has also been improved to explore a broader range of geometrical configurations. An improved sample stack helps automate and expands the possibilities (in size/weight) of in-situ apparatus that can be easily installed on the instrument, as well as adding a laminography option.

A number of new contrast options have also been added: a velocity selector as well as a double crystal monochromator provide versatile energy selection. A grating interferometer allows the characterization of heterogeneities on the scale of 0.1  $\mu$ m to 10  $\mu$ m and above through dark-field imaging, while differential phase contrast can be employed to differentiate even modest variations in the refractive index. The new instrument also has a native integration of neutron polarization equipment in order to perform vectorial tomographies of magnetic fields.

This presentation will detail the upgrade of NeXT and of MoTo and outline the ongoing commissioning and early results from these new options.



Figure 1. A schematic representation of the upgraded instrument.

# **XtremeD: new single crystal and powder diffractometer for studies under extreme conditions**

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The ability to study materials using neutron powder and single crystal diffraction under extreme conditions such as temperature, pressure and magnetic field is becoming increasingly important [1]. Neutrons provide unparalleled opportunities when it comes to locating light elements, telling apart in the crystal structure chemical elements – neighbours in the periodic table, studying magnetic ordering phenomena, etc. The ability to exploit the above properties of the neutron under extreme conditions, where interatomic interactions and coordination are significantly altered by external stimuli, opens up unique possibilities to explore structure-property relationships in a wide range of conventional materials as well as look into more exotic properties and structures, unreachable under ambient conditions.

XtremeD is a new Collaborative Research Group (CRG) instrument recently built to this end at the Institut Laue-Langevin, Grenoble, France and operated by the Spanish National Research Council (CSIC). Because the amount of sample will necessarily be small, the instrument was designed to maximize neutron flux and improve neutron detection efficiency through the combined use of variable beam focussing on the sample and the last generation <sup>3</sup>He multi-wire 2D detector (Fig. 1), respectively. A wide range of the incident beam wavelengths (0.9 - 4 Å) produced either by a HOPG or Si monochromator will be available. Depending on the instrument resolution needed for each experiment, the monochromator take-off angle can be varied from 40° suitable for magnetism to 120° suitable for the crystal structure determination. As it stands, XtremeD can host the majority of the ILL sample environment equipment with particular focus on high-pressure cells (currently up to 30GPa) and magnetic fields (currently up to 15T) both in the powder and single-crystal modes.

This talk will present and discuss the design and first results obtained on XtremeD during the hot commissioning phase.



**Fig. 1.** Photo of XtremeD in its low take-off configuration hosting an ambient powder stage [1] J. A. Rodríguez Velamazán et al, Journal of Physics: Conference Series, 325 (2011) 012010.

## Latest results from WASP the widest angle neutron spin echo spectrometer

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Neutron Spin Echo (NSE) instruments are the highest energy resolution neutron spectroscopy instruments. WASP the Wide Angle Spin echo is the youngest member of this family. WASP's magnetic configuration is unique, it is the single functioning instrument of its kind. After four cycles of user operation the first results were published. 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. Hopefully these examples will inspire you to attend the HIRES workshop at ILL to explore how Neutron Spin Echo can be used in synergy with other techniques.

## "New ZETA<sup>+</sup> on IN22"

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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 [1-3].

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  $\approx 0$  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<sup>-5</sup>. 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.

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#### **Physics Informed Steering of Triple-Axis Spectrometers**

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We investigate new concepts for enhancing the data acquisition efficiency of scanning type instruments exploring a multidimensional feature space. We compare machine-learning algorithms and probabilistic methods in order to minimize the number of experimental data points, which are required either to reconstruct an intensity landscape or to directly determine models and model parameters down to precisions defined by the scientists. Data acquisition, interpretation and modeling can become part of a closed loop, typical of the concept of Autonomous Experimentation [1]. This approach is opposite to data-driven methods [1], which try to maximize the amount of experimental data and to extract data patterns and physical information with the help of neural networks or similar AI techniques.

Taking classic triple-axis spectrometers (TAS) as representative for scanning type instruments, we test various algorithms on TAS to explore a scattering function  $S(Q, \omega)$  in the four-dimensional feature space, spanned by the momentum space, Q, and the neutron energy transfer axis,  $\hbar\omega$ . The scanning can be agnostic, meaning that no physical information on the scattering system is available, or it can be domain informed, i.e. some prior physical knowledge assists the steering process to find relevant positions in the feature space. Gaussian process (GP) regression [2] and log-GPR [3] are examples of agnostic scanning. Very recently we tested physics instructed GPs with a mean function given by the maximum likelihood estimate (MLE), similar to recent x-ray scanning experiments [4], and a parametric Bayesian approach, where initial estimates on relatively simple spin-wave models were gradually improved with every incoming data point. We decided on several metrics in order to compare between the different models and, based on this analysis, discuss their respective strengths and weaknesses of the algorithms. The general high potential of domain informed algorithms opens very interesting perspectives for efficient data steering processes in future, if dedicated decision policies are developed based on available theoretical knowledge.

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#### IN13+: more flux, more science

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IN13 is a backscattering spectrometer working with thermal neutrons with wavelength 2.23 Å corresponding to energy of 16.45 meV. The energy resolution is of the order of 8  $\mu$ eV and the energy window is  $\pm$  100  $\mu$ eV. In addition the value of the incident energy gives the availability of high momentum transfers (Q < 4.9 Å-1), much larger than obtainable with currently worldwide available spectrometers. The instrument fills the energy gap between IN6-SHARP or IN16B and IN5 at ILL, IRIS and OSIRIS at the Rutherford-Appleton laboratory (RAL) in United-Kingdom, BASIS at ORNL or HFBS-NCNR at NIST in the US, and DNA at J-Parc in Japan.

The major drawback of IN13 being the low neutron flux on the sample, implying long acquisition times that put limits on the experimental data obtainable and requires important sample masses, a major upgrade was undertaken during the last 2 years. This included the guide refurbishment, the design of a new monochromator, a new deflector and further additional minor improvements.

The instrument has been dismounted in late 2021 and recently remounted. Preliminary test were done before summer and first official experiments are planned from September on. I will report here the concept of the upgrade and very first results.

#### **Competing interactions in Dysprosium garnets**

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Amongst geometrically frustrated architectures is the hyperkagome lattice, which consists in a twisted spatial arrangement of corner-sharing triangles and which can be found in rare-earth (R) garnets  $R_3B_2C_3O_{12}$  [1]. Mean-field calculations on a hyperkagome lattice, for an effective anisotropic pseudospin  $S = \frac{1}{2}$  (characterized by general  $g_{xx}$ ,  $g_{yy}$ , and  $g_{zz}$  Landé factors), generate a very rich magnetic phase diagram, encompassing complex phases, including disordered ones, when magnetic anisotropy departs from the strong Ising case [2]. Tuning the rare-earth magnetic anisotropy in garnets is the key to exploring this phase diagram, and can be achieved through appropriate distortions of the rare-earth oxygen environment, the latter being controlled by the B and C ionic size.



Figure 1: (a) Variations, with the surrounding oxygen ( $x_0$ ,  $y_0$ ) coordinates, of the energy (in meV) of the first crystal electric field (CEF) excitation for a Dy<sup>3+</sup> in its dodecahedron. The boundaries delimiting the different Ising phases are marked by contour lines. (b) Enlargement of (a) around a set of experimental data displayed as pink squares (the higher the energy of the first CEF excitation, the stronger the Ising anisotropy).

In dysprosium garnets (R = Dy), our neutron scattering results show that the Ising character of  $Dy^{3+}$  is weakened by larger ions on the C site (Figure 1), but even in  $Dy_3In_2Ga_3O_{12}$ , where dipolar interactions should be the weakest because of the large Dy-Dy distance, multiaxial ( $\mathbf{k} = 0$ ) magnetic ordering is still observed around  $T_N$ = 1.1 K [3].  $Dy_3Ga_5O_{12}$  remains somewhat of a mystery, as it is the only Dy garnet with a subkelvin  $T_N$  (360 mK). An extensive investigation of the magnetic phase diagram and magnetic excitations of  $Dy_3Ga_5O_{12}$  will be presented, allowing one to understand the unique interplay between magnetic exchange, dipolar interaction and XY anisotropy specific to this compound.

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# Tuning properties in multiferroic with pressure and magnetic field

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Systems containing strongly correlated electrons exhibit a wide range of extraordinary characteristics. It is the presence of these correlations that enables phenomena such as the transition from insulating to metallic behavior or the emergence of superconductivity. In numerous instances, multiple of these unique properties can either coexist or compete within the same or neighboring phases. When two distinct orders are present in such proximity, they exert an influence on each other. This intricate interplay often leads to outcomes that transcend a simple combination of the two properties, opening up possibilities for novel and remarkable characteristics. This is exemplified in multiferroic compounds, where new connections emerge, allowing the manipulation of properties through external parameters.

I will present the RMn2O5 family of manganites (with R representing rare earth) as an example. These materials exhibit both magnetic order and ferroelectricity. First, I will clarify the source of the connection between magnetism and electricity : the magneto-electric coupling. Then, I will demonstrate how we can adjust their characteristics by applying pressure or a magnetic field, and I will suggest the fundamental mechanisms at play. Throughout these findings, we will observe that neutron scattering plays a central role, whether in investigations related to temperature, pressure, or magnetic field effects.



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# Multi-k or single-k magnetic order in the frustrated spinel compounds GeNi<sub>2</sub>O<sub>4</sub> and GeCo<sub>2</sub>O<sub>4</sub>?

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Non colinear magnetic structures, such as skyrmions, hedgehogs, receive a renewed interest for spintronic applications [1]. An ingredient for the realization of these complex structures is magnetic frustration. It has been shown recently that they are stabilized in some spinel compounds, of generic formula  $AB_2O_4$  [2,3]. These materials can accommodate a magnetic atom on the B sites, which form a pyrochlore lattice. The Ge-spinel compounds, with B=Co, Ni, Fe, are particularly interesting as their complex ordering is expected to arise from competing interactions up to the sixth neighbors [4]. The Co and Ni members of the family order magnetically with a k = (1/2, 1/2, 1/2) propagation vector. In a single-k picture, the magnetic structure can be described as alternating kagome and triangular ferromagnetic planes, with the spins on the kagome planes perpendicular to the <111> directions and the spins on the triangular planes also perpendicular to <111> for Co and either perpendicular or along <111> for Ni [4, 5,6,7], and planes of the same kind are antiferromagnetically coupled. However, theoreticians recently found a non-coplanar triple-k configuration in zero magnetic field [8]. Starting measurements, spin-wave modeling and single-crystal neutron diffraction data that a 2-k flattened hedgehog structure may be stabilized for the Ni compound. For the Co compound, a structural distortion associated to the magnetic transition allows the single-k structure.

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#### High Spin-Low spin transition inducing phase separation in a helical magnet

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MnGe, obtained from high temperature, high pressure (HT, HP) synthesis, is a helical magnet of the B20 cubic structure, with short helical wavelength  $\lambda_h = 29$  Å, close to 6 unit cells. It shows exotic features such as pressure induced High Spin-Low Spin (HS-LS) transitions, invar-like behaviour, and giant topological Hall effect among others [1]. Mn substitution for 3d or 4d atoms (Co, Fe or Rh) leads to helical structures with very long periods (up to 4000 Å) [2], with skyrmions phases under applied magnetic field, as for MnSi.

*What about Ru*? We performed SANS, magnetization, and X-ray diffraction measurements in  $Mn_{1-x}Ru_x$ Ge. Surprisingly, here the helical wavelength remains very short and almost independent of x (Fig), whereas no skyrmion phases are stabilized by a magnetic field. Quantitative analysis of the SANS data shows that the helical moment undergoes a step-like decrease from a HS state of  $1.8(1)\mu_B/f.u$ . to a LS state of  $0.4(1)\mu_B/f.u$ . The helical coherence length decreases from 400 Å to about 100 Å in Ru-rich samples, but despite this magnetic disorder, the periodic structure persists up to x=0.90 at least. The variation a(x) of the lattice constant does not obey the Vegard law. More importantly, a phase separation occurs in the region of the HS-LS transition (0.2 < x < 0.5), involving two B20 phases with different Ru contents.



Evolution of the helical wavelength in  $Mn_{1-x}Ru_xGe$  with Ru concentration x. a) compared with Ru concentration of the lattice constant a. b) compared with  $Mn_{1-x}tM_xGe$  systems where tM is a transition metal (Fe, Co or Rh).

*Why is*  $Mn_{1-x}Ru_xGe$  so different? We suggest what follows. Starting from the HS metal MnGe, the Mn moments acquire a LS state as Ru content increases, up to diamagnetic RuGe. Both LS and HS states are metastable, being formed at (HT, HP), then quenched at RT during the synthesis. Such a HS-LS transition is induced here by *chemical* negative pressure, the concentration x, in contrast with the HS-LS transition induced under *applied* pressure in pristine MnGe. In the intermediate range 0.2 < x < 0.5 where both HS and LS state coexist, it leads to a phase segregation between HS Mn-rich regions and LS Ru-rich ones. Such effect recalls the phase segregation observed in Laves hydrides, which occurs thanks to a pressure induced HS-LS transition [3]. Up to now observed mostly with light H or D atoms, this magnetic-driven phase separation could occur here due to the high mobility of the Mn/Ru atoms during the synthesis.

More generally, magnetic phase diagrams in  $Mn_{1-x}tM_xGe$  are linked with changes of the band structure and subsequent magnetic interactions. The evolution from MnGe to RuGe, a low gap semi-conductor, strikingly differs from that towards FeGe, CoGe or RhGe with metallic character. In the metallic systems, it reflects a change from competing ferromagnetic (F) and antiferromagnetic (AF) near neighbour interactions to a F regime where Dzyaloshinskii-Moriya interactions act as perturbations. In  $Mn_{1-x}Ru_xGe$ , the short period, robust up to very high Ru content, suggests that the F/AF competition is still at play, but mediated by a lower density of charge carriers in the Ru-rich region. Band calculations together with transport data could check this interpretation.

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#### Unusual magnetic ground state of the cobalt honeycomb magnet BaCo<sub>2</sub>(AsO<sub>4</sub>)<sub>2</sub>

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In cobalt oxides, the interplay between crystal field, spin-orbit coupling, along with lattice geometry has proven to induce a rich variety of exotic states and magnetic properties [1-4]. In these systems, the  $3d^7 \text{ Co}^{2+}$  ions in an octahedral environment are in a high spin state with S = 3/2 and effective  $l_{eff} = 1$  moments and form a jeff =  $\frac{1}{2}$  Kramers doublet ground state. Owing to the delicate balance between spin-orbit coupling and magnetic interactions, which are usually of the same order of magnitude, numerous studies on a variety of cobalt magnets have shown a plethora of complex magnetic ground states and excitations [1-6].

The two-dimensional honeycomb compound  $BaCo_2(AsO_4)_2$  (BaCoAsO) is an example of a honeycomb-lattice cobalt magnet which has been extensively studied for many years, yet its magnetic ground state has long remained unsolved [7-12]. In this study we combined neutron scattering, magnetization measurements at very low temperature, along with linear spin wave calculations and Monte Carlo simulations to investigate its magnetic ground state. We found that the competition of interactions up to the fourth nearest-neighbour induces an exotic magnetic structure with topological defects. Moreover, a detailed crystal field analysis also suggests that Kitaev-like bond-dependent interactions may be relevant to explain the unusual magnetic ground state and excitations in this compound.

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#### Spin dynamics and Thermal conductivity in KTb<sub>3</sub>F<sub>10</sub>

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In recent years, the study of the coupling between the 4f electronic density and phonon modes has aroused a lot of attention. Such interactions are suspected to play a key role in a number of issues, ranging from highly frustrated magnetism to the design of smart materials in optics. In the celebrated Tb2Ti2O7 frustrated magnet, for instance, interaction between 4f quadrupoles and lattice vibrations may be responsible for the

lack of long-range order, as well as for the rise of new magnetoelastic modes, which remain unexplained so far [1,2,3]. Even if such hybrid modes have been reported for a long time [4,5], they are still topical as they may also be at the origin of the newly discovered "phonon Hall effect"

or "thermal Hall effect", which consists in the appearance of a sizeable thermal conductivity perpendicular to both the applied magnetic field and heat flux [6,7]. In c lose analogy with the classical Hall effect in conductive materials, the phonon Hall effect could result from an "anomalous" phonon velocity due to the remarkable topological properties of phonon bands. Actually, several theoretical works advocate that these properties may be due to the coupling between CEF and lattice degrees of freedom [8].

Terbium potassium fluoride KTb3F10 is another promising novel model system in which to investigate this physics. Our first investigations by means of neutron scattering show that the Tb ions occupy the vertices of a network of corner-sharing octahedra (cubic Fm-3m space group). Furthermore, our study shows that ground state is a crystal field singlet, with a peculiar field induced structure resulting from site dependent easy plane anisotropies (Figure 1 et Figure 2). The Tb3+ CEF scheme encompasses an excited doublet at 2.7 meV which is suspected to play a key role in the anomalous low thermal conductivity. Indeed, acoustic phonons, which are the mostly responsible of the heat transport, are likely scattered by this low energy magnetic fluctuations.



Figure 1 : Inelastic neutron scattering measurements on  $KTb_3F_{10}$  at PSI-Eiger – 2  $K - k_f = 2.662 - Q(200)$ 



Figure 2 : Calculation of the neutron scattering cross section on  $KTb_3F_{10}$  - 2 K - Q(2 0 0)

Such kind of interaction could lead to the unexpected shape of the thermal conductivity curve in this compound.

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# Investigation of the remarkable magnetic behavior of itinerant electrons in (Sc,Nb)Fe<sub>2</sub> compounds and their deuterides

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Since their discovery decades ago, Fe-based Laves phases have attracted a lot of attention due to their unusual magnetic and electronic properties and their technologically important multifunctionalities such as giant magnetoresistance, colossal magnetostriction, and giant magnetocaloric effect [1-2]. Indeed, they have been used as model compounds to investigate the itinerant-electron behavior of Fe 3*d* electrons as well as the localized character of rare-earth elements magnetism. In these intermetallics, it is well established that the iron magnetism is highly sensitive to the local environment of the Fe atoms (coordination number, short range order, nature of the near neighbors, bonding type and atom location in the crystal sites) [2-3]. Here we focus our attention on the pseudo-binary compounds (Sc,Nb)Fe<sub>2</sub> and their hydrides (deuterides), which crystallize in the hexagonal MgZn<sub>2</sub>-type structure (C14, space group  $P6_3/mmc$ ). Interestingly, ScFe<sub>2</sub> is a ferromagnet whereas NbFe<sub>2</sub> is an itinerant system exhibiting a quantum critical point (QCP) [4-5]. The identification of QCP in NbFe<sub>2</sub> opens up the new phenomenon of quantum tricriticality for experimental studies in a whole class of systems with buried or avoided ferromagnetic QCP.

We present a thorough experimental study of the magnetic and structural properties of the (Sc,Nb)Fe<sub>2</sub> alloys and their hydrides (deuterides) by combining magnetization, neutron and X-rays powder diffraction experiments. Magnetization results have demonstrated an extreme sensitivity of the Fe magnetism to the substitution of Nb for Sc and also to the hydrogen (or deuterium) insertion. Nb for Sc substitution induces dramatic changes on the magnetic properties such as a strong decrease of the ordering temperature and a significant reduction of the Fe magnetic moment. The insertion of hydrogen (deuterium) has huge effects on the structural and magnetic properties. As it can be clearly seen from figure 2, the saturation magnetization is doubled for ScFe<sub>2</sub>H<sub>3.58</sub> and ScFe<sub>2</sub>D<sub>3.14</sub> when compared to the parent compound ScFe<sub>2</sub>. At low temperatures, Sc<sub>1-x</sub>Nb<sub>x</sub>Fe<sub>2</sub> and their deuterides exhibit a ferromagnetic (FM) ordering of Fe moments. In most Sc<sub>1-x</sub>Nb<sub>x</sub>Fe<sub>2</sub> compounds, the magnetic moments for Fe(2a) and Fe(6h) atoms are aligned along the *c*-axis at low temperatures (Fig.1). By contrast, neutron powder diffraction (NPD) measurements performed (on D1B CRG instrument at ILL) on deuterides have revealed that Fe magnetic moments lie within the basal-plane of the hexagonal lattice (Fig.3). The easy magnetization direction changes from axial to basal-plane upon deuterium insertion. Furthermore, a peculiarly anisotropic lattice expansion as well as large negative thermal expansion (NTE) were discovered in some

Magnetism



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#### Multipolar interactions and spin dynamics in Tb<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub>

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Despite the large number of studies carried out over the last 2 decades on rare earth pyrochlore materials [1-8], the physics of terbium-based compounds, notably Tb<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub>, remains puzzling. This particular material shows no long-range order down to sub-kelvin temperature, yet exhibits static and dynamic magnetic correlations that still defy our understanding. We present here an in-depth study that sheds new light on this issue. In the first step, we have studied the low energy sector by inelastic neutron scattering, revealing the dispersion of a magnetic mode (0.1-0.3 meV) and completing the results reported in [9]. Our study is supported by RPA calculations of the S(Q,E) scattering function, taking into account quadrupole-quadrupole interactions among the elements of the ground CEF doublet [9,10,11]. We could reproduce the dispersion of this mode and concluded that Tb<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> is close to the boundary between a quantum spin ice and a quadrupolar ordered phase. In a second step, we also considered the dispersion of the first excited crystal field doublet, located at about  $\Delta = 1.5$  meV above the ground doublet. RPA simulations, based on an anisotropic bilinear exchange parameters. These results demonstrate the key role of both quadrupolar and dipolar interactions, which are undoubtedly at the heart of the unexplained properties of Tb<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub>.

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## Plenary talk by the President of the SFN PhD award jury:

#### Spin texture in a chiral magnet: experiment and theory

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Iron langasites are magnetic model systems that allowed us to study structural and spin chirality and the relationship between them [1,2]. In this presentation, I will show the consequences of chirality on the multiferroic properties of this compound, associated to a very long wavelength spin texture, evidenced experimentally under magnetic field [3]. I will also present a new theoretical study based on the minimal ingredients identified in these compounds, including magnetic frustration and Dzyaloshinskii-Moriya interaction, that predict novel spin textures and topological objects built upon the chirality vector [4].



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# Plenary talk by the laureate of the SFN PhD award:

will be announced during the JDN.

# Polyelectrolyte-based hydrogels: tuning structure and properties by ion specific effects and addition of charged anisotropic nanoparticles

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Hydrogels are at the forefront of scientific attention in the biological and biomedical fields providing the basis for stimuli-responsive artificial tissuesm or indeed in the food industry. We study physically crosslinked hydrogels based on ionene polyelectrolytes, positively charged polymer chains, for which we have observed strong counterion-specific effects in solution previously [1]. The nature of the ionene counterion modifies significantly the critical gelation concentration (cgc) and the elastic moduli of the hydrogels (Figure 1). Small angle scattering data (SAXS, SANS) reveal one, or more mesh sizes for the cross-linking polyelectrolyte networks, depending on the counterion nature. For gels with fluoride counterions, a transition between two mesh sizes is accompanied by a rapid increase in the elastic modulus [2].



Figure 1: Elastic moduli (G') of hydrogels based on ionene polyelectrolytes with different counterions:  $F^-$  (triangles), Cl<sup>-</sup> (squares) and SO<sub>4</sub><sup>2-</sup> (circles). The curves start at the critical gelation concentration (cgc) for each system. A very rapid increase of G' for the 6-F hydrogel is accompanied by a transition between two different distances between cross-linking nodes (mesh sizes) in the gel as revealed by a combination of SANS and SAXS [2].

Introduction of negatively charged clay nanoplatelets (100nm in lateral size, 1nm in thickness) into ionene-based hydrogels is achieved. Nanoplatelets organise in a regular face-to-face stacking manner, with a repeat distance following rather closely the hydrogel mesh-size (20-30nm) [3]. This distance is *independent* of the clay platelet concentration in the gel. SANS measurements, under the contrast matching condition of the clay particles, reveal that the hydrogel mesh-size is not modified in the presence of the clay particles. The degree of nanoplatelet ordering in the hydrogel is very sensitive to the negative charge location on the clay platelet, which is different for a given clay type. Interestingly, this resembles the observations in pure clay gels [4]. In both cases, charge location closer to the nanoparticle surface induces a higher degree of ordering in both types of gel.

Increased nanoplatelet ordering leads to an improvement of the elastic properties of the hydrogel, especially for concentrations close to the cgc. On the contrary, the presence of dense clay aggregates (tactoids), destroys the hydrogel network [3]. Introduction of anisotropic particles paves the way towards hydrogels with anisotropic permeation properties.

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#### From nanoporous material evolution in solution to water behavior in confinement

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The understanding of the evolution of nanoporous materials filled of or in aqueous solution such as membranes, cementitious materials, polymers for fuel cells, biominerals, clay materials, secondary minerals, corrosion products and spent nuclear fuels, is important to explain and to predict their macroscopic behaviour. This macroscopic behaviour is governed by the spatial distribution, the transport and the chemical reactivity of species present in confined solution which are mainly water molecules and ions.

In nanoconfined media, the system length scale crosses the classical key length scales describing energetics and equilibrium of electrolyte solutions next to surfaces. In this case, classical concepts fail to account for scale specific phenomena. Thus, to better understand the evolution of nanoporous materials, the descriptions of water molecules and ion properties need to be updated.

Here, we propose to illustrate how the understanding of water and ion behavior in confinement, can help to explain the alteration of glass in aqueous solution [1]. Indeed, when glass is in contact with a solution, a nanoporous silica-based material called "gel" is formed. Depending on the glass composition, this gel limits the reactive transport of water and thus, the glass and gel hydrolysis. To explain this phenomenon, our scientific approach was to determine the predominance of the effect of size, surface composition [2], ions nature [3][4] and their distribution in confinement [5], on water properties. By the use of silica model systems presenting various single-digit nanoconfinements (pore size and pore wall surface) and electrolyte with cations presenting rising kosmotropic properties, water structure, dynamics and reactivity [6] at a picosecond timescale were characterized using infrared spectroscopy, quasi-elastic neutron scattering, small angle X-ray scattering and hard X-ray reflectometry and related to the glass alteration kinetics.



Figure 1 : Scientific approach used to understand the effect of gel formed during glass alteration on the alteration of glass.

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# Stabilization of a liquid foam by ionic charges: simultaneous multiscale study in Fourier space and real space

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Foams are ubiquitous in daily life. This fluid, however, never reaches thermodynamic equilibrium and is constantly undergoing structural change until the fluid's gas and liquid components are separated. Physical and chemical phenomena occur at various times and scales, which makes it challenging to correlate how the foam ages. However, it is figuring out how to adjust its formulation so that it can regulate both its viscoelastic characteristics and its lifespan in accordance with the necessary uses.

It is important to describe a created foam in its whole, including the size of the supramolecular self-assemblies, the inter-bubble layer, and the bubbles (on a millimeter or centimeter scale) (at the nanometre scale). To track the multi-scale evolution of a developing foam, it is necessary to use a variety of experimental methodologies simultaneously and "in operando". In order to overcome this difficulty, a special cell has been created to demonstrate, first, the potential data collection as a function of time of a free-draining foam investigated utilizing simultaneously small angle neutron scattering (SANS at ILL/D33), electrical conductivity, and optical imaging. The combination of the findings and the creation of a specific analysis code allowed us to follow the kinetics of the different surfaces independently of the inter-bubble films, the Plateau border, and their time lag, and to describe a disjunction pressure isotherm as a function of the average film thickness within an evolving foam [1]. This novel approach was employed in a study that examined the stability of a foam made with an ethoxylated fatty alcohol as a non-ionic surfactant and in the presence of either ionic charges supplied by either a second ionic surfactant (sodium dodecyl sulfate) or nanometer-sized ions (nano-ions) with low charge density. In the first instance, the ionic species stabilizes the foam by attaching to the surfactant film via a hydrophobic interaction with the help of its fatty chain, whereas in the second instance, the association of the nano-ions with the surfactant monolayer is brought about by a potent (super)chaotropic effect. [2,3]. Despite the fact that both species tend to lengthen the lifetime of a non-ionic foam, minute similarities and differences are highlighted and explored by correlating findings, particularly macro- and nanometric quantities, and by linking them to the rheological response of the interfacial layers. Very recently, the study of foam and nano-ions has opened up exciting possibilities for extraction separation applications using the concept of superchaotropic flotation [4].



Fig: foam'up setup on D33 at ILL!

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# Insights into the microscopic mechanisms responsible for phase separation in a biphasic acidic solution for metallic ion extraction

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Ionic-liquid-based acidic aqueous biphasic solutions (AcABSs) recently offered a breakthrough in the field of metal recycling. The particular mixture of tributyltetradecylphosphonium chloride ([P4,4,4,14]Cl), acid, and water presents the unusual characteristic of a lower solution critical temperature (LCST), leading to phase separation upon a temperature rise of typically a few tens of degrees. Here, we dig into the mechanisms driving the counter intuitive phase separation upon temperature rise observe in this system. Using small angle neutron scattering, we could identify the micellar structure under various conditions of acid and temperature, observing the micelle aggregation eventually leading to the phase separation in an apparently similar mechanism.

Fine titration of the chloride ions present in the solution proved the exothermic adsorption of these ions at the micelle surface, increasing with temperature. Such ionic adsorption, similarly to acid addition, induces a screening of electrostatic interactions between the micelles, indirectly driving the phase separation. Such a mechanism of ionic or molecular adsorption at the micelle surface could be generalised for numbers of molecular systems presenting a Lower Critical Solution Temperature.



*Figure 1 : Ternary phase diagram of the [P4,4,4,14]Cl, water and chlorhydric acid solution, and sketch of the ionic liquid cation. The phase diagram is given for deuterated compounds for the need of neutron scattering. S1, S2 and S3 solutions are the investigated solutions.* 

<sup>[1]</sup> G. Meyer, R. Schweins, T. Youngs, J.-F. Dufrêche, I. Billard and M. Plazanet, How Temperature Rise Can Induce Phase Separation in Aqueous Biphasic Solutions, J. Phys. Chem. Let. (2022), 13, 2731–2736.

# Probing the emergence of electrostatic attraction between like-charged phospholipid membranes

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Counterintuitive electrostatic attraction between similarly charged macroions arises when multivalent counterions are present. Recently, it has been demonstrated that monovalent counterions alone can trigger attraction among fully charged phospholipid membranes [1]. This phenomenon has been attributed to the low dielectric constant of interfacial water [2]. To gain a deeper understanding of the interplay between hydration and electrostatic forces, we investigated the interaction among an even number of phospholipid layers deposited on a silica substrate. In this geometry, specifically at the air/solid interface, we were able to control osmotic pressure applied on the layers by adjusting air relative humidity. Using the D17 neutron reflectometer at ILL, we measured the thickness variation in a humidity chamber for membranes with varying charged surface density. From this data, we deduced the disjoining pressure between the layers. Our findings reveal an experimental crossover between a repulsive mean field regime and an attractive strong coupling regime.

[1] Tetiana Mukhina, et al, *The Journal of Physical Chemistry Letters* **2019** *10* (22), 7195-7199 [2] L. Fumagalli, et al, *Science* **2018** *360* (*6395*), *1339-13* 

#### Light responsive wormlike micelles

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"Smart" materials have the ability to change their structure and functionality in response to an external stimulus. The triggers can be diverse such as mechanical stress, electric or magnetic field, light, temperature, pH etc. We are interested in smart light-responsive wormlike micelles (WLM) [1] able to change their rheological properties upon UV irradiation.

The light-responsiveness of these systems originates from the incorporation of a photochromic group (azobenzene, Azo, figure 1a) in the chemical structure of the surfactant. UV irradiation provokes a reversible isomerisation from a *trans* stable apolar state to a *cis* metastable polar conformation that changes the polarity and the hydrophobicity of the molecule (figure 1b) leading to a global structural reorganization of the system.

In this work, we focussed on a simple system made of Azo-surfactants (figure 1b) and sodium benzensulfonate as cosurfactant (figure 1 c) in water. This mixture forms a gel-like state in absence of irradiation (dark) that can be switched 'off' or 'on' upon UV or blue light irradiation (figure 1d). The goal of the study is to give a precise picture of the structural modifications that cause such a reversible sol-gel transition.



Figure 1. a) Azobenzene and photo-isomerisation between trans (stable apolar) and cis (metastable polar) isomers. b) Azo-surfactant. c) sodium benzensulfonate d) gel-like behaviour

In this talk, we will present Small Angle Neutron Scattering experiments performed on PAXY at the LLB in dark and upon UV. These measurements will be completed by SAXS and cryo-TEM experiments.

[1] Z. Chu, C. A. Dreiss, Y. Feng, Chem. Soc. Rev., 42 (2013) 7174-7203

# Determination of the structure and rheology of a fluid allowing self-consistent subduction during convection in the laboratory

<u>*M. Pépin*</u><sup>12</sup>, C. Alba-Simionesco<sup>1</sup>, A. Davaille<sup>2</sup> and H. Remise-Charlot<sup>12</sup> <sup>1</sup> Laboratoire Léon Brillouin, UMR 12 CEA-CNRS-Paris Saclay

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Subduction of the lithosphere is a main feature of Earth's mantle plate tectonics and convective dynamics. To date, we have found only one fluid able to produce in the laboratory self-consistent subduction during convection: colloids of silica nanoparticles (Ludox®) [1]. The latter provides a large diversity of regimes depending on the nanoparticles volume fraction and the history of the material. From fluid to solid right through to gel, many structures can be observed [2]. It is important to identify which conditions are necessary to generate subduction.

Convection in this colloids can have a thermal origin (when the tank is heated from below) and/or a solutal origin (when Ludox®'s water evaporates). Regardless of its nature, convection creates heat and mass transfers which affect the fluid structure's organisation and lead to concentration gradients. These concentration gradients produce different fluid characteristics (Newtonian or shear-thinning, yield stress...), which in return, influence the convective patterns. Finally, it also leads to a stratified system with a denser and stiffer skin on the surface (figure 1). This stratified system allows episodes of self-consistent subduction: the skin formed on the surface breaks and sinks into the less-viscous fluid bellow. With the help of thermal analysis (Differential Scanning Calorimetry, DSC, and Thermogravimetric Analysis, TGA) and Small-Angle Neutron Scattering (SANS), [3], we can evaluate volume fractions and fluid texture (e.g. typical size of aggregates, formation of a more or less compact skeleton...) for the different states present in the convective system (liquid, paste, skin and subducted skin). This states at a given time can be compared to thin-layer drying during time (without convection and homogeneous in bulk) [4]. Then, these measures can be linked to the fluid's rheology. Thus, we can determine the rheological conditions for which the convective system is able to trigger subduction.



Figure 1. Foreground : sample of surface skin. Background : Tank for Ludox® convection

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<sup>[2]</sup> E. Di Giuseppe, A. Davaille, E. Mittelstaedt, M. François, "Rheological and mechanical properties of silica colloids: from newtonian liquid to brittle behavior" Rheologica acta, 05 (2012) 451-465.

#### COSAN nano-ion as novel solubilization agent of hydrophobic compounds

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The solubilisation of hydrophobic organic molecules in water is relevant in many fields (pharmacy, fragrancy, detergency...). Many studies have been conducted on the use of surfactants, macrocycles or co-solvents to achieve this goal [1].

Cobalta-bis-dicarbollide ( $[Co(C_2B_9H_{11})_2]^{-}$ ), otherwise known as COSAN, is an inorganic and ionic nanometric molecule characterized by a very low charge density. Although missing a traditional amphiphilic structure, COSAN stands out for its surfactant-like characteristics, such as surface activity and self-assembly ability (vesicles, micellar and L $\alpha$ /L $\beta$  phases depending on temperature and on its concentration in water) [2].

In the current study [3], we investigated how COSAN affected the solubilization of hydrophobic organic molecules of diverse types and logP values in water, and compared it to sodium dodecylsulfate (SDS), a classical anionic surfactant. In contrast to conventional surfactants, COSAN nano-ion has a higher solubilization efficiency for 2-butanol when COSAN concentration is below its critical aggregation concentration in water. We have used scattering techniques (SANS/SAXS/DLS) to observe the formation of nanometric co-assemblies of COSAN and 2-butanol in water and to decipher, using spectral techniques, an unconventional solubilization mechanism.



**Figure:** (a) The compounds that can be solubilized efficiently by COSAN (yellow region) and the COSAN / 2-BuOH coassemblies (blue region) on the log P scale. (b) SANS (red) and SAXS (green) spectra in absolute scattering unit of 0.04 mol/L NaCOSAN in  $D_2O$  in the presence of 3 mol/L hydrogenated 2-BuOH. Full lines represent simultaneous fits of the two scattering spectra using a biaxial ellipsoid model.

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- [2] P. Bauduin et al., Angew. Chem. Int. Ed., 34 (2020), 34, 2026-38.
- [3] I. Chazapi et al., J. Coll. Int. Sci., 638 (2023), 561-568.

## High Current Accelerator-driven Neutron Sources - State of the art

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Significant progress have been made on the last decade in the fields of (i) high current proton accelerators, (ii) the understanding of the moderation process of cold neutrons, (iii) time-of-flight neutron instrumentation. It has been realized recently that the combination of progress in these different fields has opened the opportunity the prospect of building new types of neutrons sources using low energy accelerators (in the range of a few tens of MeV) which could achieve performances in neutron scattering techniques which are comparable to medium power research reactors or spallation sources. Such sources have been dubbed HiCANS High Current Accelerator-driven Neutron Sources. Several institutes across Europe and also across the world are considering the possibility to build HiCANS sources to replace ageing research reactors or as a technical solution to build national neutron sources at a reasonable cost. In Europe these institutes have gathered within the ELENA association to work around the concept.

I will present the current state of the art of CANS and HiCANS across the world and the different concepts, which allow achieving useful performances for neutron scattering.

## The HERMES - LLB outstation at the JULIC Neutron Platform

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High current Compact Accelerator-driven Neutron Sources (HiCANS) have risen in recent years as a possible answer to the closure of various research reactors in Europe. HiCANS projects in numerous laboratories around Europe are currently at different stages of their development. The JULIC Neutron Platform (JNP) at Forschungszentrum Jülich (FZJ) was conceived as the testbed for the german HiCANS project, the High Brilliance Source (HBS) [1]. Taking advantage of an existing cyclotron at FZJ, a target-moderator-reflector assembly was explicitly built to test the different components of HBS and their interplay. This platform is able to deliver neutron pulses in the 100  $\mu$ s-2 ms range and is very well suited to evaluate the feasibility of neutrons scattering experiments at a HiCANS.

In this scenario, the Laboratoire Léon Brillouin (LLB) is focused on evaluating the performance of neutron techniques around this novel type of source. For that purpose and through a collaboration with the Jülich Centre for Neutron Science, the HERMES reflectometer was installed in 2022 at the JNP. HERMES [1] is a time-of-flight horizontal reflectometer that operated at the Orphée reactor until 2019 and was mainly employed for soft-matter studies. Since its installation and first tests at JNP in 2022, several improvements have been planned and implemented at HERMES in order to exploit its maximum performance. Our current goal is to perform reflectivity experiments with supermirrors as a proof of concept, as the flux at the JNP is several orders of magnitude lower than the original Orphée flux or the one expected for a HiCANS. Nevertheless, Monte-Carlo simulations showed that an instrument as HERMES operating at a HiCANS could match the performance of similar instruments at medium power research reactors. This work is part of the collaboration within ELENA and LENS on the development of HiCANS. It has been funded by the "CANS Inflexion" program at the CEA and the "IPHI-Neutron" SESAME project of the Île de France region.

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<sup>[2]</sup> F. Cousin, F. Ott, F. Gibert, A. Menelle, Eur. Phys. J. Plus, 126 (2011) 109.

## **Diffraction around HiCANS: example of the PRESTO instrument**

#### Xavier Fabrèges,

Laboratoire Léon Brillouin, UMR12 CEA-CNRS, 91191 Gif sur Yvette Cedex, France

In this presentation, the case of diffraction on time-of-flight (ToF) neutron sources will be explored with an emphasis on the performance expected from diffractometers on HiCANS. The first part will present the specificities of ToF diffraction and the parameters driving the instrument design. The second one will be devoted to the PRESTO diffractometer developed specifically for the French ICONE project.

PRESTO is a flexible and versatile diffractometer aiming to cover the needs of the magnetic materials community (multifunctional materials, frustrated magnets, molecular magnets, ...). A complete set of simulations, from neutron transport to virtual experiments, have been carried out to estimate the performance and scientific capabilities of the instrument on powder and single crystal samples.

# Spectroscopie : Etat de l'art et nouvelles opportunités

#### <u>Quentin Faure</u>,

Laboratoire Léon Brillouin, UMR12 CEA-CNRS, 91191 Gif sur Yvette Cedex, France

High-intensity accelerator-driven neutron sources (HiCANS) are proposed as a possible route to make an access to neutrons easier than today's sources such as spalliation source or nuclear reactors. This talk is devoted to spectroscopy. More specifically, it aims to discuss the needs of the neutron community in order to target a viable spectrometer in a possible future HiCANS facility.

I will first describe neutron spectroscopy in a broad manner and then focus onto what can be achieved scientifically nowadays in various fields (hard condensed matter, soft matter).

Secondly I will present a broad panorama of spectrometers, in particular new ones such as SHARP+ (ILL-France), MUSHROOM (ISIS-UK) or CAMEA (PSI-Switzerland). Finally, this presentation will lead to some discussion about "our" needs and which spectrometers we want for the French community.

# The ICONE Conceptual Design Report

#### Marie Plazanet,

LIPhy, University Grenoble-Alpes and CNRS UMR5588b

In 2022, the CEA and the CNRS asked to a small working group gathering neutron scattering experts from different universities and from the Laboratoire Léon Brillouin, as well as experts in accelerators to evaluate the possibilities which could be offered by a HiCANS type sources. A document titled "ICONE, UNE NOUVELLE SOURCE DE DIFFUSION NEUTRONIQUE FRANCAISE » has been produced. The document discusses various aspects ranging from the actual use of neutron scattering techniques in various scientific fields, the landscape of the current French neutron scattering community, the potential of HiCANS for neutron scattering and the current possible technical implementation of a HiCANS for the French community.

## The IKUR-FUN initiative in the Basque Country – Raison d'être and first steps

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The IKUR 2030 Strategy was recently launched by the Department of Education of the Basque Government. An initial investment of 100 M€ over the period 2021-2030 has already been committed in four specific priority areas: neurobiosciences; quantum technologies; high-performance computing & artificial intelligence; and neutron & neutrino science. In this latter context, the IKUR-FUN initiative (from the Spanish Fuente Ultracompacta de Neutrones) was born in late 2022. Its primary *raison d'être* is predicated upon the pressing need to explore and assess in depth the requisite levels of neutron provision for academia and industry in the Basque Country, with an emphasis on capitalizing from the latest developments in increasingly compact neutron sources and the opportunities that they offer. To this end, a multidisciplinary working group has been assembled, with an initial focus on instrument development, sustainable materials, and industrial applications. This contribution describes the overall rationale and drivers behind the IKUR-FUN initiative and its first steps during these early days. Some of these include ongoing & mature research & development programmes in neutron and neutrino science by current IKUR-FUN actors, as well as joint scientific & technological demonstrators in close collaboration with existing neutron sources elsewhere in the world. These activities constitute a necessary first step towards a fully fledged business case in support of the (much-anticipated) arrival of neutrons to the Basque Country in the foreseeable future.

\* On behalf of the IKUR-FUN working group.

## Why are the results of 'Total Scattering' experiments always so exciting?

#### Simon A.J. Kimber<sup>1</sup>,

# <sup>1</sup> Formerly at the Université Bourgogne Franche-Comté, Dijon, France; c/o Pôle Emploi, Agence Dijon Nord, Dijon, France.

The 'total scattering' method has recently become a popular means of looking beyond the average crystal (and magnetic) structures determined by diffraction. This method is based upon a Fourier transform of powder scattering data, to a real-space pair distribution function. This 'PDF' includes all diffuse scattering from disorder, and even magnetic correlations. It is hence sensitive to nanoscale symmetry breaking, and other short-range effects [1]. In the author's experience, these experiments *always* yield interesting results, especially when applied to materials with phase transitions. Some of the more prominent ones include e.g. emergent dipoles in thermoelectrics [2]; 'nematic' correlations in superconductors [3]; persistent local dimers [4] etc. However, some total scattering results are incompatible with e.g. spectroscopic results [5].

In this poster I will revisit some of the fundamentals underlying total scattering experiments using neutrons. Two examples will be given: 1) The study of local magnetic correlations in the layered two-dimensional magnet  $CuMnO_2$ ; and 2) An energy-resolved total scattering study of correlations in thermoelectric GeTe.

For CuMnO<sub>2</sub>, magnetic total-scattering reports two-dimensional spin correlations above the Néel temperature [6]. However, we show by energy analysis, that this is a result of the high energy scale of spin-wave dynamics. These are comparable to the incident energy used on (even t.o.f) diffractometers, distorting the measured diffuse scattering. By applying low-energy polarised neutron scattering, we show that the quasi-static spin correlations are actually one-dimensional, as expected from the distorted orbitally-ordered coordination of  $Mn^{3+}$  sites [7].

For GeTe, we show how t.o.f. neutron spectrometers can be used to determine *energy resolved* pair distribution functions. [8] This effectively yields atomic correlations on a picosecond timescale. The excellent thermoelectric performance of this material was previously claimed to be a result of symmetry breaking disorder at high temperature. Instead, we show that it is the result of highly anharmonic and anisotropic dynamics, with very large atomic displacements. This result unifies the average structure, spectroscopic, and local structure pictures, for this class of materials.

So why are the results of total scattering experiments always so exciting? At least partially, it is because of instrumental factors (lack of energy resolution, low incident energy...). In addition, the Fourier transform method 'repackages' diffuse inelastic scattering in unexpected ways. Finally, some approximations in modelling softwares are overly simplified. Taken together, these factors can give the *appearance* of local symmetry breaking, and hence imply exotic physics.

So why should we do total scattering experiments on crystalline materials *at all*? This technique remains uniquely sensitive to nm scale chemical disorder. In addition, our results support a fundamental theory of coupling between strain and atomic fluctuations in energy materials. This information is present in the PDF in a model independent manner. Finally, our results suggest future directions for neutron instrumentation. Many of our most pressing energy challenges rely upon materials with unusual dynamics, and a mixture of coherent and incoherent effects. Neutron scattering techniques, like the one we develop in [8] are uniquely able to extract the underlying atomic correlations, and are hence highly complimentary to e.g. large scale molecular dynamics simulations.

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## New Analysis Frameworks for FWS-QENS of Colloid Suspensions

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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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## A thermal neutron three-axis spectrometer IN8-Thermes at ILL in evolution

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The three-axis spectrometer IN8 offers to ILL users advanced conditions for studies of thermal excitations in single crystals and liquids. The instrument performance and flexibility are ensured by the use of large doublefocusing monochromators and analysers with independently variable and remotely controlled horizontal and vertical focusing (bending) of the crystal reflecting planes providing high counting rate even for small and lowscattering samples. The new monochromator unit consists of 4 different exchangeable crystal planes including two planes built from mosaic crystals of pyrolytic graphite and copper with the principal reflections PG002 and Cu200 and the other two planes assembled with elastically bent perfect silicon crystals set to make use of the reflections Si111 and Si311 with prohibited second-order diffraction harmonics. The further step in renovation of the spectrometer is a classical single-detector secondary spectrometer set-up called THERMES (THERMal Excitations Spectrometer) now commissioned at IN8. The new instrument benefits from a compact design that permits a larger accessible dynamic range (wider available angular ranges in the existing experimental zone) with particular attention paid to neutron shielding including special construction of the detector diaphragm. The most recent development includes a beryllium filter option for high-resolution studies of vibrational dynamics in hydrogen containing materials. A relatively low and structureless instrument background provides access to vibration energies in the range from a fraction of one meV up to hundred meV. The achieved performance of the new option is illustrated by recent measurements of organic compounds.

# Neutron scattering sum rules, symmetric exchanges, and helicoidal magnetism in MnSb<sub>2</sub>O<sub>6</sub>

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MnSb<sub>2</sub>O<sub>6</sub> is based on the noncentrosymmetric P321 space group with magnetic Mn<sup>2+</sup> (S = 5/2,  $L \approx 0$ ) spins ordering below  $T_N = 12$  K in a cycloidal structure. The spin rotation plane was found to be tilted away from the *c*-axis [1] resulting as a helicoidal ground state, which we refer as the tilted structure. In our previous diffraction study [2] we found no evidence that this tilted structure is favored over the pure cycloidal order (referred as the untilted structure) [3]. The ground-state magnetic structure, expected to be built and originate from seven nearest-neighbor Heisenberg exchange constants, has been shown to be coupled to the underlying crystallographic chirality with polar domain switching being reported. In this work, we apply neutron spectroscopy to extract these symmetric exchange constants [4]. Given the high complexity of the magnetic exchange network, crystallographic structure and complications fitting many parameter linear spin-wave models, we take advantage of multiplexed neutron instrumentation to use the first moment sum rule of neutron scattering to estimate these symmetric exchange constants. The first moment of neutron scattering provides a way of deriving the Heisenberg exchange constant between two neighboring spins if the relative angle and distance of the two ordered spins is known. We show that the first moment sum rule combined with the known magnetic ordering wavevector fixes six of the seven exchange constants. The remaining exchange constant is not determined by this analysis because of the equal spatial bond distances present for different chiral exchange interactions. However, we find this parameter is fixed by the magnon dispersion near the magnetic zone boundary, which is not sensitive to the tilting of the global magnetic structure. We then use these parameters to calculate the low-energy spin-waves in the Néel state to reproduce the neutron response without strong antisymmetric coupling. Using Green's response functions, the stability of long-wavelength excitations in the context of our proposed untilted magnetic structures is then discussed. The results show the presence of strong symmetric exchange constants for the chiral exchange pathways and illustrate an underlying coupling between crystallographic and magnetic "chirality" through predominantly symmetric exchange. We further argue that the excitations can be consistently modelled in terms of an untilted magnetic structure in the presence of symmetric-only exchange constants.

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<sup>[2]</sup> E. Chan, J. Pásztorová, R. D. Johnson, M. Songvilay, R. A. Downie, J. W. G. Bos, O. Fabelo, C. Ritter, K. Beauvois, C. Niedermayer, S.-W. Cheong, N. Oureshi, and C. Stock, Phys. Rev. B **106**, 064403 (2022).

<sup>[3]</sup> R. D. Johnson, K. Cao, L. C. Chapon, F. Fabrizi, N. Perks, P. Manuel, J. J. Yang, Y. S. Oh, S.-W. Cheong, and P. G. Radaelli, Phys. Rev. Lett. **111**, 017202 (2013).

<sup>[4]</sup> E. Chan, H. Lane, J. Pásztorová, M. Songvilay, R. D. Johnson, R. Downie, J-W. G. Bos, J. A. Rodriguez-Rivera, S.-W. Cheong, R. A. Ewings, N. Qureshi, and C. Stock, Phys. Rev. B 107, 144420 (2023)

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	Mon 9 Oct.		Tues 10 Oct.	Wed 11 Oct.		Thurs 12 Oct.
9:00- 11:00		BIOLOGY Plenary - Keynote 9:00-10:10	MAGNETISM Plenary - Keynote 9:00-10:10		Workshop COMPACT SOURCES (6 conf)	
		INSTRUMENTATION 10:10-10:50 (2 conf)	MAG 10:15-11:00 (3 conf)	<b>SOFT</b> 10:15-11:00 (3 conf)		
11:00-			Coffee	Coffee		Coffee
11:30 11:30- 12:30		INSTRUMENTATION 11:20-12:30 (4 conf)	SFN PhD Award Ceremony President of Jury Laureate		Workshop COMPACT SOURCES Roundtable	
12:30- 14:00	0- Registration - Buffet 0		Lunch	Lunch		Lunch
14:00-	Registration - Coffee <b>Opening</b> 14:30-14:50		Posters	SOFT MATTER Plenary – Keynote 14:00-15:10		
16:00			14:00 – 15:30			
<b>CONDENSED MATTER</b> Plenary – Keynote 14:50-16:00			<b>SOFT</b> 15:15-16:00 (3 conf)	MAG 15:15-16:00 (3 conf)		
16:00- 16:30	5:00- Coffee 5:30		Free time	Coffee		
16:30- 17:00	<b>CM</b> (2 conf)	<b>BIO</b> (2 conf)		ENSA – AG SFN		
17:00- 18:30	17:00-Session NEWS18:30from FACILITIES, 2FDN					
18:30- 19:30	8:30- Welcome cocktail – 9:30 Posters		Musical performance			
19:30	19:30 Dinner		Dinner	Dinner		

