Beamline Name: **NIMROD**

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**This proposal requests a Near and InterMediate Range Order Diffractometer (NIMROD) as a day one instrument on the Second Target Station at ISIS. Uniquely, NIMROD will provide continuous access to particle separations ranging from the interatomic (< 1Å) through to the mesoscopic (> 300Å). This is the characteristic dimension of fullerenes and small protein molecules, *i.e.* the true nanoscale. As such, the instrument will open up major opportunities for novel and timely science, in areas where the primary aim is to relate molecular-level structure to the phase and function of materials.**

The properties of many scientifically and technologically important materials arise from a subtle balance between short-, medium- and long-range interactions. Traditionally the structural correlations on these length scales are probed using separate wide- and small-angle diffraction experiments. TS-II at ISIS now provides a unique opportunity to build a diffractometer that can probe a broad range of structural correlations simultaneously. This approach makes possible the development of a coherent picture of the complex relationship between structure and properties. The rationale is to relate changes in local molecular environment to larger scale processes, such as protein folding in solution, confinement in microporous media, and phase behaviour and nucleation. In summary:

- NIMROD is a unique instrument specification, which relies on the longer wavelengths of TS-II to increase the upper limit of the accessible correlation lengths, while also extracting atomic resolution from the shorter wavelengths. The instrument bridges the traditional gap between SANS and wide-angle neutron scattering, by using a common calibration procedure for all $Q$-scales. The data obtained from NIMROD will therefore enable the development of detailed and well-constrained models of complex scattering systems.
- NIMROD is backed by a broad-based, internationally recognised, user community. For example, in the recent International Assessment of University Research in Chemistry in the UK (EPSRC & RSC, 2003) its use of central neutron facilities was highlighted: "*UK chemists were among the first worldwide to exploit the power of neutron scattering and synchrotron radiation to probe the behaviour of complex systems: for example, the structure and dynamics of aqueous solutions, ionic liquids, and polymers at interfaces. Work in these areas continues to be competitive internationally*."
- NIMROD will strengthen the synergy between experiment and theory, and is supported by internationally leading theoreticians. Importantly, the instrument covers length scales that are only now being viewed as accessible to atomistic computer simulations. In this context, we note that our community is responsible for recent advances in quantitative data analysis of diffraction from disordered materials. Techniques such as Reverse Monte Carlo (RMC) and Empirical Potential Structure Refinement (EPSR) allow us to produce real-space molecular models of complex systems, which can be compared directly with the experimental data.

The beneficiaries from the design of this instrument include the principle scientific disciplines, as well as more applied areas such as chemical engineering, oil and gas recovery, environmental science, renewable energy, separation technology, food science, biomaterials and pharmacology.

**Scientific Case:**

NIMROD will enable new science and technology wherever the molecular and mesoscale structures of disordered, or partially disordered, materials are related to their properties and function. As such, the case for this instrument underpins the core science areas of TS-II: Advanced Materials, Soft Condensed Matter, Biomolecular Sciences, and Chemical Structures, Kinetics and Dynamics. In each of these areas, potential users of NIMROD describe specific new examples relevant to their research interests. This demonstrates broad-based enthusiasm for the instrument, and in the context of the TS-II timescale these letters of support also provide a forward-look into growth areas served by NIMROD.

It is clear that understanding of disorder over the NIMROD length scales will, in the future, become increasingly necessary to resolve major scientific and technological challenges. Core issues centre on the behaviour of biological molecules in solution, and the need to control the properties of increasingly complex liquids, glasses and composite/template materials. In each case, the sub-Ångstrom resolution structural data available from NIMROD are a prerequisite to detailed understanding. Further opportunities will stem from the ability of the instrument to exploit site-specific isotope substitution on dilute species, to first and second order difference levels. For example, measurement of the local and mesoscopic structure of the proton environment via H/D labelling is a key to understanding hydrogen-bonding and associated liquids, and in developing hydrogen storage media.

## Complex and Confined Liquids

Complex fluids and solutions are ubiquitous in science and technology: NIMROD offers outstanding new opportunities for the study of these systems. We anticipate major drives to understand and control the solvent mediated interactions that lead to solute association and conformational changes, and to design chemically selective solvents. Further impetus is given by the recent synthesis of nanoporous media of well-defined geometry, which opens up new possibilities in studying important effects in confined fluids. These substrates have dual status as an arena in which to investigate novel physical and chemical phenomena in reduced-dimensions, and as a test-bed for storage, separation, release, and catalysis technology. All of these areas demand structural information on the nanometer length scales that are targeted with NIMROD. When allied with high stability detector arrays, the instrument realistically opens the possibility for routine studies of ternary and higher complexity molecular mixtures, at chemically and biochemically relevant solute concentrations.

- ***Biological Molecules in Solution.*** NIMROD will excel in structural studies of biomolecules in solution, and will supersede the currently available worldwide suite of neutron instruments. It would, for the first time, enable us to relate changes in solvent structure directly to biosolute conformation. In a recent example of such work, performed on SANDALS, it was shown that pressure induced unfolding of myoglobin in aqueous solution is associated with changes in the water structure. However, this work was frustrated by the upper length-scale accessible on the instrument, a limit that would be significantly increased on NIMROD. Another example of biomolecular relevance that has recently captured the interest of the user community is the study of disaccharides, and their utilization as bioprotectants through effects such as glassification.
- ***Confined Fluids.*** A large number of natural and industrial processes depend on the properties of confined fluids, with numerous fundamental and practical questions remaining unanswered. When combined with new classes of nanoporous materials, such as MCM silicas, studies on NIMROD are likely to be pivotal: all the relevant length-scales are accessible to this one instrument. Confinement allows one to deeply undercool liquids, and thereby to enter regions of the phase diagram inaccessible to the bulk liquid. The community will exploit this to probe the role of structural frustration and adsorption of systems as diverse as complex liquids, glass-formers, polymer melts, and hydrogen. High-resolution (sub-Ångstrom) studies of the solid-liquid interface are planned within the two-dimensional pores of lamellar hosts, such as clays. Crucial questions

centre on the nature and extent of layering and solvent density. Here again, the ability of NIMROD to extend atomic resolution over hundreds of Ångstroms will lead to qualitatively new science.

- ***Electronic Liquids.*** Current understanding of electron localisation/delocalisation in the liquid state is hampered by a dearth of structural information over the length-scales that are crossed as one moves from an insulating (electron localised) to conducting (electron delocalised) state. The study of electronic liquids, such as metal-ammonia type solutions or liquid semiconductors, is therefore an area in which NIMROD will play a leading role. Furthermore, this instrument would allow us to probe directly the structure of the exotic species that exist in these systems, for example polarons, bipolarons, electron channels, and excitonic atoms.
- ***Ionic Liquids and Mesophase Systems.*** Room temperature ionic liquids are an environment friendly medium, of very low vapour pressure, in which one can control the selectivity of many organic reactions. To date, detailed structural studies of such liquids have been limited to short chain materials. NIMROD would enable us to study much longer chain lengths, thereby giving access to new classes of ionic liquids. For example, by increasing the amphiphilic nature of the cation, ionic liquid crystals can be formed. These have extensive mesophase ranges, and are thermally very stable. In addition, they have potential as oriented solvents that can impart selectivity in reactions by ordering the reactants.
- ***Molecular Liquids.*** Understanding multicomponent liquids is an increasingly important aspect of modern solution chemistry. As the number and complexity of molecular species increases, the necessity to probe longer length scales rises. Often, indirect in-solution molecular effects can play a critical role in chemical phenomena such as reaction product chirality, and polymorphism. The need to understand the role of intermediate range order in solvent media is another area of rising importance. Many chemical and biochemical processes require biphasic solvents for efficient operation, especially when reaction intermediates and products have different solubilities in non-polar or polar media. No existing neutron instrument has the range of characteristics required to probe such processes: NIMROD will therefore make a unique contribution.

**Functional and Composite Materials**

NIMROD will probe the important structural correlations in a wide range of disordered and partially disordered solids. This will allow us to understand the relationships between structure and properties, and increasingly to tailor materials to their function. Examples include optically and biologically active glasses, ionic conductors, advanced polymers, electrode materials, and selective molecular storage/release media. The relevance of NIMROD also extends to many advanced composites, where nanometer-scale particles or inclusions are embedded in a solid or liquid matrix. This broad class of materials includes colloidal and liquid crystal dispersions, sol-gel systems, polymer-inorganic composites, magnetic media, and intercalation compounds.

- ***Porous Media and Intercalates***. The proposed development of NIMROD is particularly timely for studies of this class of materials, which includes templated glasses, nanoporous polymer networks, zeolites, and graphitic and inorganic intercalation compounds. Key targets in this research are the ability to control the architecture of the host material, and the host-guest interactions. This will have impact in applications such as hydrogen storage, battery electrodes, supercapacitors, and organic and radioactive waste containment. Again the benefits of NIMROD are clear – it will enable us to expand the host superlattice to hundreds of Ångstroms while retaining atomic resolution.
- ***Optical, Biocompatible, and Conducting Glasses.*** Amorphous and glassy solids find increasing application across a wide range of modern technological applications. Examples include materials such as a-Si:H as an amorphous semiconductor, rare-earth doped fibre optics in amplifiers or lasers, and amorphous magnetic materials. To date, there is little information on the key functional mesoscopic length scales which relate to the correlation lengths characteristic of conduction

electron mean free paths, optical and compositional inhomegeneities, voids, and the length scale of magnetic interactions. Instrumental capabilities in this area are also expected to make a significant impact in improving our understanding of the glass transition in electrolyte glasses. An exciting emerging theme is that of bio-active and bio-compatible glasses, for use in tissue growth and replacement.

- ***Sol-Gel Materials and Colloidal Dispersions.*** Sol-gel systems are finding an increasing number of applications, including optical coatings, filters, or ultra low expansion materials. In addition to local structure, low-$Q$ data are vital to obtain information about their composition, homogeneity and mesoscopic structure, and thereby to tailor their useful properties to specific tasks. Similar arguments make NIMROD well suited for studies of those colloidal dispersions of particles such as fullerides, and metallic and ice clusters, in which the characteristic structural correlations extend to a few hundred Ångstroms.

**Phase Behaviour and Nucleation**

A forte of NIMROD is its ability to measure, on the same sample and at the same time, structure over a wide range of length scales in the region of a phase transition (liquid-liquid separation, crystallisation, metal-nonmetal transition, magnetic ordering, ionic conductivity in glasses), as a function of thermodynamic parameters or composition. This information is a prerequisite for unravelling the underlying mechanisms in numerous nucleation, growth and phase separation processes.

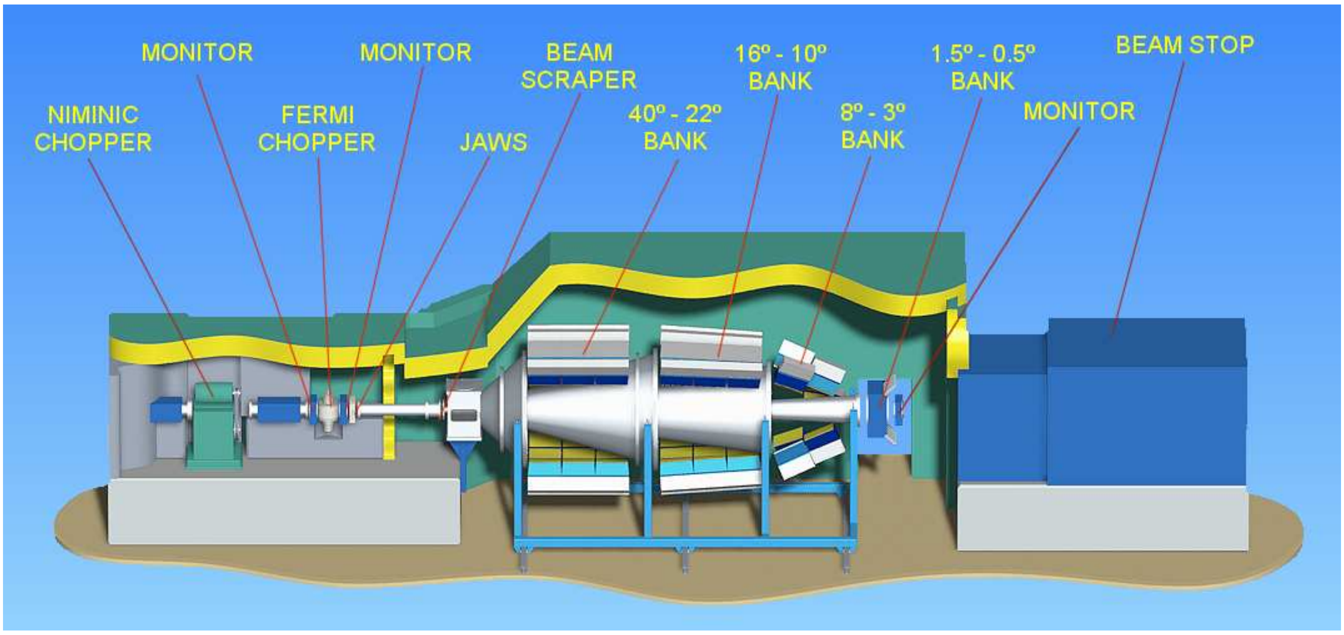
- ***Clathrate formation.*** Understanding the formation, decomposition and inhibition of gas clathrate hydrates is critical to tackling technological challenges posed by these materials, from pipe blocking to exploiting methane resources in sediments and permafrost. There is also new interest in the control of growth and luminescent properties of semiconductor clathrates. This area of research requires structural data from NIMROD, to follow disordered guest-host and guest-host-additive systems from the early stages of formation to the evolution of crystalline structures from these amorphous phases.
- ***Nucleation.*** The ability to measure small- and wide-angle diffraction simultaneously will allow us to focus on the mechanisms of a number of nucleation and growth processes, particularly the growth of crystals from pure liquids and solutions. The structure and properties of many solid materials is strongly dependent on the history of crystallisation, leading to polymorphs with widely different properties. Specific examples are that of ice formation in the presence of additives, such as sugars, and growth of metal crystals from the melt.
- ***Phase Behaviour of Multicomponent Liquids.*** There is currently no instrument that allows one to measure the mixing and phase behaviour of multicomponents. Recent experiments point towards chemically relevant microsegregation between organic and aqueous phases, but the important length scales are missing from the picture. The instrument would also open up the new field of density-driven phase changes in liquids and amorphous solids at constant composition, and allow observation of structural transitions among surface layers of longer-chain hydrocarbons adsorbed from multicomponent solutions. The latter systems are common to many commercial detergents, as they are cheap and offer high performance. In this context, many of the restrictions on molecule size and shape would be lifted.

In summary, NIMROD is a unique instrument that bridges the accessible length scales traditionally covered by small-angle and wide-angle techniques. The instrument therefore opens up whole fields of qualitatively new science, and complements current initiatives for fabricating functional materials and nanometre scale devices.

**Outline Design Specification:**

The fundamental idea behind NIMROD is to obtain diffraction data over a wide range of *Q*, both low and high, in a single experiment. Target Station II, with its 10Hz repetition rate and optimised target-moderator configuration, is an ideal source specification for NIMROD:

- NIMROD requires an incident flight path of at least 20m, and a secondary flight path up to 7m, to achieve a sufficiently small beam divergence and penumbra that the smallest scattering angles are reached with adequate resolution. Building a shorter flight path instrument with the same *Q* coverage and resolution would significantly compromise the count rate at all *Q* values.
- As a consequence NIMROD will require a time frame of at least 68ms to access the full wavelength range, 0 - 10Å, in a single pulse. It is therefore ideally suited to the 100ms time frame of TS-II, without the need for the pulse removing and frame overlap choppers that would be needed on TS-I.
- The low-*Q* capability of NIMROD is only possible by virtue of the excellent long-wavelength flux of the optimised coupled moderator available on TS-II. In the crucial low-*Q* region NIMROD outperforms SANDALS by a factor of up to 10 in count rate, even after taking account of the 1 in 4 pulse rate of TS-II compared to TS-I, and with better resolution. Such performance is not available on any of the TS-I moderators.



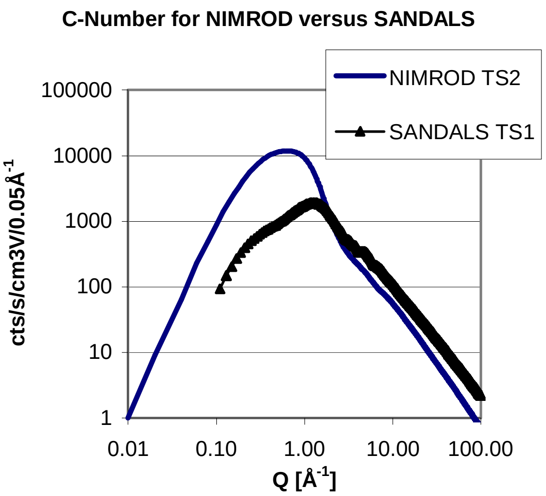

NIMROD will build on the successful SANDALS detector technology with specialised ZnS/glass sandwich detectors that are typically 70% more efficient than the equivalent $^3$He detector of the equivalent size. The detector electronics will follow the highly stable GEM design, which should deliver the required 0.1% stability over 24 hours as required to make full use of the available count rate on this diffractometer. This will increase the accessible concentration range for isotope difference experiments by a factor of 3 for second order difference experiments and a factor of 10 for first order difference experiments compared to SANDALS.

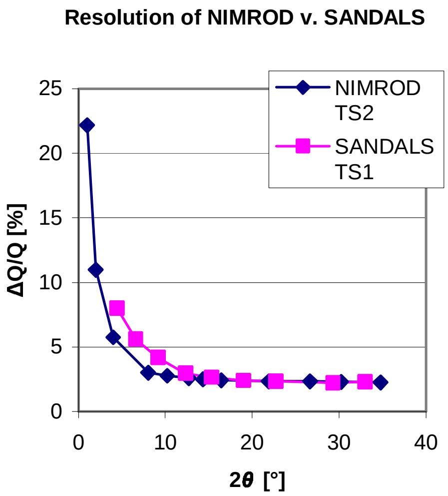
NIMROD will view the coupled cold moderator, which produces excellent fluxes of cold neutrons. Given that the resolution requirement (ΔQ/Q) is ~2% for most of the Q range, and that at the low scattering angles of NIMROD the resolution is dominated by geometric considerations, the broad pulses of this moderator do not affect the resolution significantly. For disordered solids, it is proposed to build a 90° detector bank with resolution ~0.5% to significantly enhance the performance at high-*Q*. The contributions from this higher angle bank have not been included in the performance characteristics given below, since detectors at 90° scattering angles can only be used for hydrogen-free samples.

| | |
|---|---|
| **Moderator** | Coupled cold |
| **Incident Wavelengths** | 0.05Å – 10Å |
| **Q-range** | $0.02Å^{-1}$ – $100Å^{-1}$ |
| **Resolution** | ~10%ΔQ/Q, 2θ = 0.5°-5°; 2%ΔQ/Q, 2θ = 12°-90°;0.5%ΔQ/Q. |
| **Total Length** | 30m |
| **$L_1$, $L_2$** | 20m, 1-7m |
| **Flight path** | Straight/tapered |
| **Detectors** | 10 x 200 x 20mm in rows parallel to beam, and over full range of azimuthal angles, ±90°. <0.1% stability required. |
| **Beam size** | 30mm wide x 30mm high |
| **Detector tank** | Vacuum, no beam windows visible by detectors |
| **Sample environment** | Standard, multi-position sample changer. |

## Performance

The estimated count-rate and resolution of NIMROD is compared with the current SANDALS in the following graphs.




C-number measures the count rate expected for $1cm^3$ of vanadium placed at the sample position. The projected numbers are based on the most recent estimates of target/moderator performance for both target stations and takes account of the different frequencies of the two target stations. At low Q NIMROD outperforms SANDALS, while at high Q SANDALS is better by a factor of 2 or 3 compared to NIMROD. If a way could be found to extract an epithermal neutron beam directly from the reflector, and combine this with a guide for long wavelength neutrons, then the performance of NIMROD could be enhanced even further. In practice experience with liquids on SANDALS indicates that data beyond $20Å^{-1}$ are rarely needed, and up to this Q value NIMROD is still highly competitive.

## Other Features

One idea currently being investigated is the possibility of putting a Fermi chopper (and corresponding NIMONIC chopper) in the incident beam line, with a view to doing low resolution inelastic scattering measurements on some samples. Placzek (inelasticity) corrections remain an unsolved problem for hydrogen-containing materials and it could be a useful feature to have the ability to look at the inelastic response of some materials in the angle range being used by the diffraction pattern. In addition a rotating Debye-Scherrer collimator is proposed which will serve to reduce low angle backgrounds substantially. Some form of tapered neutron guide will almost certainly be needed in the incident beam to correct for gravity effects on the longer wavelength neutrons.

## Letters of support received (attached as an appendix):

| | *Name* | *Affiliation* | *Area of interest* |
|---|---|---|---|
| 1. | Dr Ashok Adya | University of Abertay Dundee | Microporous media and electrolytes |
| 2. | Dr. Christiane Alba-Simionesco | Universite Paris-Sud | Confined and supercooled liquids and glasses |
| 3. | Dr. Paul Anderson | University of Birmingham | Porous media, composites and hydrogen storage |
| 4. | Prof. Carla Andreani | Universita Roma2 | Hydrogenous molecular liquids |
| 5. | Dr Adrian Barnes | University of Bristol | Electronic liquids and liquid metal alloys |
| 6. | Dr. Chris Benmore | Argonne National Laboratory | Functional and composite materials |
| 7. | Dr. Daniel Bowron | ISIS | Hydrogen bonded liquids and solvation |
| 8. | Dr Piers Buchanan | Kings College London | Superionic liquids and clathrate hydrates |
| 9. | Prof. Eugene Bychkov | Université du Littoral | Functional glasses |
| 10. | Dr. Stuart Clarke | University of Cambridge | Liquid adsorption and colloidal dispersions |
| 11. | Dr. Jason Crain | University of Edinburgh | Phase behaviour of molecular liquids |
| 12. | Prof. Roger Davey | UMIST | Nucleation and crysytallisation |
| 13. | Prof John Dore | University of Kent | Liquids under confinement |
| 14. | Dr. Sofia Diaz Moreno | ESRF | Molecular liquids and reaction media |
| 15. | Dr. Luis Fernandez Barquin | Universidad de Cantabria | Magnetic nanaparticles and composite materials |
| 16. | Prof. John Finney | University College London | Biomolecular liquids and nucleation |
| 17. | Prof. Henry Fischer | Laboratoire LURE, Paris | Large molecules and biological solutions |
| 18. | Prof Neville Greaves | University of Aberystwth | High temperature liquids and silicate glasses |
| 19. | Dr. Alex Hannon | ISIS | Structure of glasses |
| 20. | Prof. Jean-Pierre Hansen | University of Cambridge | Theory of liquids |
| 21. | Dr. John Harding | Chair, CCP5 | Computer simulation of condensed matter |
| 22. | Dr. Chris Hardacre | University of Belfast | Ionic liquids and nucleation |
| 23. | Dr Simon Hibble | University of Reading | Functional materials and nucleation |
| 24. | Prof. Robert Hillman | University of Leicester | Nanostructured and porous media |
| 25. | Dr Diane Holland | University of Warwick | Advanced functional materials |
| 26. | Dr. Uwe Hoppe | Universitat Rostock | Crystallisation of glass ceramics |
| 27. | Dr. Kathy Johnson | University of Liverpool | Complex liquids and particles in solution |
| 28. | Dr Dave Keen | University of Oxford | Framework structures and disordered magnetics |
| 29. | Prof. Mike Klein | University of Pennsylvania. | Computer simulation of complex systems |
| 30. | Dr. Carolyn Koh | King’s College London | Clathrate hydrates |
| 31. | Prof. Salvatore Magazu | Universita di Messina | Cryoprotectants in biological solutions |
| 32. | Prof. Robert McGreevy | ISIS | Disordered materials |
| 33. | Prof. Paul McMillan | Royal Institution | Amorphous and nanocrystalline materials |
| 34. | Prof. Geoff Mitchell | University of Reading | Functional polymers and organic materials |
| 35. | Dr. George Neilson | Bristol University | Biological and electrolyte solutions |
| 36. | Prof Bob Newport | University of Kent | Sol-gels and bioactive glasses |
| 37. | Dr. Hugh Powell | University of Durham | Waste containment in clays |
| 38. | Dr. Silvia Ramos | ESRF | Molecular liquids and ionic solutions |
| 39. | Prof. Maria Antonietta Ricci | Chair, CNR | Complex liquids and disordered materials |
| 40. | Prof. Rob Richardson | University of Bristol | Conformation in colloids and liquid crystals |
| 41. | Prof. Peter Rossky | University of Texas at Austin | Modelling of complex and biological systems |
| 42. | Dr Philip Salmon | University of Bath | Fast-ion conductors and polymer electrolytes |
| 43. | Prof. Roger Sinclair | University of Reading | Poorly crystalline materials and magnetism |
| 44. | Dr. Neal Skipper | University College London | Electronic liquids and confined fluids |
| 45. | Prof. Mark Smith | University of Warwick | Sol-gels and bioactive glasses |
| 46. | Prof. Alan Soper | ISIS | Liquids and disordered materials |
| 47. | Dr. Jan Swensson | Chalmers University | Polymer composites and biomaterials |
| 48. | Dr Matt Tucker | University of Cambridge | Mineral physics and radionuclide containment |
| 49. | Dr Beau Webber | University of Kent | Mesostructured porous materials |
| 50. | Prof. Adrian Wright | Universiy of Reading | Nanoheterogeneities in glasses |
| 51. | Prof. Marco Zoppi | CNR, Firenze | Hydrogen storage and confined fluids |