4.1 Solid State Physics

R. Currat[1], R. Cywinski[2], C. Fermon[3], A. Furrer[4], B. Keimer[5], G.H. Lander[6], D.F. McMorrow[7], H.R. Ott[8], C. Vettier[1]

[1] Institut Laue Langevin, 6 rue de Horowitz, F-38042 Grenoble 9, France
[2] Department of Physics and Astronomy, University of Leeds, Leeds LS2 9JT, United Kingdom
[3] Laboratoire Léon Brillouin, CEA, Saclay, F-91191 Gif-sur-Yvette Cedex, France
[4] Laboratorium für Neutronenstreuung, ETH Zürich and PSI Villigen, CH-5232 Villigen PSI, Switzerland
[5] MPI für Festkörperforschung, Heisenbergstraße 1, D-70569 Stuttgart, Germany
[6] Institut für Transurane, Postfach 23 40, D-76125 Karlsruhe, Germany
[7] Condensed Matter Physics and Chem. Dep., Risø National Laboratory, P.O. Box 49, DK-4000 Roskilde, Denmark
[8] Laboratorium für Festkörperphysik, ETH Zürich, CH-8093 Zürich, Switzerland

Abstract

Solid state physics encompasses fundamental research that has underpinned much of the technological progress in the last 50 years. Recent trends include the emphasis on complexity, including organic materials, and reduced dimensionality down to the scale of quantum dots. In the large variety of instruments used in solid-state physics, scattering has a special place as it gives information on **spatial** correlations. Within scattering techniques, neutrons are unique as they are able to provide, simultaneously, information on **both** the **static** and **dynamical** correlations. We discuss these advantages for neutrons, stress the materials-driven nature of this approach, and present a selection (by no means complete) of flagship experiments that will be possible only at the ESS. We conclude with a discussion of the best instruments and pulse structure for frontier experiments at the ESS.

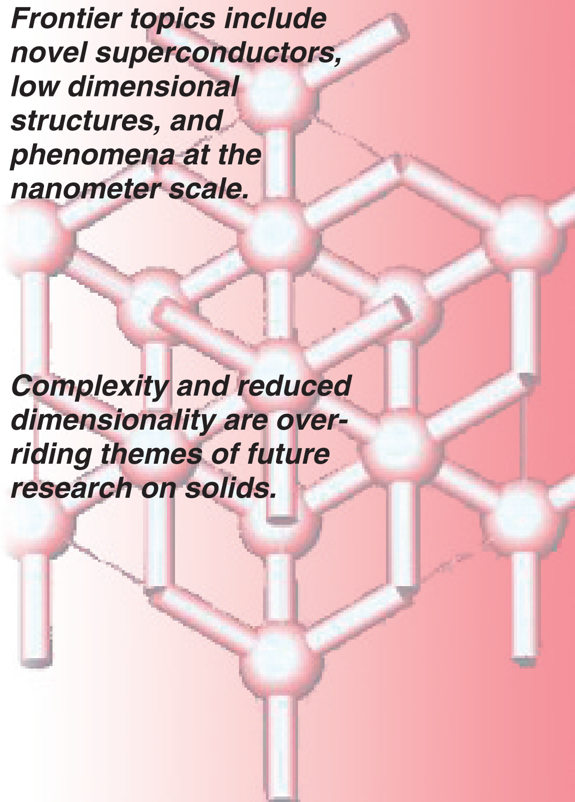
I. Introduction

This report identifies future research frontiers in solid state physics. It starts by reviewing briefly the general capabilities of neutron scattering methods for the study of phenomena in condensed matter physics, with particular emphasis on the capabilities provided by powerful modern spallation neutron sources such as the ESS. Although complementary methods such as the other scattering probe, synchrotron X-rays, and local probes like NMR, EPR and Mössbauer, provide important information, our deliberations have confirmed the unique opportunities afforded by neutrons in general and by ESS in particular. The report highlights examples of flagship experiments, and addresses the impact that the ESS can have in these frontier areas. Recommendations for instruments and target options are presented.

Advances in solid state physics are at the root of most technologies shaping today's world. Neutrons are key to our understanding of solids. The ESS will have a large impact on cutting-edge research in solid state physics.

Research in the solid state continues to have a high impact, both in basic physics as well as with respect to technological applications. Recent examples include high-T_c and other unconventional superconductors, low dimensional semiconductor structures, magnetic thin films, and materials with giant magneto-resistance. A great deal of basic research is now technology driven and much is oriented towards nanometer-scale systems. The properties of nano-patterns and self-assembled quantum dots are of great interest from both a theoretical and experimental perspective.

Frontier topics include novel superconductors, low dimensional structures, and phenomena at the nanometer scale.



The future challenge in basic solid state physics is the exploration and understanding of the collective behaviour of large numbers of interacting particles. Although future trends are notoriously difficult to predict, two important directions emerge. Firstly the tendency to higher complexity, specifically materials which have physical properties determined by competing interactions, and secondly the trend to reduced dimensionality, both by synthesizing materials with low

Complexity and reduced dimensionality are over-riding themes of future research on solids.

dimensional structural elements and by reducing the physical size of objects to surfaces and interfaces, single atom wires and dots. One basic interest in solid state physics is to establish the ground state of relevant systems. This may be done by exploring possible excitations out of the ground state, neutrons are a versatile, and often unique probe with which to accomplish this goal.

The table summarises some of the research areas that are expected to be of major interest in ten years time.

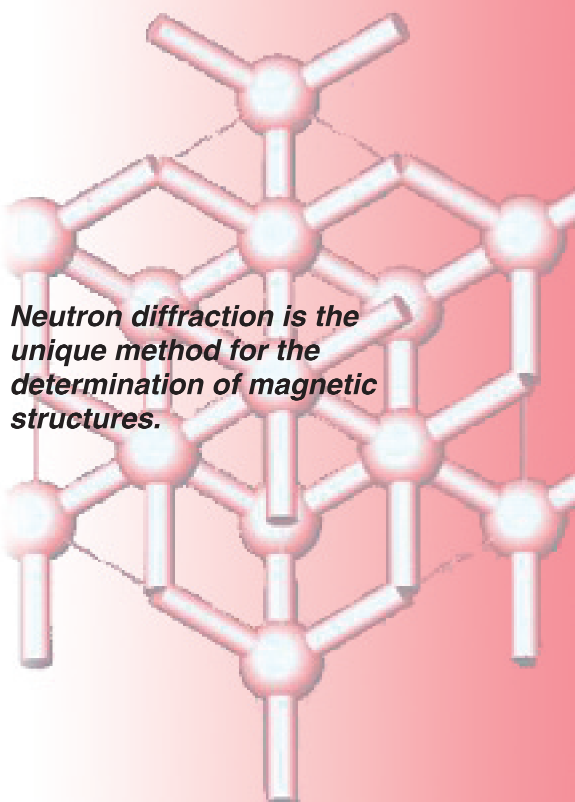
Table 1:
Frontier Research Areas in Solid State Physics

Dimensionality	Complexity	Structures and lattice effects	Non-equilibrium and time-dependent phenomena	New Materials
Quantum dot arrays Transport and magnetic properties in 1-d systems Domains walls, domains correlations, grain boundaries Surfaces and thin films	Interplay of spin, orbital and charge degree of freedom Coupled excitations Strongly interacting electron systems Flux line lattices Phase transitions, quantum critical points	Frustration Disorder, interfacial roughness Proximity effects Lattice modes Confinement	Fast response to external probes and fields Magnetic fluctuations and relaxations Tunnelling	Molecular magnets Interfaces/hybrid structures Self-organising molecular systems Novel magnets and superconductors Organic materials

II. The role of neutrons

In solid state physics, the degrees of freedom and interactions necessitate the use of a large variety of experimental methods. Bulk measurements of thermal and transport properties are invariably the first step, but several techniques are needed which are sensitive to the atomic environment and electronic structure, and which provide information on the spin state of the electron. Electron correlations in the solid state exist over vast ranges of spatial and temporal (energy) length scales. Local probes such as STM, and atomic spectroscopic studies using electromagnetic radiation provide crucial information. In addition, scattering methods (neutrons, x-rays and electrons) provide inter-atomic information on spatial correlations. However, the neutron has a unique combination of properties that make it indispensable for many problems in solid state physics. The de Broglie wavelength of thermal neutrons is on the same scale as inter-atomic spacings, allowing diffraction experiments to be conducted to locate the positions of atoms. Because of their mass, neutron have a rather low kinetic energy; they can be moderated and neutron beams are produced with energy in the range 0.1 meV to 10 eV, well matched to solid state excitations. (This is to be contrasted with x-rays, which at comparable wavelengths are much more energetic, in the keV range.) Because of their magnetic moment (spin), neutrons are sensitive to magnetic moments arising from electronic and nuclear magnetic

Because of a unique combination of properties, neutrons are a powerful and indispensable probe of solids.



Neutron diffraction is the unique method for the determination of magnetic structures.

moments. Because they are uncharged, neutrons penetrate deep into materials. Because they are weakly interacting (in contrast to electrons), measured scattering cross-sections can be compared directly to theory.

Neutrons have played a pivotal role in the investigations of phase transitions and co-operative phenomena, magnetism, structure (static and dynamics), as well as in many other fields. Particularly intriguing is the connection between phase transitions and theoretical concepts, such as symmetry breaking, order parameter, universality class, scaling and critical behaviour. In the past, the interplay between theoretical concepts and experimental observations concerning phase transitions has been extremely successful and many significant contributions have been made using neutron scattering.

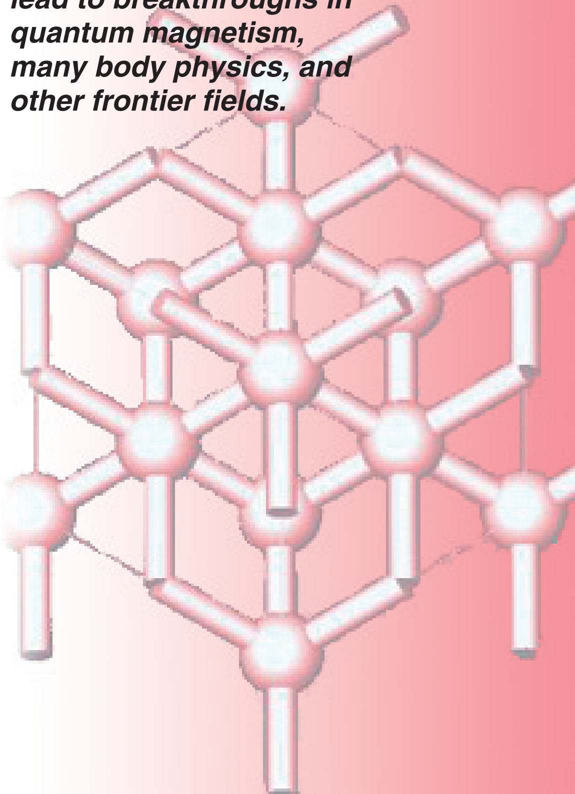
The interplay between neutron experiments and theory has driven the development of many new concepts in solid state physics.

A vast panoply of neutron techniques have contributed to this work. Neutron diffraction (from powders and single crystals) is a basic, but essential, technique, providing information on chemical and magnetic structures. Neutron reflectometry using polarised neutrons has given us a clearer picture of the growth and the physics of magnetic thin films and superlattices. Inelastic neutron scattering is the only probe that provides a complete picture of both structural and magnetic dynamics in solids. Emerging techniques include analysis of three dimensional polarisation, and the direct mapping of the full dynamical susceptibility over the entire Brillouin zone. All of these techniques suffer from the intrinsic low brilliance of neutron sources; as a result, the materials studied must have a sizeable volume and/or sizeable scattering density. The (lateral and vertical) spatial resolution is restricted to a few 100 µm while the temporal resolution is in the 0.1 sec range. The advent of the ESS will offer entirely new capabilities to explore spatial and temporal properties of condensed matter with µm and ms resolution, respectively.

Low intensity continues to be the major limitation of neutron research. Due to its high flux, the ESS will open up entirely new opportunities.

Here we emphasise a few topics where neutron scattering techniques are expected to play a major role. In ***magnetism***, significant advances are expected in synthesizing molecular and organic magnets, i.e. solids built from structurally well-defined clusters containing magnetic ions in a complex environment. These are of both fundamental importance, and with respect to potential application in magnetic storage devices. New developments are also expected in exploring novel magnetic phases and their dynamics in low-dimensional systems. The study of ***phase transitions*** will continue to be a major field of research with neutron based techniques. Systems of high complexity, exhibiting extreme many body effects (e.g. unconventional superconductivity) and low dimensional features are known and expected to undergo a large variety of phase transitions. Their exploration using neutron techniques will provide crucial insights into the microscopic mechanisms causing these phenomena. Of high current and most likely future interest is the relationship between spin polarisation and transport of conduction electrons in specially tailored materials, ***spintronics***. High

Research at the ESS could lead to breakthroughs in quantum magnetism, many body physics, and other frontier fields.



intensity neutron beams will play a central role in elucidating the spin polarisation and dynamics of these electrons.

III. Future opportunities – flagship areas

The ESS will lead to breakthroughs in three distinct ways:

a) to allow scientists to address new problems, and to ask new questions.

b) to provide new tools to tackle problems at the research frontiers.

c) to offer high quality experimental data for unambiguous discrimination between theoretical models.

In the following, we present selected flagship areas which are representative of the topics listed above.

Dynamics of superlattices, thin films, wires and dots

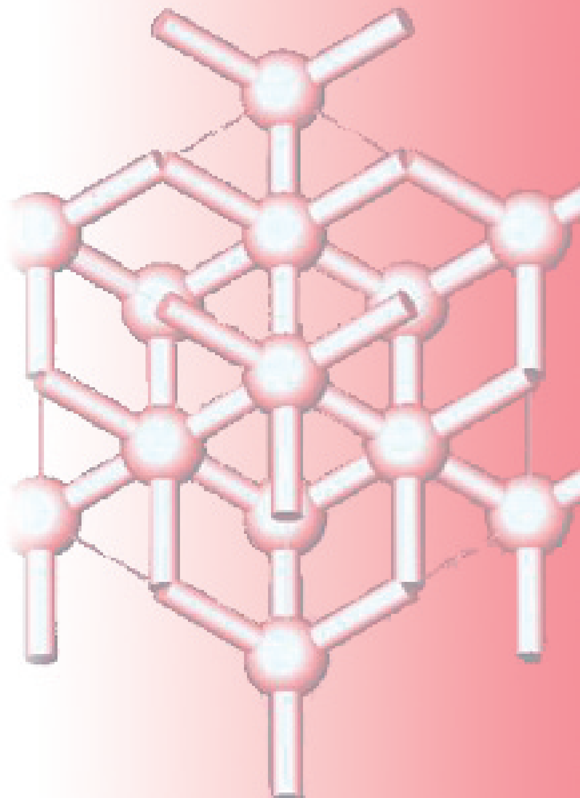
Following the discovery of giant magneto-resistance in 1988, the physics of micro- and nano-structured magnetic materials has become a field of intense activity. Thin films and superlattices, as well as wires and dots are now extensively studied for their fundamental properties and their potential applications in systems like sensors and magnetic random access memory devices (MRAMS). Understanding the dynamics of these systems will continue to be a key challenge. In contrast to Brillouin light scattering and ferromagnetic resonance techniques, neutron scattering gives access to the whole Brillouin zone.

Neutron beams at the ESS will provide maps of the magnetic polarisation and dynamics of nano-structured materials and devices.

Brillouin light scattering has provided the first dispersion curves in magnetic dots, but is limited to 30 GHz. Until now, there is no theory able to explain the excitations measured in dots, even in simple NiFe square dots. An experimental input at higher frequencies appears to be essential in understanding the dynamics of these systems. A reflectometer at the ESS will offer the capability of measuring spin wave spectra in very thin films, wires and dots, and will certainly have an important impact on the field of nano-magnetism.

In contrast to alternative techniques, neutrons provide access to a wide range of energies and momenta.

The observation of magnetic inelastic scattering from superlattices is presently at the limits of neutron technology. An interesting experiment has been performed recently at the ILL on a Dy/Y superlattice, where the effects of folding on the inelastic response function due to the superlattice periodicity have been observed. Such neutron experiments allow the exchange coupling parameters, both within a single layer and between the layers, to be deduced. However, this will require a considerable increase in intensity, coupled with better resolution, especially if technologically important films such as transition-metal superlattices are to be examined and understood.



Molecular magnets

A typical example of a molecular magnet is Mn_{12} acetate with total spin quantum number S = 10, giving rise to thousands of excited spin states which can only be disentangled by high resolution neutron spectroscopy. For instance, the lowest lying group of spin states comprises (2S + 1) = 21 levels of energies $\eta\omega \leq 1.2$ meV as shown in Figure 1 below. Mn_{12} acetate exhibits quantum tunnelling between these spin states which can be tuned in a controlled manner by an applied magnetic field. This opens the way to a novel class of information storage systems on the molecular level. Unfortunately, the quantum tunnelling in Mn_{12} acetate is restricted to temperatures of a few Kelvin. The search is on for materials that would preserve the virtues of Mn_{12} acetate at liquid nitrogen temperature.

Molecular magnets could serve in atomic-scale information storage systems. Neutron scattering is a unique probe of their excitation spectra, whose accuracy will be tremendously enhanced by the ESS.

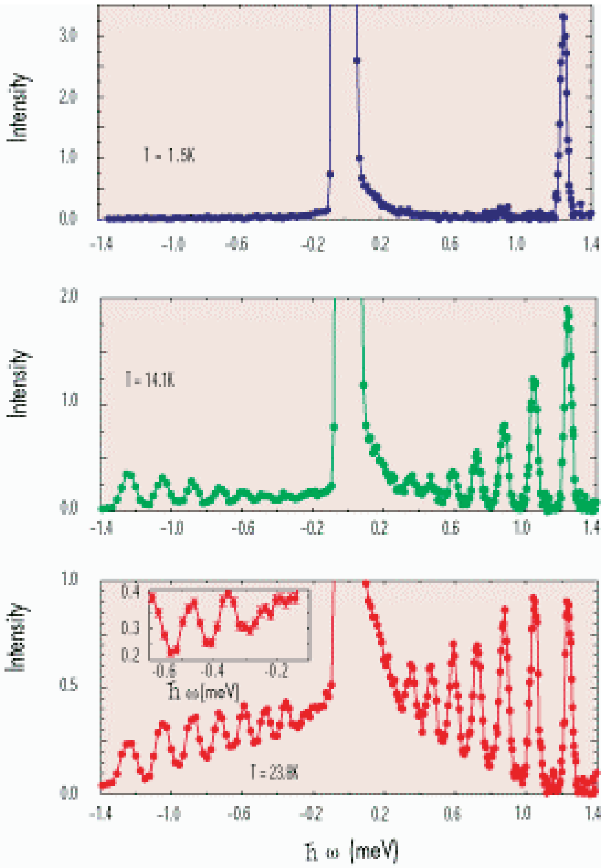
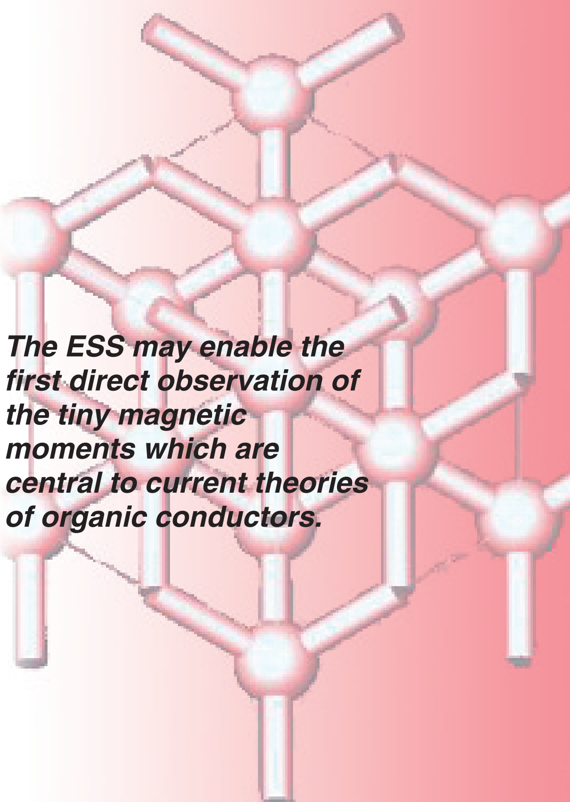


Figure 1: Energy spectra observed at three different temperatures in Mn_{12} acetate (Courtesy of I. Mirebeau).

Spin density waves in organic materials

Among the low-dimensional electronic systems, the charge transfer Bechgaard salts $(TMTTF)_2X$ and $(TMTSF)_2X$ (X = PF_6, AsF_6, SbF_6, SCN) show the richest phase diagrams with almost all known electronic phases: a metal, a paramagnetic insulator, spin and charge density wave states, a spin-Peierls state and finally an unconventional superconducting state.

The ESS may enable the first direct observation of the tiny magnetic moments which are central to current theories of organic conductors.



Other salts in the same family have been shown to exhibit the quantum Hall effect. The phase diagrams have been mapped out mostly based on transport and specific heat data as well as NMR results.

For the spin density wave (SDW) phases, detailed NMR predictions exist concerning the wavevector of the modulation and the amplitude of the ordered moment. Yet, so far, direct neutron evidence for a SDW superlattice peak is missing, due to a combination of the weak magnetic moment (~ $0.08\mu_B$), the unfavourable magnetic form factor and small sample sizes. Experiments of this type will become possible with the intensity available at the ESS, allowing a direct determination of the SDW amplitudes and periodicities, and thereby opening up an entirely new area of research.

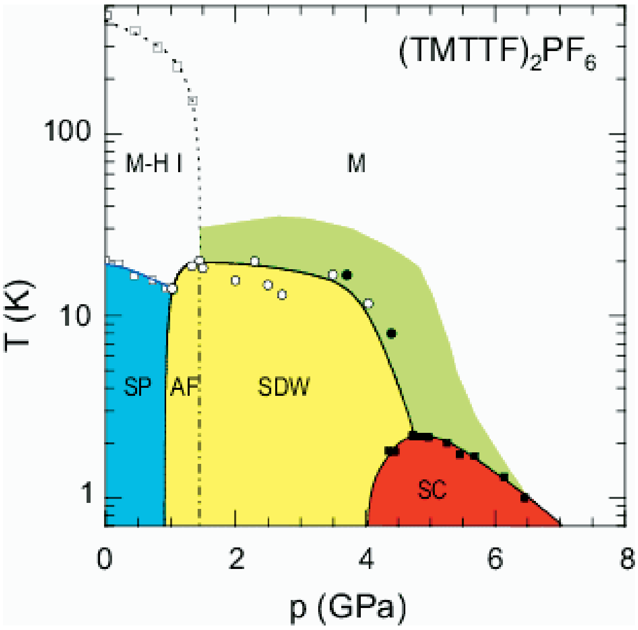
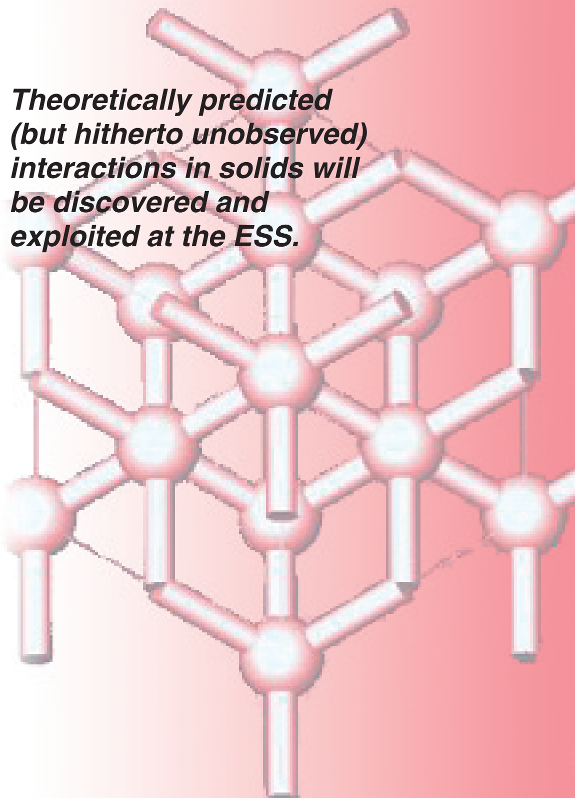


Figure 2: Phase diagram of the Bechgaard salt $(TMTTF)_2PF_6$ deduced from resistivity (Courtesy of D. Jaccard).

Revealing exotic interactions

The properties of magnetic materials are usually described in terms of bilinear spin interactions. Neutron spectroscopy with its dipole selection rule $|\Delta M| = 1$ has been the technique of choice to measure the magnetic excitation spectrum and thereby, to allow the direct determination of the magnetic exchange coupling constants. However, there are numerous examples such as molecular magnets, high T_c cuprates and f-electrons compounds where higher-order interactions (e.g. quadrupolar, octupolar, three- and four-body exchange) are relevant, but their sizes could so far not be determined directly. In principle, neutron scattering allows the direct observation of higher-order term transitions. However, the associated transition matrix elements are typically two orders of magnitude smaller than for dipolar scattering. Such novel experiments would be made possible by the ESS.

Theoretically predicted (but hitherto unobserved) interactions in solids will be discovered and exploited at the ESS.



Coupled excitations

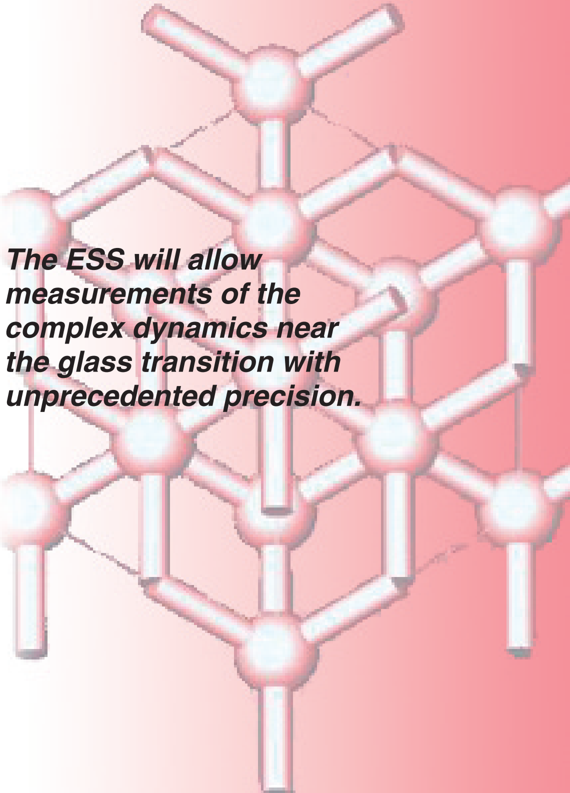
Inelastic neutron scattering at the ESS will open a new window on the electronic structure of metals and superconductors.

Excitation phenomena in solids can be classified into single particle continua and collective modes. Because of intensity constraints, neutron scattering experiments have been almost exclusively limited to collective modes. Recent investigations of two-spinon continua in insulating one- and two-dimensional quantum magnets are pushing the limits of current sources. At a chopper spectrometer at the ESS with wide reciprocal space coverage, detailed maps of single particle Stoner continua in metals and superconductors will be obtained up to energies of the order of the Fermi energy. In systems where correlation effects are strong (which are currently at the forefront of condensed matter science) it will be possible to extract a wealth of information on the band dispersions, Fermi liquid parameters, superconducting coherence effect, etc., that is currently inaccessible. The high neutron flux at the ESS will also enable high resolution measurements of the intrinsic lifetimes of collective modes over the entire Brillouin zone. Although predictions for the lifetimes of magnetic and lattice vibrational excitations (for instance, due to electron-phonon scattering) have been available for many years and are becoming ever more accurate, they could thus far be tested only in a few special cases.

Physics of defects at the dilute limit

Defects are ubiquitous in solids, and high intensity neutron beams at the ESS will provide incisive information about their microscopic structure.

The increasing sophistication of “first principles” theoretical calculations of the fundamental electronic structure, total energies and atomic short range order, places stringent demands upon the accuracy of experimental measurements of the extended atomic and magnetic defects around impurity atoms in metals, alloys and compounds. Experimentally, information on this problem can be obtained only from diffuse neutron scattering experiments, with polarisation analysis. The associated cross sections are extremely small and counting times are often prohibitively long. Moreover, such experiments should ideally be carried out at extreme dilution to circumvent the often intractable problem of non-linear superposition of overlapping defects. These experiments are crucial for a full solution of the defect problem and experimental corroboration of the most sophisticated of our “first principles” band theoretical calculations, but is not feasible at present neutron sources.



Spin glass dynamics

The ESS will allow measurements of the complex dynamics near the glass transition with unprecedented precision.

It has been said that the deepest and most interesting unsolved problem in solid state physics is probably the nature of glass and the glass transition. Indeed the status of the glass transition as a true thermodynamic transition is still questioned. Spin glasses provide a simple analogue of the structural glasses yet the *accurate* measurement of relaxation processes in spin glass systems is at the very limits of what is feasible at present using spin echo facilities. A wider Fourier time, coupled with significantly improved counting statistics, is required to determine the precise functional form of the relaxational dynamics, both for comparison with structural glasses and to discriminate between the proposed theoretical models. In addition the measurements should also be performed over a wide range of magnetic dilution, the lower ranges of which are entirely inaccessible at present. The implications of such studies are profound, as many of the spin glass relaxational models are finding applications in areas as diverse as virus mutation, protein folding and the travelling salesman problem.

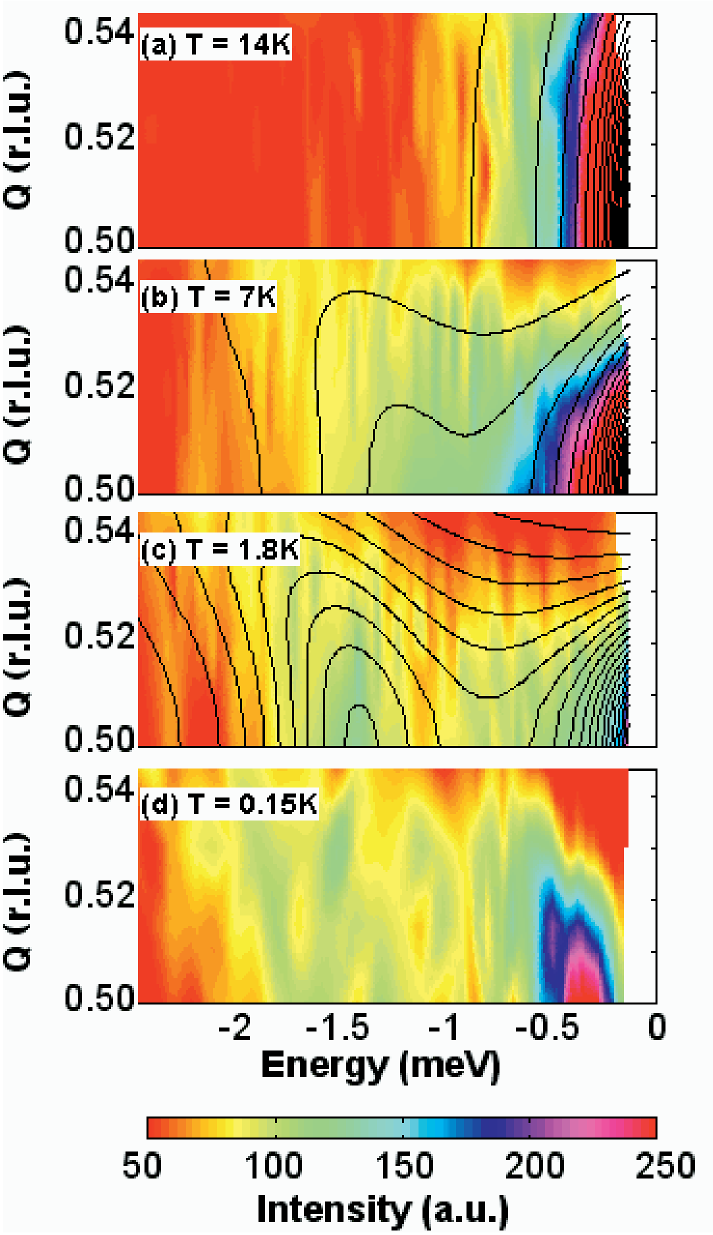
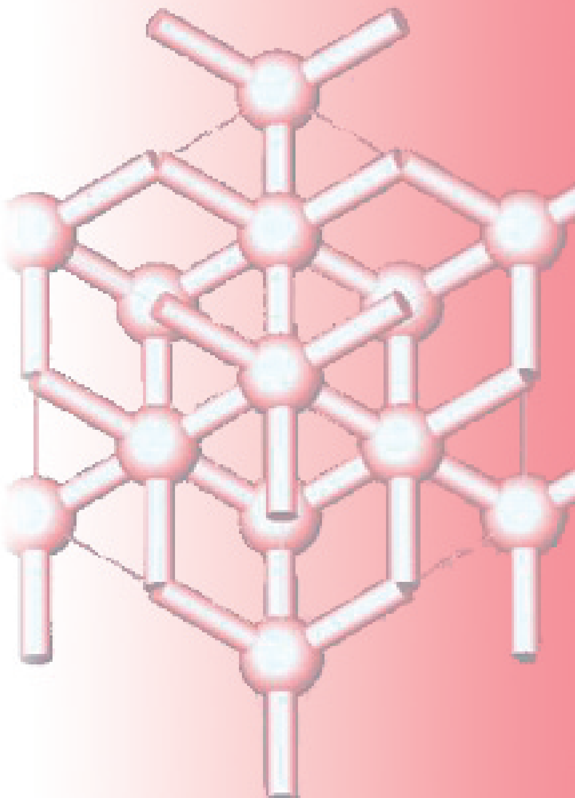


Figure 3: Temperature dependence of the magnetic response of UPd_2Al_3 near the superconducting phase transition. Above the phase transition, there is coexistence of quasi-elastic scattering and dispersive modes. In the superconducting state the quasielastic scattering is replaced by a low-lying inelastic mode (data taken from (Courtesy of N. Bernhoeft).



Quantum phase transitions

Of special interest are situations where either the competition between different interactions prevents the system from readily adopting a well-defined ground state, or where a restriction in spatial dimensionality does not allow for long-range ordering phenomena. Interactions of similar, medium or large magnitude may lead to very complicated phase diagrams. Cases where all the interactions are of similar strength and weak are very challenging. Here the situation may occur that the phase transition only sets in at $T = 0K$, in the quantum critical regime. A related example would be the important and technically challenging experiment on a material such as the recently discovered ferromagnetic superconductor UGe$_2$. Inelastic scattering experiments need to be performed at low temperature (< 0.5 K) and at pressures of up to 3 GPa.

Experiments at the ESS will set new benchmarks for extreme conditions of external pressure and magnetic field, thus providing important insights into zero-temperature phase transitions driven by quantum fluctuations.

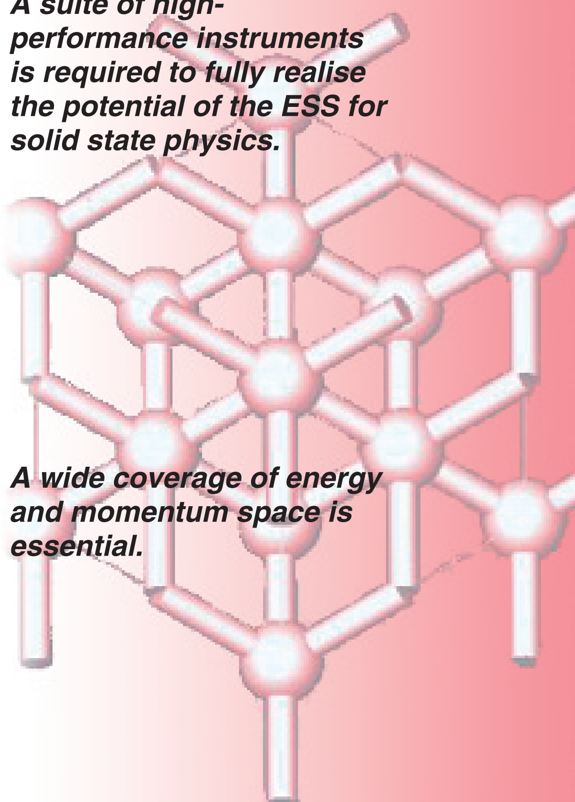
The key requirement is to map out the inelastic response function over a wide range of momentum space and energy as a function of temperature, external pressure and applied magnetic field. While neutron scattering can make unique and essential contributions to our understanding of the mechanisms underlying these phenomena, the sample volumes that can be subjected to these extreme conditions are necessarily small. At current neutron sources, inelastic scattering studies are hence restricted to a few special cases. Detailed investigations of the dynamic aspects of magnetic field and/or pressure induced quantum phase transitions require the ESS.

It should be stressed that in such experiments the positions in momentum space where the maxima will occur are unknown, so that a wide “mapping” technique is required. Many new materials call for this approach; unavailable at present, but a planned development at the ESS.

IV. Instrument requirements at ESS

The requirements for instrumentation in solid state physics are based upon two principle demands: that of probing (Q,ω) space for excitations, and that of providing a detailed structural (atomic and magnetic) characterisation of the sample. Whilst the former demand can only be met by a suite of inelastic scattering spectrometers, the latter must be met by a range of rather disparate total- and elastic scattering instruments, namely powder and single crystal diffractometers, diffuse scattering instruments and reflectometers. In all cases polarised incident neutrons and polarisation analysis are either essential or a distinct advantage, and the instruments should be capable of accepting extreme sample environments (e.g. pressure, magnetic field, temperature etc.).

A suite of high-performance instruments is required to fully realise the potential of the ESS for solid state physics.



The key scientific topics we have highlighted demand a coverage of Q space from $0.1 \leq Q \leq 12Å^{-1}$ and an energy transfer range of six orders of magnitude, from μeV to eV. This can be achieved through an instrument suite consisting of a

A wide coverage of energy and momentum space is essential.

backscattering spectrometer, a variable resolution cold chopper spectrometer, thermal and high energy chopper spectrometers, and a constant-Q spectrometer. The resulting coverage of Q-ω space is illustrated in the diagram below.

A single crystal diffractometer is an essential component of the suite for determination for the structural and magnetic order and for crystallographic studies of multilayer systems. Polarised incident neutrons and high magnetic fields at the sample position will enable magnetic spin density determination.

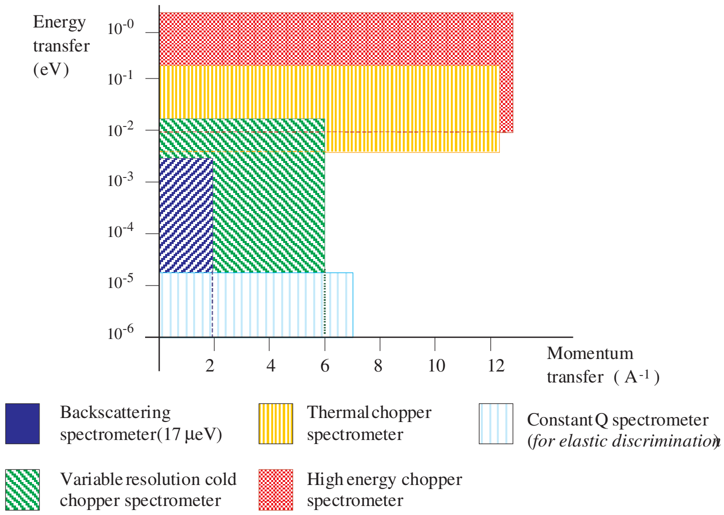
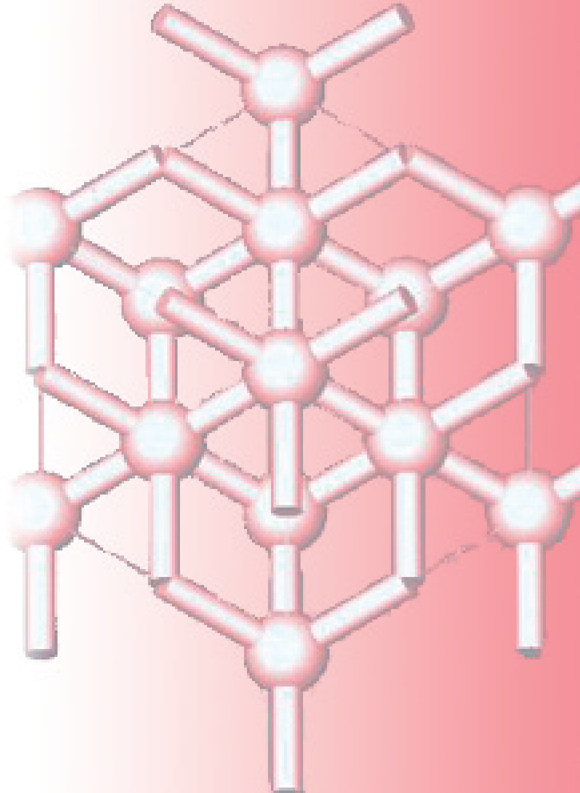


Figure 4: Required coverage in the Q-ω space.

Solid state physics places several conflicting demands on powder diffractometry. Firstly the growing complexity of magnetic structures (e.g. spirals and spin density waves) studied by neutron diffraction requires high resolution (Δ d/d > 0.1%) across a Q-range from a lower limit of at least 0.3 Å^{-1} to 12 Å^{-1}. The efficient mapping of the evolution of magnetic and structural phases in parameter space defined by pressure, temperature and magnetic field is best met by a diffractometer following the GEM design at ISIS.

The study of the structures of artificial films, multilayers and mesoscopic structures requires a reflectometer optimised for intensity rather than resolution. Polarisation analysis and surface capabilities are essential. A dynamic range of 10^8 up to a Q of 0.512 Å^{-1} represents a real advance in reflectometry and should be considered to be a design goal.



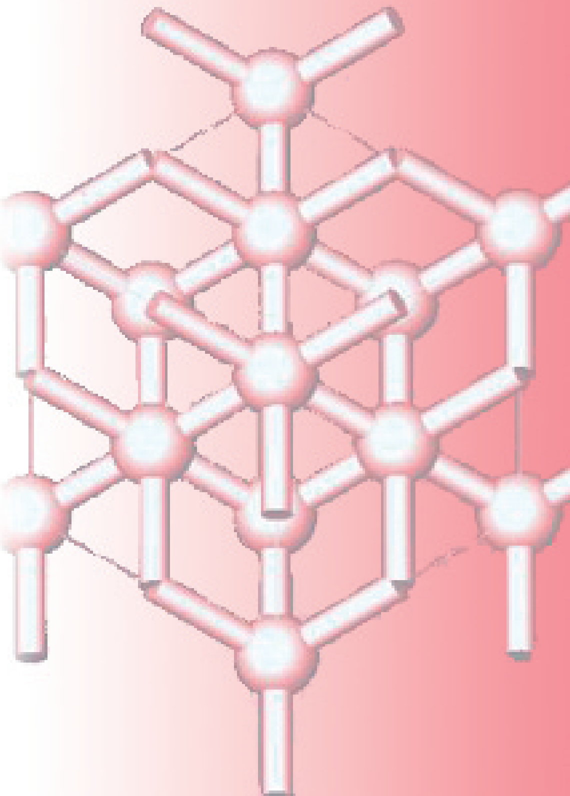
Our conclusions are summarised below:

Priorities for instruments:

First Priority:
Chopper Medium Resolution
Chopper High Resolution
Cold Chopper
High Intensity Reflectometer + Energy Analysis
Magnetic Powder Diffractometer

Second Priority:
High Resolution Powder Diffractometer
Chemical Single Crystal
High Resolution NSE
Medium Resolution Backscattering
Diffuse Scattering Diffractometer
Constant Q

The instruments needed for solid state physics are best placed at the 50 Hz 5 MW and $16^2/_3$ Hz 5 MW target stations.



Achievements of neutrons in solid state physics

- Almost everything which is known about magnetic structures of electrons– from the early demonstration by Shull of antiferromagnetism in simple systems, to the complex magnetic structures being developed for new magnetic materials – has come from experiments with neutrons. Complex magnetic orderings which include spirals, fans, and cycloids, and asymmetric magnetisation have been unravelled by neutron diffraction experiments.

- Pioneering experiments have been made on nuclear spin ordering in Cu and Ag, which are antiferromagnets below 70 nK and 600 pK respectively. These observations of one of the weakest interactions in a solid is a significant step towards defining the ultimate ground state of electronically non-magnetic materials.

- Polarised neutron reflectometry of surface and interface magnetism in thin films and multilayers is providing technologically relevant information. Important examples include the observations that the magnetic order can propagate through non-magnetic layers and that giant magnetoresistance is not necessarily associated with antiferromagnetic coupling. Magnetic roughness can be separated from the chemical roughness.

- Neutron diffraction provided the first microscopic evidence for flux line lattices in conventional superconductors and played a major role, especially at higher magnetic fields where no other technique can image flux lines, in accounting for the large dissipation in the high-T_c materials.

- Neutrons have provided the definitive crystal structures in high temperature superconductors, which have served as the basis of all considerations of the superconducting mechanism and have led to production of better quality samples. Neutrons have precisely located the positions of the oxygen atoms, where the charge carrying holes reside. Of particular importance has been the demonstration of the charge transfer concept of hole doping with oxidation, and that the superconducting temperature was related to the evolution of structural order. Neutron spectroscopy has provided unique information on the nature of magnetism in high temperature superconductors, on the interplay between magnetic fluctuations and superconductivity and on the role of the lattice dynamics.

- Inelastic neutron scattering has provided unique information on the interaction and anisotropy energies which determine the Hamiltonian operator. Extensive measurements on magnons and crystal field excitations over a wide range of frequency and momentum space have revealed e.g. the interaction of the spin waves with the single particle excitations (Stoner continuum) in iron. Similarly, measurements of phonons and their density of states have improved our understanding for example of the role of phonons in the martensitic phase transition in the IVb metals.

- Major contributions to our understanding of model systems for statistical physics in one, two and three dimensions, include the verification of the Haldane conjecture, determining the properties of the Haldane gap, and the discovery of solitons, as the characteristic elementary excitation of strongly non-linear magnetic systems. This also includes the observation of solitons in solids and the demonstration of all the expected, but previously not observed, properties.

- Outstanding results from space and time-dependent studies of correlated electron systems include proof of the importance of correlations for the spin dynamics and the antiferromagnetic character of the spin correlations. The interplay of electronic degrees of freedom (charge, spin and orbital order of electrons) has been amply evidenced.

- One of the most fundamental questions in condensed matter physics concerns the liquid and solid phases of ^{3}He and ^{4}He. Experiments with neutrons have provided unique and important information on, for example, the Fermi liquid parameters of ^{3}He, the Bose condensation of liquid ^{4}He and the magnetic structure of solid ^{3}He. The importance of understanding these quantum systems was highlighted by the award of the 1996 Nobel prize in physics for the discovery of superfluidity in ^{3}He by Lee, Osheroff and Richardson.