Developments in instrumentation and science at the ISIS pulsed neutron source.

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Abstract

New neutron scattering instruments at ISIS of interest to the disordered materials community include: GEM, the new diffractometer for both powder and disordered structure measurements; OSIRIS, a project for high resolution measurements, both dynamic and structural, using both unpolarised and polarised neutrons and an upgrade to IRIS, the quasielastic spectrometer. There are also plans for the provison of a second target station.

Data from GEM taken during its commissioning show that it is ideally suited for in-situ crystallization measurements as well as amorphous structures. New amorphous structures measured on the LAD and SANDALS diffractometers include ion conducting glasses and polymers, and amorphous metallic alloys. In the field of quasielastic neutron scattering, results are presented on: the diffusive dynamics in a quasicrystalline alloy; the dynamics of polymer electrolytes; and diffusion in fast ion conducting glasses.

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1. Introduction

This paper provides a brief review of developments in instrumentation and new science at the ISIS Pulsed Neutron Source which are of interest to the disordered materials community. The second section looks at three instruments, the third section describes three new areas of science, while the last, brief, section looks to the future.

2. Instrument developments

The instrument portfolio at ISIS is continually changing: many instruments are improved; some of the older instruments are replaced; and new instruments built. As examples, three instruments have been selected as being of interest for disordered materials : GEM representing a replacement, OSIRIS as new instrument and IRIS as an upgrade.

2.1 GEM

The Liquids and Amorphous Diffractometer (LAD) was the first diffractometer to be built for ISIS and was removed at the end of 1998 to make way for the GEM diffractometer. The basic objective of the GEM design is to provide an instrument which combines high resolution over a wide Q range with the high count rate required to study samples under varying conditions such as temperature, pressure and chemical reaction. Experience at ISIS has demonstrated that time-of-flight diffraction gives the benefit of access to a very wide range of d-spacings (or Q) with constant resolution for all $\Delta d/d$ (or $\Delta Q/Q$). In addition, fixed detector geometry simplifies the provision of specialised sample environments. The special features of GEM include a very large detector area, high degree of collimation of both incident and scattered beams and high detector stability over a large solid angle. GEM has been designed to be both a good-resolution, high-flux powder diffractometer for crystalline samples, and a wide-angular-range, high-flux, low-background instrument for disordered materials studies. The combination of high count-rate and good resolution will advance areas such as magnetic diffraction, isotopic difference experiments, time-resolved and small sample diffraction. Following a period of scientific commissioning, GEM is now a fully scheduled instrument. As part of the commissioning, one of the experiments involved in situ crystallization from an amorphous precursor on samples previously measured on a reactor source. High quality data were collected at 3 minute intervals and showed better resolution at high-Q (using the backward angle detectors) than the reactor data.

2.2 OSIRIS

The OSIRIS project will explore the instrumental horizons available with the cold neutrons from a pulsed source and especially the totally new avenues available to polarised neutrons on these sources. On pulsed sources, polarisation techniques offer great potential for high resolution studies, and lack of opportunity and technical developments has left the field unexploited. By exploiting the combination of sharp pulses, white beams and cold neutrons from ISIS, high resolution measurements, both dynamic and structural, can be carried out using both unpolarised and polarised neutrons. Following on from commissioning of the diffraction detectors in 1998, during 1999 the instrument was scheduled to 50% for diffraction. The rest of the time was used for polarisation developments and the building of the inelastic section.

The polarised neutron option will open up new fields in pulsed neutron scattering such as magnetic scattering and allowing the separation of the coherent and incoherent scattering.

2.3 IRIS

IRIS, the high-resolution quasielastic spectrometer, was another of the first instruments on ISIS. It has two analyser systems - graphite and mica - and the project to replace the graphite analyser with a larger, helium-cooled, analyser is nearing completion. This should give a factor ~ 2.5 increase in count-rate with an associated improvement of signal-background from 1350:1 to 5000:1. This will greatly improve the quality of data obtained from nonhydrogenous materials such as quasicrystals (section 3.1) or increase the throughput of hydrogenous materials such as polymers (section 3.2).

3. New science

Over the years the science programme on ISIS has also changed with some areas disappearing to be replaced by new fields especially those with possible technological applications. Three examples of new science are highlighted : two have technological links related to ionic conduction - polymer electrolytes and fast ion conducting glasses.

3.1 Quasicrystals

Recently the first experimental evidence for atomic hopping in a quasicrystalline structure was found [1]. Using the isotope substitution technique it was possible to conclude that the observed quasi-elastic scattering components in icosahedral $Al_{62}Fe_{25.5}Cu_{12.5}$ were originating from the motions of both Cu and Fe atoms and that these motions were confined in space.

Since then there have been further results using quasielastic neutron scattering experiments on the $Al_{50}Cu_{35}Ni_{15}$ [2]; a system that can be considered as a one-dimensional quasicrystal. Measurements were carried out both on the IRIS spectrometer at ISIS, RAL, and at the MIBEMOL spectrometer at LLB, Saclay. In the IRIS measurements three $Al_{50}Cu_{35}Ni_{15}$ samples were investigated, one containing natural elements, the second containing natural Al and Cu and 'null matrix' Ni, and the third containing natural Al, the copper-65 isotope and 'null matrix' Ni.

The results show that frequent jumps of Cu and Ni atoms on different time scales are taking place in the $Al_{50}Cu_{35}Ni_{15}$ alloy at high temperature between different interstitial lattice positions. As this alloy has a deformed cubic structure, this finding indicates that hopping motions are not restricted to occur in systems with quasicrystalline local order but is probably generally present in Al-based intermetallics at high temperature.

3.2 Polymer electrolytes

The relation between mechanical and electrical relaxation in polymer/lithium-salt complexes is a fascinating and still unresolved problem in condensed-matter physics, yet has an important bearing on the viability of such materials for use as electrolytes in lithium batteries. At room temperature, these materials can be biphasic: they consist of both fluid amorphous regions and salt-enriched crystalline regions. Ionic conduction is known to occur predominantly in the amorphous fluid regions. Although the conduction mechanisms are not yet fully understood, it is widely accepted that lithium ions, coordinated with groups of ether oxygen atoms on single or perhaps double polymer chains, move through re-coordination with other oxygen-bearing groups. In PEO-LiTFSI, the formation and disruption of these coordination bonds must be accompanied by strong relaxation of the local chain structure [3]. Quasielastic neutron scattering on the PEO/lithium-salt electroluytes probes the relaxation on a nanosecond timescale, and shows [4] that at least two processes are involved: a slow process with a translational character and one or two fast processes with a rotational character. Whereas the former reflects the slowing-down of the translational relaxation commonly observed in polyethylene oxide and other polymer melts, the latter appears to be unique to the polymer electrolytes and has not been observed before. A clear picture emerges of the lithium cations forming crosslinks between chain segments and thereby profoundly altering the dynamics of the polymer network.

3.3 Fast ion conducting glasses

The understanding of the microscopic mechanism for ion conduction in glasses is a long-standing problem in glass science. Several properties of ion conducting glasses, such as the mixed alkali effect, the immobile salt effect and the non-Arrhenius temperature dependence of the conductivity, are not well understood. In order to get a structural basis for understanding these properties, neutron diffraction experiments performed on LAD at ISIS have been combined with x-ray diffraction experiments and reverse Monte Carlo modelling to produce microscopic models of the complex structures of ion conducting glasses [5].

From the structural investigations of $A_xB_{1-x}PO_3$ glasses (A,B=Li,Na,Rb) the mixed alkali effect can be qualitatively understand and shown to be a natural consequence of the fact that the two kinds of alkali ions have distinctly different local environments, which are preserved in the mixed glasses. This leads to a large energy mismatch for cationic jumps to dissimilar sites and thus makes such jumps unlikely to occur at room temperature. Furthermore, the two kinds of ions are randomly mixed with no particular preference for pairing or clustering of similar or dissimilar ions. However, the overall distribution of alkali ions (independent of type) is not entirely random - rather the ions are located in thin low-dimensional pathways which run between the phosphate chains. These pathways are partially blocked and so the long range mobility (i.e. the d.c. conductivity) is reduced considerably compared to the corresponding single alkali glasses.

4. Future prospects

As already mentioned the instrument portfolio at ISIS is continually changing, but the most exciting prospect for ISIS is the plan to provide a second Target Station. This project, if funded, would provide a new suite of some 18 instruments optimised to use long wavelength neutrons.

In addition, the RAL site will also see the arrival of the new synchrotron (DIAMOND) funded jointly by the Wellcome Trust and the UK and French governments. RAL will then become a centre of excellence for the study of condensed matter using a range of experimental techniques.

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