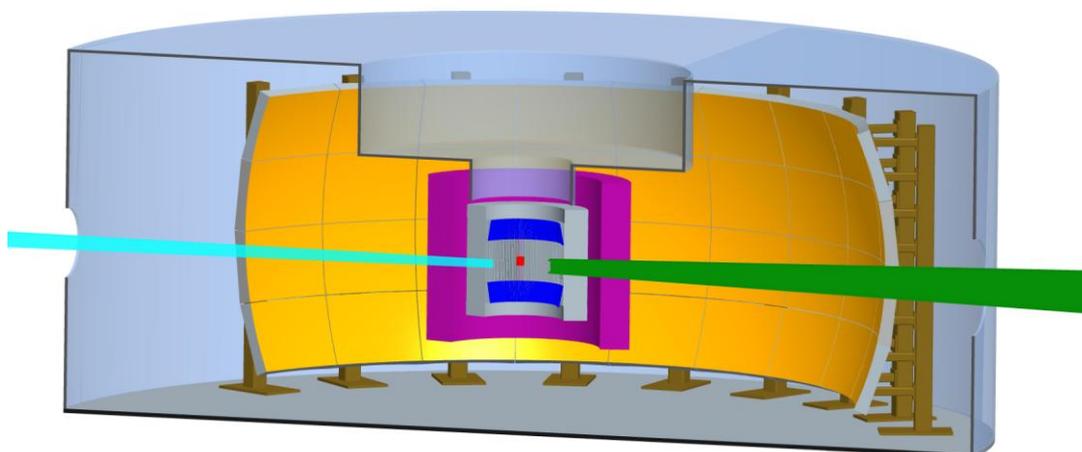


FIRES: A Novel Backscattering Spectrometer for ISIS

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Executive Summary

FIRES is a novel neutron spectrometer designed to enlarge by one order of magnitude the spectral window presently available at the ISIS Pulsed Neutron and Muon Source. The extension to microelectron-volt resolution or, in the time domain, to nanosecond relaxation timescales will open up new vistas in a wide range of research areas including biology and soft matter, energy conversion and storage, chemical catalysis, and quantum magnetism. With a unique combination of high spectral resolution and wide dynamic range, FIRES provides a step-change in low-energy spectroscopy at ISIS and beyond. Three potential sites have been identified to position this new spectrometer. The preferred option is a beamline at the south side of Target Station I. This report explores in detail why and how FIRES is to become an integral part of the ISIS instrumentation suite.

1. Introducing FIRES

FIRES will be a novel neutron spectrometer to replace IRIS at the ISIS Facility, Rutherford Appleton Laboratory, United Kingdom [1]. It will achieve an order of magnitude higher energy resolution compared to the currently operational ISIS backscattering neutron spectrometers OSIRIS and IRIS. Access to a spectral resolution well into the micro electron-volt (μeV) domain will open up a plethora of fresh opportunities in a wide range of scientific areas including biology, energy, environmental, and fundamental condensed-matter research. In this way, FIRES responds to one of the top priorities of the UK Science and Technology Facilities Council to *exploit and upgrade ISIS for maximum impact in key research areas*, as detailed in its latest strategy document [2].

In addition to the above science drivers, a growing user community, presently well over forty UK-based principal investigators, use the ISIS quasielastic spectrometers OSIRIS and IRIS on a regular basis. With an average of six days per experiment, the UK-based research community alone would already provide a distinct oversubscription for FIRES with just one proposal per group per year. As a natural replacement for IRIS, FIRES is also well aligned with current plans to extend the capabilities of the OSIRIS spectrometer into the TeraHertz domain, an initiative supported by ca. 150 research groups worldwide [3]. By exploiting modern chopper designs and state-of-the-art neutron technologies, FIRES will extend and complement the existing ISIS instrument suite. In turn, a revitalised spectrometer will renew and expand its user community.

This document is a compilation of efforts by the ISIS Quasielastic Neutron Scattering (QENS) Section in partnership with the user community to bring the FIRES project forward. As such, it has been organised as follows: Section 2 presents the science drivers and explains in some detail why

FIRES is indeed a much-needed addition to the existing instrument suite at the ISIS Facility. As each scientific area has been presented in detail, the reader might want to skip to the relevant subsection for details on how FIRES will address new scientific challenges in their particular area of expertise. Section 3 provides a technical specification of FIRES, as well as a comparison with similar instrumentation at ISIS and elsewhere. Section 4 presents a discussion of possible location scenarios and associated resource requirements. We close by presenting a series of recommendations on the best way forward for the FIRES initiative in the medium and long terms.

2. Why FIRES: Science Drivers

Exploring complex matter on nanometer length scales has become a central theme in contemporary condensed matter research, particularly in soft matter and biology. Probing longer length scales requires that the corresponding time scales are also increased. As a result, a strong scientific demand presently exists for higher-resolution neutron spectroscopy instrumentation. In addition to the above paradigm shift, a vast range of central issues requiring improved spectroscopic capabilities remain to be addressed not only in fundamental condensed-matter physics (strongly correlated electron systems, quantum fluids) but also at the interface between chemistry and physics (energy conversion and storage, ionic conduction, confined fluids).

The FIRES instrument addresses these challenges with a ten-fold increase in spectral resolution compared to current capabilities at ISIS, to reach nanosecond relaxation times while retaining a useful momentum-transfer (Q) range, from 0.1 up to 2 \AA^{-1} . As a result, three orders of magnitude in dynamics can be swept in a single measurement, from ps to ns

timescales. Since these timescales match state-of-the-art capabilities in molecular dynamics (MD) simulations, a profound understanding of complex materials will ensue from this technical development.

In contrast to existing backscattering instruments, FIRES will provide an unprecedented versatility, stemming from its ability to trade neutron flux for spectral resolution. It also offers a superb energy resolution over a wide range of energy transfers, opening up fresh and unexplored opportunities for high-resolution neutron spectroscopy at low energy transfers. The examples covered in this scientific case comprise all aspects of this innovative spectrometer design.

2.1. Biology and Soft Matter

The current trend in soft condensed matter and biology is to study materials of increasing complexity [4]. As such, an accurate separation of dynamical processes is key to both the analysis and subsequent interpretation of experimental data. Motions can span a wide range of timescales, from the very slow relaxation associated with viscous flow, to fast, sub-picosecond vibrations. As a result, it is often mandatory to combine information from several experimental techniques in order to attain a comprehensive and consistent understanding of the dynamical response in such systems. QENS is a unique technique as it offers simultaneous frequency and spatial information on the motions of specific moieties in multi-component systems (via H/D substitution). This information is directly comparable to the output of molecular dynamics (MD) simulations, a partnership which is necessary for the study of complex materials.

Polymers

The physical and chemical environment surrounding polymers can be tailored to alter the properties of these materials in fundamental and unexpected ways. Exploring these effects, particularly under conditions of strong spatial confinement, is a thriving area of research in soft condensed matter, ranging from nanocomposites for new material formulations, to polymer electrolytes for applications in energy conversion and storage.

Dynamic heterogeneity in thermodynamic polymer blends is a well-established phenomenon [5]. It is observed in polymer blends characterised by largely different glass-transition temperatures T_g leading to very different dynamical behaviour (e.g., polyethylene oxide / polymethyl methacrylate mixtures). QENS has established that there is a temperature range where the high- T_g component is frozen on the timescale of the segmental dynamics of the low- T_g component. In this situation, this low- T_g fraction evolves dynamically within the frozen, or confined, network provided by the high- T_g material [6,7]. In order to cover the different relaxation times characterising either component in the blend a wide dynamical range is necessary.

Polymer thin films have been used to explore the effects of confinement on the glass transition. These studies have revealed atypical and subtle phenomena such as the dependence of T_g on film thickness and thermal expansivity. It has been found that a lower T_g is not always related to the higher mobility characteristic of these soft materials. This finding contradicts our current understanding of the glass transition process. To further understand these phenomena, it has been necessary to combine QENS measurements on polystyrene thin films using two spectrometers with different energy resolutions, namely, HFBS (NCNR, USA)

and OSIRIS (ISIS, UK). These results have been interpreted in terms of cooperatively rearranging regions [8].

Ionic conduction mechanisms in polymer electrolytes are of central importance for the subsequent use of these materials in battery electrodes [9,10]. For instance, in polyethylene oxide - LiClO₄ mixtures, the lithium ions are thought to be coordinated to the oxygen anions, and ionic conduction involves a rearrangement of the oxygen environment. This is accompanied by a strong relaxation of the local chain structure. Changes in the local chain dynamics are typically in the ns range, well within the reach of FIRES.

The development of the 'bulk heterojunction' approach for polymer solar cells using conjugated polymers such as poly-3-alkyl-thiophenes (e.g, P3HT), has been the subject of renewed interest in the exploitation of carbon-based materials for energy applications [11]. Blends of P3HT and C₆₀ derivatives are not only low-cost alternatives to current technologies, but also the functional groups along the conducting backbone provide solubility and thus enhanced polymer processability [12]. Nevertheless, their structure has to be controlled and their dynamics understood in order to avoid a reduction of charge-carrier mobility leading to degradation of device performance. Since these polymers are intrinsically semi-crystalline, backbone and side-chain mobilities span ps-ns timescales.

Biological Molecules

Proteins are an integral part of biological systems, and ultimately responsible for a plethora of intra-cellular biochemical processes. Proteins are not rigid molecules and their mobility, internal motions, and conformational transitions are key to their biological function. Understanding the interplay between structure, dynamics, and

function in these complex macromolecules clearly necessitates further input from state-of-the-art experimental probes and, in particular, QENS [13,14]. Recent QENS experiments at ISIS have been performed on more realistic biological systems such as, for example, the study of water dynamics in *E. coli in vivo* [15] and human red blood cells [16]. Once more, these works reflect an increasing trend to tackle soft and biological systems of increasing complexity.

Proteins are also an important component of cell membranes. Exploring the relationship between internal protein motions and ultimate catalytic function, as well as the effect of lipid environment on enzymatic activity, are areas of intense study, as well as heated controversy [17]. A large body of experimental data suggests that the so-called 'dynamical transition' in proteins (a sharp rise in the atomic mean-square displacement above ~200 K), appears to be slaved to that of the local solvent. On the other hand, the physical properties of the lipid membrane such as lateral stress and mobility also affect the catalytic rate of enzyme turnover, therefore suggesting that a complex interplay between membrane and protein motions lies at the heart of catalytic and biological function [18]. An instrument like FIRES will enable the study of protein motions and lipid mobility within the cell membrane as a function of hydration level and protein concentration, by providing direct experimental access to these conformational changes (see Figure 1). Likewise, lipid membranes cannot be understood without taking into account their aqueous environment. The structure and dynamics of bound and bulk water provide important clues on the flexibility and transport properties of membranes. QENS gives important insight into the translational and rotational dynamics of the bound and bulk water in these biological environments [19].

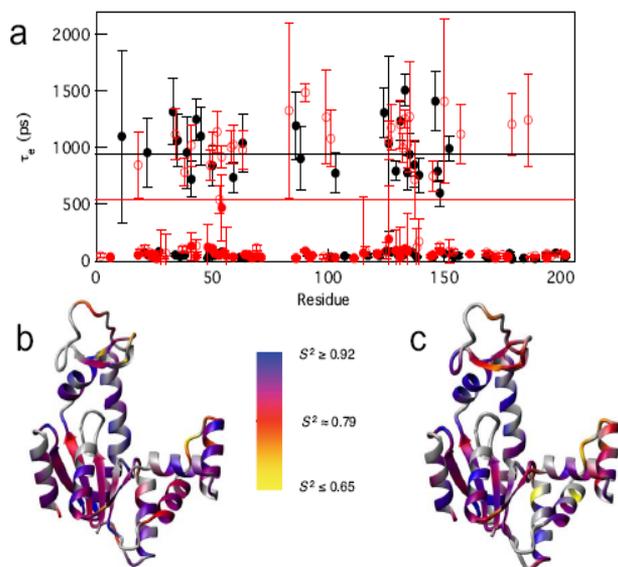


Figure 1: a) Relaxation times for internal motions of the protein apo thermoAdk at 50 °C (black filled circles), 80 °C (red filled circles), and 80 °C in the presence of 20% glycerol (red open circles). Addition of 20% glycerol slows down the overall rotational diffusion at 80°C to the value observed at 50 °C without glycerol. The order parameter S_2 in the absence and presence of 20% glycerol is shown in (b) and (c), respectively. Large differences in S_2 are seen in the lid regions where many residues display complex internal dynamics on multiple timescales. Adding glycerol slows the overall tumbling of the molecule. For more details see Ref. [18].

Smaller biological molecules such as hormones, neurotransmitters, toxins, or drugs are also of immense relevance to biology. Neutron diffraction has been used recently to unravel the solution structure of neurotransmitters, yet little information is available on how such molecules move and aggregate in solution. Bioprotectants such as the disaccharides are yet another important class of small molecules of biological interest [20]. Disaccharides such as trehalose are so-called cryptobiotic substances, i.e., they allow proteins to survive in hostile conditions, such as high or low temperatures or in the absence of oxygen. They protect the surface of biological molecules by acting as a stabilizer

against the surrounding environment. At present, two different theories can explain the physico-chemical mechanisms behind bioprotection: while the first one suggests that the sugar acts as a substitute for water, the second ascribes the protective properties of trehalose to its glassy characteristics. Using QENS in combination with H/D contrast-variation techniques allows the measurement of either the water or the sugar dynamics [21,22]. Access to relaxation processes near the glass transition requires high resolution spectrometers such as FIRES. Furthermore, neutron data and MD simulations spanning experimentally accessible timescales will both be necessary to distinguish between these two theoretical scenarios (see Figure 2).

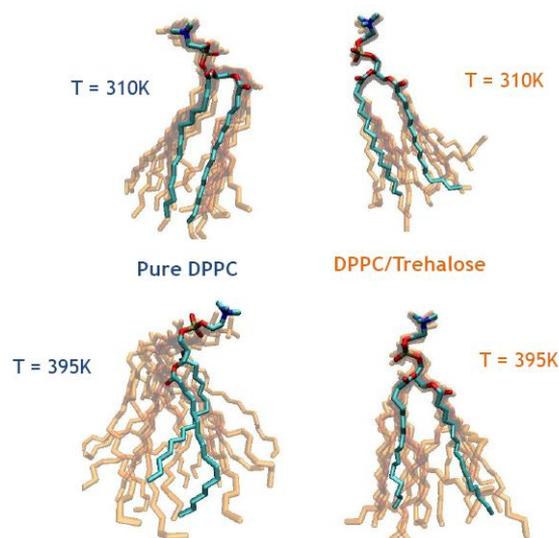


Figure 2: Snapshots of molecular dynamics simulations of DPPC lipid molecules with and without trehalose [4].

Last but not least, the effects of external stimuli such as hydrostatic and chemical pressure on biological activity and function are key to understanding Alzheimer's disease or the adaption mechanisms of organisms under extreme conditions, e.g., extremophiles at deep-sea level. Recent neutron experiments at high pressures indicate a slowing down of protein motions in the 1-100 ps timescale [23] accompanied by a reduction of the fraction of mobile

protons [24]. Simulations reveal that the effect of pressure actually depends on the timescales under consideration. While volume reduction increases structural hindrance and slows down motions at times >10 ps, it also increases the effective collision frequency of neighboring atoms leading to fast diffusive motions in the sub-ps regime.

Pharmaceuticals

Self-assembled 'soft' nanostructures are being used in a number of important applications in medicine and pharmacology. One example is the use of hydrogels as drug-delivery systems. Hydrogels are water-insoluble networks of cross-linked polymer chains such as polyvinyl alcohol, a compound used in surgical threads. Inside these polymer networks, a huge amount of water is stored (up to 99% of the matrix weight) making the hydrogel structure flexible and suitable for scaffolding in tissue engineering. The cross linking of the polymer chains is a decisive factor for the functioning and biocompatibility of the hydrogel. In drug-delivery applications, the drug release is strongly dependent on the translational mobility of the caged water [25,26]. QENS is superbly suited to probe the characteristic timescales for water mobility both under confinement and in the bulk, to explore the efficacy of newly synthesised hydrogels with enhanced physical and chemical stability. Moreover, the high spectral resolution afforded by FIRES is required to separate water diffusive motions from the segmental dynamics of the hydrogel matrix.

Food Science

Freeze-drying (or lyophilisation) is a routine industrial process to preserve food [27]. Fruits and vegetables contain up to 90% water, the rest being primarily carbohydrates (sugars). A large fraction of the water is in direct contact with the carbohydrate matrix. The strong interaction

of water with biomolecular surfaces leads to a restricted motion of the former. The presence of the carbohydrate molecules results in a breaking of the hydrogen-bonded network as well as a reduction of water mobility with increasing sugar concentration. The structure and dynamics of frozen and dried biological material may differ greatly from that of the starting materials. Using contrast-variation techniques, QENS provides exquisite insight into the dynamics of the hydrogenous components of food and its surrounding aqueous environment before and after the freeze-drying process [28].

2.2 Quantum Matter

Strongly Correlated Electron Systems

Quantum magnets are arrays of atoms each carrying a low intrinsic spin angular momentum, typically $S=1/2$ or $S=1$. As such, they are archetypes to unravel the ultimate consequences of quantum mechanics in strongly interacting many-body systems. Novel ground states of the entire many-body ensemble may be realised by varying the dimensionality, the nature of the magnetic interactions, or the strength and directionality of external fields. Examples include quantum spin liquids or magnon Bose-Einstein condensates [29]. Understanding the properties of such 'Quantum Matter' represents an enormous intellectual challenge, as well as requires the development of new experimental techniques and facilities. The enhanced energy and wavevector resolution of FIRES will make significant contributions to many areas of quantum magnetism where a detailed knowledge of spectral line shapes can provide insight into new physical phenomena, including: how quantum phase transitions occur, what stabilises the bound states of quasiparticles, how magnons become unstable in strongly fluctuating spin systems, or when magnons fractionalize into spinons.

At temperatures approaching absolute zero, the ground state of a many-body system changes in a fundamental way as a function of external stimuli such as pressure or magnetic field. In this case, transitions are driven by quantum fluctuations (allowed by the Heisenberg uncertainty principle). Unresolved issues range from the very nature of quantum critical fluctuations to how emergent quasiparticles transform upon the crossing of critical points in the phase diagram [30]. Moreover, quantum magnets can be used to realise a wide variety of universality classes for phase transitions, including the emergence of exotic quantum symmetries near the quantum critical point in an Ising chain (see Figure 3) [31], or Bose-Einstein magnon condensation in gapped dimerized systems upon the application of a magnetic field or external pressure.

The high energy and wavevector resolution provided by FIRES can be exploited to address these fundamental questions, including whether quasiparticles are well-defined (or ‘sharp’) at the transition or, alternatively, acquire finite lifetimes due to the presence of additional scattering and decay channels. High spectral resolution is required to determine lifetimes accurately and to test theories at a quantitative level. Real-life perturbations also enrich the physics of quantum phase transitions. For example, in quasi-1D Ising magnets, weak interchain couplings can transform the spectrum of critical fluctuations for gapless solitons into a series of gapped stable bound states [32], similar to the problem in particle physics of quark confinement into mesons. High energy and wavevector resolution would allow direct experimental tests of such predictions and the discovery of entirely new physical phenomena.

Quantum spin ladders such as those found in the compound $(\text{Hpip})_2\text{CuBr}_4$ have also attracted considerable theoretical interest yet experimental data remains sparse. In our context, $(\text{Hpip})_2\text{CuBr}_4$ has the

appealing feature that the dominant energy scale – the so-called “rung exchange” – lies around 1 meV. For this family of quantum magnets, such a low-lying excitation spectrum will allow the mapping of the entire phase diagram with an applied magnetic field for the first time. Given the low energy scale of the excitation spectrum and the need to perform a quantitative analysis of spectral line shapes, these experiments are ideally suited for FIRES.

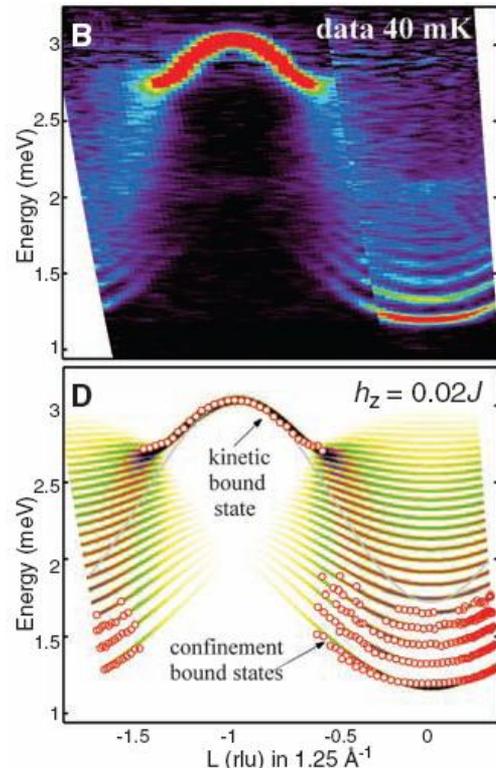


Figure 3: Map of spin excitations in the quasi one-dimensional Ising ferromagnet CoNb_2O_6 measured on OSIRIS [31]. The lower part shows model calculations.

Frustration

Frustration in a many-body system occurs when all interaction energies cannot be simultaneously minimized. Generally, frustration prevents the occurrence of magnetic order or strongly reduces the magnetic ordering temperature compared to what would be expected from a consideration of exchange interaction

energies alone. Novel ground states have already been observed, including spin liquids, spin ices, resonating valence-bond states, cooperative paramagnets, and unconventional magnetic ordering (non-collinear, partially ordered, or short-range structures).

Geometrically frustrated low-spin antiferromagnets are a good example where a detailed analysis of spectral line shapes has played an essential role. Theory proposes these materials as candidates for spin-liquid states where elementary excitations are not the conventional $S=1$ magnons, as in ordered magnets, but are topological $S=1/2$ spinons [33]. A broad scattering continuum is a direct manifestation of strong quantum fluctuations leading to fractionalization of conventional $S=1$ spin waves into a pair of deconfined $S=1/2$ spinons at higher energies. Understanding under what circumstances spinons exist is of fundamental importance. A groundbreaking experiment would be to observe how spinons become confined again into spin waves at the lowest energies, i.e., a sharp mode would split off the bottom of the continuum at the lowest energies. The tighter energy and wavevector resolution of FIRES would be ideally suited to probe this novel physics.

Spin ice is a material class where the arrangement of spin moments is topologically identical to proton disorder in water ice. These systems show spin fluctuations down to low temperatures and over a wide range of relaxation times. Nuclear-spin excitations have recently been discovered in these materials [34]. The existence of these low-frequency hyperfine interactions opens up the exciting possibility of manipulating nuclear spins through the electronic-spin system. To date, not much is known about the nuclear-spin system and its resulting excitation spectrum. Nuclear-spin excitations lie in the μeV range and require a very high energy resolution to isolate them

from the quasielastic dynamics of the electronic spin system.

Nanomagnetism for Spintronics and Quantum Computation

Molecular magnets consist of clusters of few interacting magnetic ions arranged in well-defined topologies such as rings, wheels, or horseshoes. In these so-called Single Molecule Magnets (SMMs), the slow relaxation and quantum tunnelling of the magnetisation observed at low temperatures has opened new possibilities for possible applications in information storage and quantum computing [35]. Research efforts in this field are highly interdisciplinary, with important contributions from both chemistry and physics. On the chemistry front, new materials are synthesised and subsequently characterised with a variety of physical probes. The aim of these studies is to gain a quantitative physical understanding of the relaxation and tunnelling phenomena in order to increase operating temperatures.

High resolution inelastic neutron scattering is one of the most powerful techniques for the study of SMMs [36]. In contrast to other techniques such as bulk magnetisation and heat-capacity measurements, neutron spectroscopy provides direct access to the electronic Hamiltonian responsible for the unusual magnetic properties of SMMs. Exchange and anisotropy interactions typically lead to energy splittings as low as a few μeV , energies, beyond the reach of current instrumentation at ISIS. An increased energy resolution will lead to a much more detailed picture of the interplay between exchange and anisotropy interactions in SMMs and, thus, to a more robust understanding of their unusual magnetic behaviour.

Quantum Liquids and Solids

Simple quantum liquids, such as ^3He and ^4He , have been of fundamental importance for the understanding of strongly correlated

fermionic and bosonic systems. In view of their simple and well-known interactions, the Helium isotopes provide an ideal test bed for many-body theories which can then be ported to more complex scenarios in solid-state physics. In fact, inelastic neutron scattering studies on the hybridization between fermionic and bosonic excitations in ^3He - ^4He mixtures has provided a great deal of insight into excitation spectra and quasiparticle couplings, paving the way to a better understanding of complex, strongly correlated electron materials [37].

An example of interacting excitations is the Pitaevskii conjecture for liquid ^4He , which states that the energy of a single-particle excitation at any Q cannot exceed the two-roton energy [38]. On IRIS, partial confirmation of this conjecture has been obtained in the high- Q region [39]. However, the conjecture should also be valid at lower Q values, where it appears from low-resolution triple-axis and time-of-flight measurements as if the maxon energy actually exceeds the energy of two-roton excitations at high pressures. A solution of this long-standing problem would be achievable on a spectrometer like FIRES.

Another long-standing problem in the physics of Helium concerns the energy shift and line width of the roton excitation in superfluid ^4He . While the line width is well described in terms of scattering processes caused by thermally excited rotons (the so-called Landau-Khalatnikov mechanism), the energy shift is not. However, the two quantities are governed by the same Feynman diagrams and, thus, should be intimately linked. On the experimental front, low-temperature data measured on neutron spin-echo and reactor-based backscattering spectrometers do not agree with data at higher temperatures taken on time-of-flight spectrometers. The specification of FIRES is such that the energy resolution would cover the whole temperature range of interest, leading to a single, self-consistent data set to test opposing theoretical scenarios.

Recent experiments suggest that Helium can exist in a supersolid phase [40]. Supersolidity describes the coexistence of solid and superfluid properties. Whether this novel state is related to Bose-Einstein condensation (as in the fluid phase) is still the matter of intense debate. The existence of elementary excitations, like phonon-roton dispersion, might provide the key for a truly microscopic understanding of the above findings. FIRES is the ideal tool to search and characterize the associated excitation spectrum in this new phase of matter.

Quantum-mechanical effects are also crucial to describe the bulk properties of the condensed hydrogens where crystal plasticity is dominated by zero-point-energy effects and quantum rotations. Under ambient conditions, solid para-hydrogen remains rotationally disordered down to fractions of a Kelvin. Owing to its simplicity, intermolecular interaction potentials able to deal with equation-of-state data up to fairly high densities are known with great accuracy. This makes hydrogen an ideal test bed for comprehensive studies whereby a controlled amount of crystal disorder can be introduced via the use of para-ortho hydrogen mixtures over a wide range of pressures and temperatures. The ability of FIRES to probe both quasielastic and low-energy inelastic modes (associated with centre-of-mass motions and internal rotation, respectively) will certainly provide new insights into the nature of rotational disorder and quantum-mechanical effects in the condensed phases of this fundamental diatom.

2.3. Functional Materials

Complex Liquids and the Glass Transition

The response of materials to extreme conditions (high temperatures and pressures) embraces fundamental questions in material science and in adjacent fields

such as Geology and the Earth Sciences. Materials of prime interest are the oxides, which are closely tied to the geosciences and glass technologies. From a physical point of view, a number of oxides are of interest, namely, pure oxide melts consisting of only a single network-forming component (e.g. SiO_2 , B_2O_3), binary and ternary mixtures consisting of network-forming and network-modifying components (e. g. Na_2O , CaO), as well as intermediate compositions (e.g. Fe_2O_3 , Al_2O_3). The oxide matrix arises from the linking of network-forming entities with highly directional covalent bonds. In the case of binary and ternary mixtures, there is also a component bound to the network (so-called 'modifier') characterised by much faster motions. In the alkali-silicate melts, QENS has been employed to develop a fundamental understanding of the microscopic origin of fast-ion transport and its relation to the strongly non-linear dependence of the viscosity on composition typically found in these materials. Combined MD simulations and QENS have substantiated earlier models which suggest that the alkali ions move along preferential pathways in the network [41].

Typical geological melts are characterised by at least three components including the oxides Al_2O_3 and Fe_2O_3 . Investigations of ternary sodium aluminosilicates have shown that the addition of Al_2O_3 has a substantial influence on the dynamics of the oxide matrix and slows down the motion of the modifier to time scales exceeding 100 ps at typical melt temperatures of 1600 K. A decay of the correlation between pairs of alkali atoms takes place on even longer time scales and should show a peculiar momentum-transfer dependence that has not yet been accessed by experiments. Further quite intriguing questions arise regarding the role confined water plays under conditions mimicking volcanic magmas. With an instrument like FIRES, these time scales will become accessible and, further, could be combined with results

from atomistic MD simulations to understand these complex multi-component systems.

The study of strongly associated molecular liquids offers new and important clues about the relation between the liquid state and metastable phases of matter. For example, the single-particle stochastic dynamics of hydrogen fluoride, the simplest hydrogen-bonded liquid, has been recently mapped on IRIS with unprecedented detail [42]. At low temperatures, there is a marked breakdown of the celebrated Stokes-Einstein and Debye-Stokes-Einstein relations for translational and rotational diffusion, respectively. Interestingly, these deviations obey the same power-law behaviour found in prototypical models of network-forming liquids, as well as in a wide range of glasses, suggestive of similar underlying microscopic mechanisms. Extension of these studies to more complex hydrogen-bonded molecular liquids, including medium-sized biomolecules, would require an order-of-magnitude increase in energy resolution.

The additional orientational degrees of freedom present in molecules also add richness and complexity to their behaviour below the melting point. Many molecular crystals exhibit dynamic orientational disorder arising from a delicate interplay between highly directional forces (typically of an electrostatic nature) and those arising from the overlap of non-spherical electron clouds and dispersive interactions. These 'plastic crystals' are intermediate between the molten or glassy states and the fully ordered crystal [43]. Certain materials such as ethanol exhibit transitions between the 'glassy crystal' and plastic-crystal phases, and these are fully analogous to those exhibited by the canonical glass-to-liquid transition. The extended dynamic range and tighter resolution of FIRES will enable a comprehensive characterization of these molecular glasses beyond what is currently possible.

Energy Research and the Hydrogen Economy

Hydrogen, the most abundant element in the universe, has great potential as a clean and secure source of energy. Unlike fossil fuels, it can be generated from renewable energy sources and its combustion produces water, a harmless by-product in comparison to the burning of hydrocarbons. The quest for efficient hydrogen-storage materials remains a key scientific challenge with profound implications for long-term global sustainability. Recognising this need, hydrogen research has become an important part of the UK Research Councils' Science Strategy for the next decade and beyond [44].

High-resolution neutron spectroscopy at low energy transfers is an exquisite probe of the potential energy landscape and mobility of the hydrogen molecule in storage materials. Present instrumental capabilities on inverted spectrometers are best exemplified by referring to recent IRIS studies on the interaction of molecular hydrogen with carbon nanostructures [45,46] and metal-oxide nanosurfaces [47]. Access to the potential energy landscape of molecular hydrogen in these novel materials requires simultaneous quasielastic and inelastic neutron-scattering measurements. Spectral transitions associated with hindered quantum rotations, tunneling, or stochastic diffusion span the energy-transfer range 0-20 meV. High spectral resolution is mandatory in order to interpret the spectroscopic data in terms of site occupation or the strength of the interaction potential. The case of hydrogen uptake by carbon nanohorns serves to illustrate present instrumental capabilities at ISIS (cf. Fig. 4). With an energy resolution of 150 μeV at 15 meV on IRIS, it is however not possible to fully resolve the observed multiplet structure around 14.7 meV, or to distinguish between homogeneous vs heterogeneous spectral line broadenings in the neutron data. A similar situation is encountered in hydrogen adsorption in

graphite intercalates such as KC_{24} , where tunnelling transitions associated with a strong pinning of the H_2 molecule to the material give rise to spectral band heads at energies as low as 0.4-0.5 meV. These spectral features are further characterised by hyperfine splittings of a few μeV , which can only be partially resolved with current instrumentation [46], thereby hampering a full spectroscopic assignment of the neutron data. These limitations in spectral resolution will prove critical in the study of promising yet more complex hosts such as metal-decorated nanoporous carbons and metal-organic frameworks, both characterized by a multitude of adsorption sites and a fine balance between sequential chemisorption and physisorption steps in the mechanism of hydrogen uptake.

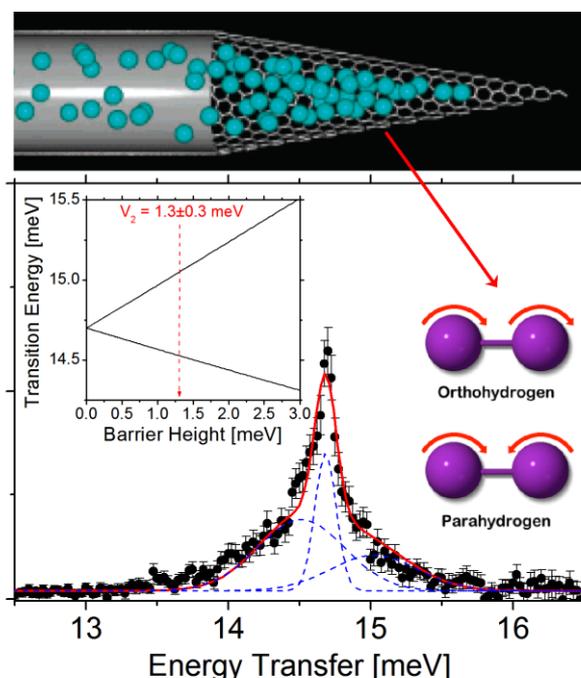


Figure 4: Inelastic neutron spectrum of hydrogen in carbon nanohorns. High energy resolution is required to differentiate between adsorption sites associated with the tubular and tip portions of the material, as shown in the cartoon above.

With its tenfold improvement in energy resolution, FIRES will constitute a step change in the exploitation of neutron spectroscopy to meet the scientific

challenges underpinning an otherwise formidable and yet unsolved technological problem.

Also within the remit of the Hydrogen Economy, new compounds which exhibit high hydrogen conduction are of considerable interest for applications in the burning of hydrogen fuel to generate electricity (fuel cells) and hydrogen sensing. Protonic conduction in perovskite-type oxides has been extensively examined, while similar information on hydride conduction in oxides is lacking. One such example is the transition metal oxide-hydride $\text{LaSrCoO}_3\text{H}_{0.7}$ [48]. This material is characterized by an exceptionally large molar conductivity compared to other proton conducting materials. QENS data shows that translational diffusion involves a jump process between lattice vacancies. Interestingly, the onset of diffusion coincides with the temperature range used for the synthesis of this material and, at temperatures above 700K, it occurs in conjunction with hydrogen loss from the sample. Tracking the onset of hydride diffusion around the onset is not possible at present, as it requires a logarithmic increase in energy resolution compared to that presently available on OSIRIS or IRIS.

In a similar vein, the study of low-frequency and stochastic motions in chemical hydrides has proven quite elusive to date. The ammonia boranes (e.g., NH_3BH_3) are isoelectronic with the hydrocarbons and can, therefore, store large quantities of hydrogen. Hydrogen desorption temperatures, however, are typically too high or too close to the decomposition temperature of the material. Also, the irreversible nature of the desorption step leads to unwanted (and toxic) by-products and a subsequent loss of recyclable material. LiNH_2BH_3 and NaNH_2BH_3 are the first two examples of a potentially large class of metal amidoborane compounds able to release molecular hydrogen at around 90 °C with no borazine emission [49]. The role of

soft vibrational modes involving the NH_2 and BH_3 functional groups in the stable crystal, or how these morph into diffusive motions leading to H_2 formation remains unexplored. To this end, it would be necessary not only to zoom in and out of the quasielastic region with a high neutron flux, but also to investigate energy transfers up to 10-20 meV as a function of temperature and H_2 release. FIRES will be able to achieve these stringent requirements better than any other instrument at ISIS.

Surface Science and Catalysis

Received wisdom dictates that neutron spectroscopy is a bulk condensed matter probe, with a sensitivity reaching millimolar levels only under very favourable circumstances (e.g., hydrogenous materials). Yet recent advances in materials science can be exploited to advance beyond this paradigm, as it is now possible to secure sufficient quantities of monodisperse nanomaterials exhibiting exceptionally large surface-to-volume ratios (100's m^2/cm^3). Low-energy neutron spectroscopy is thus confronted with the opportunity of becoming a powerful surface-sensitive technique, with immediate implications in chemical catalysis and the nanosciences.

Carbon supported platinum and platinum alloys are the most promising catalysts for low-temperature hydrogen-oxygen fuel cells. The advantages of platinum over other metals relate to its innate ability to dissociate molecular hydrogen as well as its superb resilience to oxidation and poisoning. The interaction of molecular hydrogen with platinum nanoparticles of size 2-8 nm is central to catalyst performance, the dissociation into chemically bound hydrogen atoms being a crucial yet poorly understood step in the overall electrochemical cycle. Inelastic neutron experiments on OSIRIS have probed the dissociation process and the subsequent identification of adsorption sites [50]. Key to the interpretation of these experiments was the ability to monitor both

the elastic and inelastic response up to energy transfers of ca. 20 meV. Owing to limitations in energy resolution, however, neither quasielastic broadenings of the adsorbed hydrogen phase nor the fine structure of the para-to-ortho rotational line were within the reach of these seminal experiments. For highly heterogeneous media, this information becomes very relevant for the design of more efficient catalytic materials.

Inelastic neutron scattering experiments probing the adsorption of methane on magnesium oxide (MgO) surfaces provide a textbook example of what is presently achievable on inverted geometry spectrometers like IRIS and OSIRIS [51]. Through sequential and carefully controlled layer-by-layer surface deposition, changes in the neutron tunnelling spectrum are the most direct probe of how the molecular film grows from two to three dimensions, and provides unprecedented insight into ubiquitous phenomena such as adhesion, corrosion, and heterogeneous catalysis. Moreover, the evolution of the interatomic potential can be tracked in detail and compared to ab-initio calculations, placing stringent restrictions on sound theoretical descriptions of adsorbate-substrate interactions. For larger or more complex systems (e.g., aromatic hydrocarbons, polar and hydrogen-bonded molecules) the need for improved energy resolution will become mandatory.

The chemical changes in host-guest catalytic media such as zeolites are both technologically relevant and a significant challenge to experimental and theoretical methods. As diffusion rates within the zeolite pores are critical to material performance, a sound understanding of the underlying physical chemistry is crucial to optimise a plethora of industrial processes including fluid cracking, hydrocarbon separation, or catalytic dewaxing. For the n-alkanes in nanoporous silicalite frameworks, it is known that activation energies for diffusion are low

for methane through n-hexane, but rise thereafter with increasing number of carbon atoms [52]. In conjunction with MD simulations, neutron data also indicate that these linear hydrocarbons tend to reside entirely within the interior of single-channel segments and diffuse through jumps across energetically less-favourable channel intersections. Longer molecules, on the contrary, straddle channel intersections and must undergo conformational rearrangements in order to diffuse. Most of these predictions await experimental confirmation and QENS is ideally suited for the task. As example, the fast rotational dynamics of butane and pentane isomers in MCM-22 has been recently investigated on IRIS [53]. Parallel MD simulations can reproduce the neutron data around 200K and also show how n-isomers indeed can rattle between cages more easily than their branched counterparts at temperatures more relevant for practical applications by the chemical industry ($T > 300\text{K}$). In this high-temperature regime, translational diffusion coefficients for these large molecules are still of order $\sim 10^{-11} \text{ m}^2\text{s}^{-1}$ and would therefore require the μeV energy resolution afforded by FIRES.

Superionic Conductors

Ionic conductors and mixed conductors are the basis of solid-state ionics, a rapidly growing field with many applications including batteries, chemical sensors, fuel cells, and electrochemical devices. The fast diffusion of ions through crystalline lattices is also a key factor in energy production and storage. The most fundamental property of solid-state conductors is the ionic conductivity. As this observable is directly linked to the diffusivity via the Nernst-Einstein equation, a microscopic understanding of stochastic processes necessarily involves QENS measurements over a wide momentum-transfer range. In addition to applications in hydrogen storage, the quest for better materials for solid-state batteries and solid-oxide fuel cells requires a

detailed characterization of the dynamics of structural defects associated with lithium- and oxygen-ion diffusion. As an example of the latter, solid-oxide fuel cells currently operate at 1000°C and rely on oxygen-conducting yttria-stabilised zirconia. While neutron scattering has already provided a fair amount of insight into the mechanisms of ionic conductivity [54], the availability of spectrometers with higher energy resolution will allow extension of current work by scanning a wider range of temperatures, of relevance to the optimisation of operating conditions.

From the standpoint of fundamental superionic conduction, knowledge of the temperature dependence of the ionic hopping mechanisms and pathways provides much-needed insight into those factors which promote their surprisingly high ionic conductivity. Such information will address key issues in this research area, such as the nature of correlations between diffusing ions. As an example, the study of the ionic transport mechanism in the mineral cryolite, a key ingredient in the industrial production of aluminium and a model system in the Earth sciences for diffusion in perovskites, needs a wide range in spectral resolution to cover changes in ionic mobility with increasing temperature [55]. In addition, the differentiation between competing diffusion scenarios requires a wide range of Q vectors. FIRES will meet both needs. A dynamic range of up to 1 ns also matches well the temporal window of MD simulations, which so far have proven essential to resolve specific questions about the nature of the diffusion process.

Confined Fluids

Liquids subjected to geometric confinement are ubiquitous in nature and industry. Interfacial phase transitions such as wetting, layering, and surface freezing and melting, or the effects of confinement on molecular transport coefficients and bulk transitions including superfluidity, capillary

condensation, and liquid-crystalline ordering constitute major scientific challenges both on the experimental and theoretical fronts, with immediate implications not only in physics, chemistry, and biology but also in the Environmental and Earth sciences.

How do, for example, pollutants travel through soil? QENS has been the method of choice to investigate molecular mobility inside clay sheets under physico-chemical conditions closely mimicking those found in nature, e.g., temperature, pressure, pH, or ionic strength. Industrial and household waste often contains toxic hydrocarbons including small organics like methanol and glycol, as well as a whole range of carcinogenic cyclic aromatics like phenol, paraquat, or nitrophenol, the latter being a trace species commonly found in fertilizers. In spite of the extreme two-dimensional confinement of smectite and vermiculite clay platelets, translational diffusion of the most mobile organic molecules is found to be surprisingly close to that of the bulk [56]. With current instrumental capabilities, however, it has only been possible to explore very restricted regions of the physical and chemical phase diagrams of these complex ternary systems, up to temperatures and pressures typically found 5-10 km below the Earth's crust or solute concentrations close to the saturation limit. In particular, the role of intermolecular interactions leading to aggregation phenomena would require access to rotational correlation times of at least hundreds of picoseconds. Also, a significantly better energy resolution is necessary to study larger molecules, like the asphaltenes, or extend current studies to higher pressures and temperatures corresponding to tens of km into the lithosphere. This latter aspect not only has implications in waste remediation but also in the development of increasingly efficient methods for crude-oil extraction.

Methane clathrates (or 'methane ices') are solid forms of water containing large

amounts of trapped methane. Originally thought to occur only in the outer regions of the Solar System where temperatures are low and water ice is common, very large deposits of methane ice have also been found under sediments on ocean floors. High-resolution neutron spectroscopy is ideally suited to explore the binding and phase stability of this important host-guest system. Owing to its tetrahedral point-group symmetry, methane's rotational and librational energy-level structure splits in subtle yet predictable ways according to the site symmetry and strength of the underlying intermolecular potential inside the clathrate cage. Typical spectral splittings range from μeV to meV and spread out over a wide range of energy transfers (up to ~ 1 meV for freely rotating methane). These measurements are exceedingly difficult to accomplish on backscattering instruments at reactor sources yet they are well within the reach of FIRES.

Metallic and Electronic Liquids

Metals and alloys have important applications in the aerospace and automobile industries. In particular, AlNi alloys are being studied as the primary constituent of gas turbines because they are lighter than standard Ni-based superalloys. Solidification processes in such materials involves a pronounced stretching and slowing down of relaxation times. The chemical short-range order observed in AlNi melts and the microscopic dynamics on cooling below the melting point have recently been investigated with electromagnetic levitation in conjunction with isotopic substitution techniques. Using QENS to explore in detail the relaxation processes occurring over a wide temperature range requires a higher energy resolution than what is currently available on conventional time-of-flight spectrometers.

From a fundamental point of view, metallic melts can also be regarded in many cases as hard-sphere-like and are prime systems

for answering pending questions in modern mode-coupling theories of liquids. Deep supercooling and, consequently, the dramatic slowdown of relaxation times are important data for comparison with theoretical predictions. One intriguing question is whether there is a unique temperature range at which solidification starts at the atomistic level. Motions on nearest-neighbour length scales are very sensitive to this process; at distances corresponding to the peak in the structure factor, atomic motions slow down and the associated QENS energy widths become narrower, as originally predicted by de Gennes. In the supercooled state, this process is even slower and eventually comes to a halt as the glass transition is approached from above. To access these timescales, high energy resolution is required, combined with the good resolution in momentum transfers FIRES will provide.

'Electronic solutions' are another interesting and very relevant case. These are formed when a metal dissolves in polar media, the prototypical solvent being liquid ammonia. The dissolution process releases electrons into the liquid giving rise to beautifully coloured conducting solutions. The presence of solvated electrons results in composite liquids of extraordinary properties including: very low density and viscosity, high electrical conductivity, a deep pseudoeutectic (giving rise to the lowest temperature liquid metals ever observed), concentration-driven metal-nonmetal transitions, liquid-liquid phase separation, highly conducting glassy phases, exceptionally high redox reactivity, and liquid-state superconductivity. Recent neutron and X-ray scattering experiments have elucidated the structure and dynamics of these remarkable liquids, including electron solvation, the appearance of vibrationally dressed electron states such as polarons and bipolarons, and electron delocalisation [57]. An increase of the self-diffusion coefficient with metal concentration has been attributed to the competing

influence of electron and ion solvation. Much remains to be explored, however, about how ion and electron mobility in these systems can be further exploited or controlled to drive novel chemical processes such as graphene dissolution, charging, and intercalation. To this end, the ability to map the stochastic dynamics of these materials becomes a mandatory step. With its finer energy resolution, FIRES will open up entirely unexplored areas in the compositional phase diagrams of these materials, particularly at low metal concentrations or as critical points are approached.

3. Technical Considerations

3.1 Outline Design Specification

Attaining high spectral resolution at a pulsed neutron spallation source like ISIS requires tight neutron pulses in the time domain. For inverted-geometry spectrometers, this requirement can be satisfied by positioning the instrument on either a decoupled or a poisoned neutron moderator. As a result of this choice, incident flux is compromised and fixed by construction. The recently commissioned BASIS spectrometer at the SNS (USA) has been designed and built on such a predicament.

A hitherto unexploited alternative involves the combination of an intense (e.g. coupled) cold neutron moderator and a fast chopper to produce well-defined neutron pulses of variable duration. Such a “pulse-shaping” device allows for straightforward changes to instrumental resolution by altering the neutron pulse width. Moreover, spectral resolution can be traded against intensity. This novel approach to inverted-geometry neutron instrumentation lies at the heart of the FIRES design concept.

The use of pulse-shaping choppers necessarily restricts the useful spectral

range because this parameter is determined by both the width of the moderator pulse and the distance from moderator to chopper. As detailed in Ref. [58], a suitable dynamic range and incident flux require both a coupled moderator and a short distance to the pulse-shaping chopper. Since the dynamic range is necessarily restricted by the chopper-moderator arrangement, a high-repetition-rate source such as ISIS Target Station I is the natural choice in order to maximise incident neutron flux.

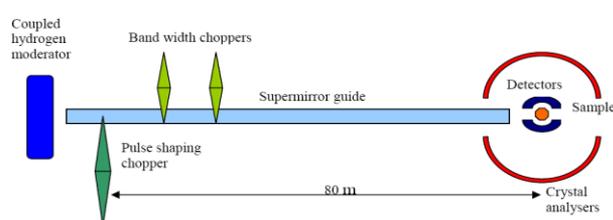


Figure 5: Schematic diagram showing the main components of the FIRES instrument.

Figure 5 shows a schematic diagram of the FIRES instrument. Neutrons travel along a supermirror guide for ca. 80 m before they are focused onto the sample position. Additional “bandwidth” or “order-selection” choppers prevent frame overlap between consecutive neutron pulses. The secondary spectrometer consists of a 2-meter-high patchwork of Si(111) crystals, placed at a distance of 2.5 m from the sample and covering an angular range of 10-150°. The corresponding momentum-transfer range amounts to $0.18 \leq Q \text{ (\AA}^{-1}\text{)} \leq 1.95$, in addition to a solid-angle coverage approaching 2 steradian on each side of the secondary spectrometer. Use of the Si(333) reflection will provide a three-fold increase in the available Q range. Position-sensitive detector technology is exploited to enhance energy resolution by reducing flight-path uncertainties. In combination with a guide insert, this detector array provides the requisite Q resolution for single-crystal experiments (for examples, see Section 2). The spectrometer will also have a high-

angle diffraction bank, thus mimicking current capabilities on OSIRIS and IRIS to perform simultaneous high-resolution diffraction and spectroscopic measurements.

The pulse-shaping chopper would be ideally located 6 m away from the spallation target, and controls the neutron pulse width. As explained earlier, this component can also be used to trade resolution against intensity. Flux gain factors of 4-5 are expected at the expense of modest changes in spectral resolution. An additional advantage of a pulse shaper stems from its highly symmetric triangular line shape, providing a clean and easy-to-model instrument response function.

In terms of spectral resolution, a chopper pulse width of 50 μs gives rise of an energy resolution of $\Delta E = 2.4 \mu\text{eV}$ for elastic scattering, whereas use of the full moderator pulse (width 200 μs) would result in $\Delta E = 6.5 \mu\text{eV}$ (see Figure 6). The spectral resolution at inelastic energy transfers is also outstanding: at the free-rotor line of molecular hydrogen (14.7 meV), it amounts to $\Delta E = 38 \mu\text{eV}$, and at the roton line of superfluid ^4He (0.74 meV), the energy resolution is $3.3 \mu\text{eV}$. For further details of the calculations see Ref. [58].

Simulations predict a useful dynamic range, R , of 250 μeV for single-pulse operation. This range is about one hundred times greater than the energy resolution, that is, $(R/\Delta E) = 100$. FIRES therefore outperforms other modern backscattering instruments characterised by $(R/\Delta E) = 60-80$. When operating without the pulse-shaping chopper, the dynamic range is determined by the source frequency and the length of the instrument, which results in a spectral range of about 600 μeV .

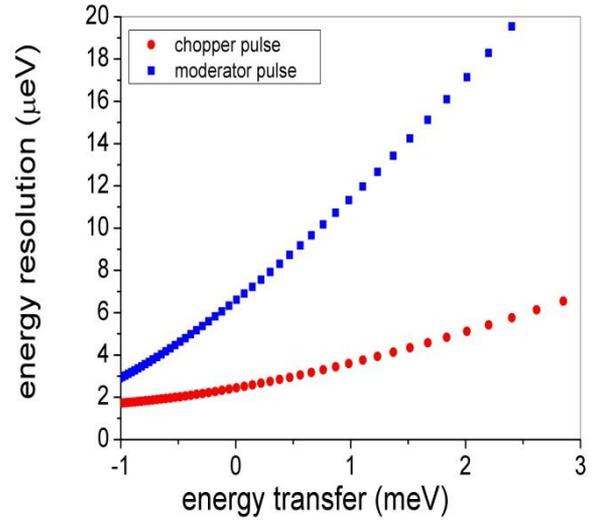


Figure 6: Calculated energy resolution for chopper pulse widths $\Delta t = 50 \mu\text{s}$ and $\Delta t = 200 \mu\text{s}$. For further details see the text.

Table I: Summary of Technical Specifications.

Moderator	Coupled hydrogen ($T=20\text{K}$)
Guide	80 m supermirror guide
Secondary spectrometer	Si(111) analysers Near backscattering: $\theta_B = 87.3^\circ$ Height: 2m Radius: 2.5 m Solid angle: 1.9 sr Beryllium filter 2 position-sensitive detector banks
Energy resolution	2.4 μeV with chopper pulse 6.5 μeV with moderator pulse
Spectral flux at sample position	$4 \times 10^4 \text{ n/cm}^2/\text{s}/\mu\text{eV}$ in high resolution mode

3.2 Layout considerations

The primary spectrometer consists of two major components, namely, the supermirror guide and the choppers. Monte Carlo simulations with the McStas simulation package have been performed for the design of a long supermirror guide [59]. One option explored in this work consists of a curved ballistic guide with an elliptic focusing section at the end (see Figure 7). It is important to note that this guide design leaves the dimensions inside the biological shield unchanged. A gain in intensity at the

sample position of a factor two compared to a pure ballistic guide design was achieved. Moreover, the elliptic focusing section resulted in a more homogenous divergence profile.

Further gains in intensity are expected for a full elliptical guide starting inside the biological shield. The dimensions of this elliptic guide would be quite large, with a maximum height of 35 cm and a maximum width of 24 cm. Preliminary results indicate a further gain factor of 3 compared to the ballistic/elliptic design presented above. This elliptic-guide configuration excludes a curved section, therefore a beam blocker would be necessary to remove prompt high-energy neutrons from the spallation target.

Pulse shaping will be achieved with a counter-rotating double-disc chopper running at a maximum frequency of 300 Hz. At this speed, a neutron pulse width of 50 μ s can be achieved with a slit width of 60 mm. This width is compatible with all potential guide designs considered to date. The fast chopper should be positioned as close as possible to the moderator. At ISIS Target Station 1, a distance of ca. 6 m from a coupled hydrogen moderator would quite conveniently place the pulse-shaping chopper just outside the biological shield, resulting in a dynamic range of about 250 μ eV. Given the length of the primary spectrometer and the source repetition rate, this dynamic range could be extended to ca. 600 μ eV. In order to exploit in full the whole dynamic range provided by the source, several pulses within a single frame could be created by the pulse shaping chopper, a technique called 'Repetition Rate Multiplication' (RRM). For an indirect spectrometer, RRM provides a straightforward extension of the dynamic range. Applying RRM will extend the dynamic range to the maximum provided by the source repetition rate. After the fast pulse shaping chopper, further choppers to restrict spectral bandwidth and avoid frame overlap will also be required.

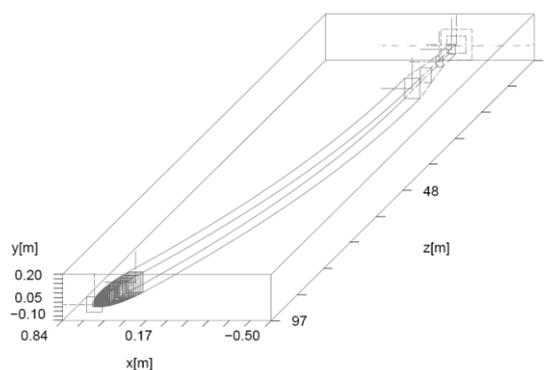


Figure 7: Curved ballistic guide combined with an elliptic focusing section.

The secondary spectrometer houses two large silicon analysers and detector banks (see Figure 8). The whole analyser assembly will sit inside a vacuum tank. The analyser bench is made of silicon wafers (Si 111) glued onto aluminium frames. The distance from the sample to the analysers is 2.5 m. To minimise flight-path uncertainties, the analyser bench is split vertically into two sections. Each section will focus scattered neutrons onto separate detector arrays consisting of position-sensitive Helium tubes. Helium tubes with a diameter and spatial resolution of 8 mm are envisaged. Position sensitivity will allow access to out-of-plane scattering.

The results of a geometry optimisation of the secondary spectrometer using 3D computer-aided design tools are shown in Figure 9. To enable the use of bulky and complex pieces of sample environment (e.g., high-field magnets, high pressures), a sample bin diameter of 600 mm has been incorporated to the design from the outset. Equally important is the need for a high Bragg-reflection angle off the Si analysers, in order to approach the backscattering condition in the secondary spectrometer.

As shown in Fig. 9, this Bragg reflection angle has been set at 87.3° , along with a

detector-array radius of 350 mm, and Helium-detector tubes of length 170 mm. This detector arrangement provides an angular resolution of 1.3° . A good momentum transfer resolution, necessary in some magnetic excitation experiments, can then be achieved by combining position-sensitive detection with a restricted incoming divergence. In addition, a Beryllium filter suppresses higher-order analyser reflections to provide access to higher energy transfers.

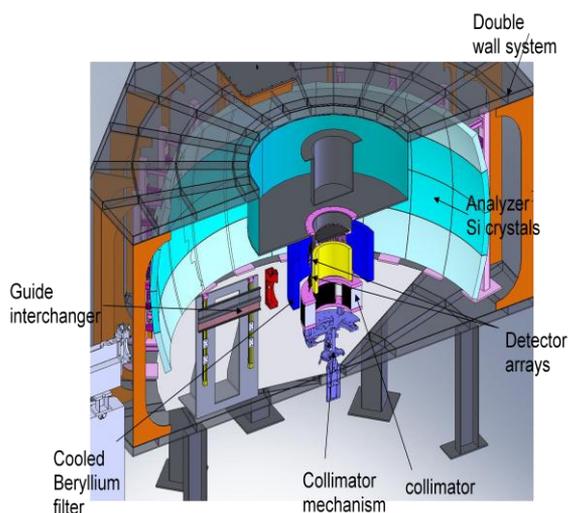


Figure 8: Layout of the secondary spectrometer.

In addition to spectroscopic capabilities, a diffraction detector bank consisting of position-sensitive Helium tubes has been accommodated at a scattering angle of ca. 170° . This detector will provide simultaneous high resolution diffraction data.

There are several options for the second side of the analyser array. It could house a different analyser crystal such as Si(311), to extend the Q-range of the instrument. Alternatively, use of the same Si(111) crystal would result in a twofold increase in count rate.

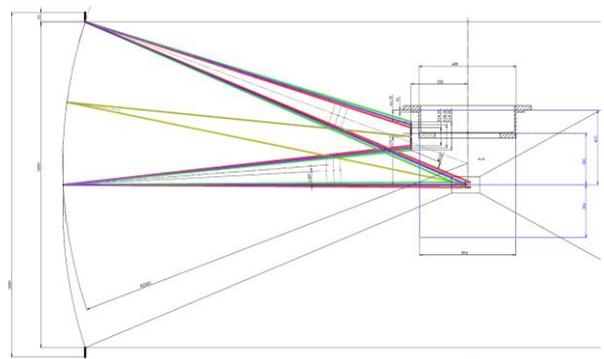


Figure 9: Plane projection showing the optimised dimensions of the analyser and detector banks in the secondary spectrometer. For a three-dimensional view, see Fig. 8.

3.3. Comparison with other Spectrometers

As detailed in Section 2, FIRES represents a much-needed addition to the existing ISIS instrument suite and science programme. At present, IRIS achieves an energy resolution in the μeV range via the use of the Mica (002). This option, however, requires 20 Å neutrons, thereby leading to a very unappealing incident neutron flux as well as severely restricted energy- and momentum-transfer ranges ($R=20$).

Compared with OSIRIS, FIRES will offer an order of magnitude higher energy resolution. Direct-geometry, time-of-flight spectrometers such as the new LET instrument at ISIS can, in principle, achieve μeV resolution by using long wavelengths. This approach, however, has immediate drawbacks in terms of intensity and momentum transfer range. Most of the science examples described in this document would suffer considerably or would be simply intractable due to these technical restrictions. A useful figure of merit to compare different spectrometers is the relative energy resolution $\Delta E/E$. Direct geometry time-of-flight spectrometers can achieve 0.01, whereas FIRES approaches 10^{-3} .

At reactor-based neutron sources, direct-geometry backscattering instruments remain the workhorse for high-resolution spectroscopy ($\sim 1 \mu\text{eV}$). Examples include IN10 and IN16 at ILL (France), as well as modern backscattering spectrometers exploiting phase-space-transformation techniques, e.g., Spheres at FRM-II (Germany), HFBS at NCNR-NIST (USA), or soon IN16B at the ILL. These instruments can yield a spectral flux of $1\text{-}2 \times 10^4 \text{ n/cm}^2/\text{s}/\mu\text{eV}$. Since all the aforementioned instruments have a similar solid-angle coverage in the secondary spectrometer, FIRES is in a position to deliver at least twice the count rate than the best backscattering instruments on reactor-based neutron sources, at the expense of a relaxed energy resolution due to its operation under near-backscattering conditions.

High energy resolution at inelastic energy transfers is difficult to achieve on reactor-based backscattering spectrometers. The approach works sometimes, as on IN10, but the choice of energy-transfer values is discrete and very limited. A wide and flexible choice in dynamic range, in particular for inelastic measurements, is the main advantage of a time-of-flight backscattering instrument like FIRES compared to reactor-based backscattering instruments. In this way, FIRES will be a unique tool to address totally unexplored scientific challenges.

Moreover, the study of dispersive excitations with high energy resolution (of order of few μeV) has become possible in the last few years with the development of neutron-resonance spin-echo (NRSE) techniques on three-axis spectrometers: ZETA at the ILL and TRISP at FRM-II. These challenging high-resolution measurements are performed in the time domain. FIRES measures in the energy domain and, therefore, gives access to the full spectral line shape. Hence, it is a more powerful tool to study excitation continua and gaps, as well as the fine and subtle

splittings that arise from homogeneous or inhomogeneous spectral broadenings. Also, zero-field spin-echo techniques are restricted to very low magnetic fields at the sample position, a limitation easily circumvented on an indirect-geometry spectrometer at a pulsed spallation neutron source.

The need for such novel high-resolution backscattering instruments has already been recognised by the two new spallation sources SNS (USA) and JPARC (Japan). The new backscattering instrument BASIS at SNS, uses a poisoned moderator to create short-time pulses. This approach to neutron production is costly. Our calculations indicate that FIRES on the current ISIS Target Station I can provide a similar flux as BASIS operating at a source power of 1 MW. The main reason behind this difference in instrument performance can be traced back to the higher peak flux of a coupled hydrogen moderator. Another distinct advantage of FIRES over BASIS is the flexibility offered by the pulse-shaping chopper to trade resolution against intensity. A factor of 4-5 in flux can be gained by using the full moderator pulse width at the expense of a factor of less than 3 in resolution.

A very similar conceptual design for an inverted-geometry instrument as that presented here for FIRES has been proposed (and is now under construction) at J-PARC in Japan. The DNA-spectrometer is virtually identical to FIRES, except that it will use a shorter primary flight path. A shorter flight path, however, will require tighter pulses from the pulse shaping chopper to achieve the same energy resolution.

4. Potential sites for FIRES

FIRES requires a cold coupled moderator for two reasons. First, a coupled moderator delivers long neutron pulses, directly

influencing the useful dynamic range of the instrument. Second, a high peak flux ensures a much-improved performance compared to decoupled or poisoned moderator concepts such as those already used in state-of-the-art backscattering spectrometers like BASIS at the SNS. This technical requirement necessarily implies an upgrade of the current ISIS Target Station 1 (TS1) to include a new coupled hydrogen moderator and, potentially, a redesign of the reflector assembly.

Possible beamlines which are currently viewing the hydrogen moderator on TS1 and which might be freed in the near future include LOQ or the CRISP/SURF instruments. In either case, the length of the FIRES instrument (about 90 m from the moderator) would bring the secondary spectrometer well into the Harwell site. Access to Harwell is likely to be a lengthy and costly process, as it necessarily involves de-licensing protocols.

A redesign of ISIS TS1 opens up new possibilities. One such possibility is to provide access to the hydrogen moderator from the South side of the target station. Using a beamline on this side would position the instrument at a similar distance from the target as the HRPD diffractometer. This solution would resolve issues related to access to the Harwell site. Nonetheless, it must be stressed that both possibilities require an upgrade of ISIS TS1 to accommodate a new coupled hydrogen moderator.

A third possibility is to place FIRES in the newly constructed ISIS Second Target Station (TS2). Here, a coupled cold moderator exists and there is also room for a long beamline. TS2 operates at 10 Hz, hence at a repetition rate 4 times lower than TS1. From a scientific viewpoint, many high-resolution experiments do not require the concomitant large dynamic range (nearly 3000 μeV) attainable on TS2. Assuming that an upgraded coupled hydrogen moderator

on TS1 has a similar performance to that in TS2, the two TS1 options considered above still offer a factor 4 higher intensity than TS2. On TS2, the wide dynamic range combined with the constraints imposed by a pulse-shaping chopper can allow for more pulses using the RRM technique. With 10 or more pulses accommodated within one source pulse on TS2, this option would require a more sophisticated chopper-slit design.

Concluding Remarks

FIRES is a novel and flexible backscattering neutron spectrometer offering a wide range of flux and resolution options. As such, it constitutes a step change in low-energy neutron spectroscopy at the ISIS Facility in the medium and long terms.

In terms of spectral resolution, FIRES will enlarge our energy window by one order of magnitude, leading to new scientific opportunities in a wide range of research areas. A judicious combination of high energy resolution and wide dynamic range provides a unique neutron instrument for high-resolution spectroscopy. With these properties FIRES will be complementary to reactor-based backscattering instruments and will in turn open new scientific opportunities outlined in the scientific case.

Three potential sites have been identified for this new spectrometer. The preferred option is a beamline at the south side of an upgraded Target station I.

With the science case and technical specification presented herein, the FIRES project has reached the level of maturity necessary to take this instrumentation initiative a step closer to reality.

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