Beamline Name: polREF

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Summary:

polREF is a polarised neutron reflectometer for studying the *inter* and *intra* layer magnetic ordering of surfaces and thin film systems. These magnetic systems lie at the heart of many of the technological devices that are now being developed in areas such as computer storage media, field sensing and looking further forward such fields as quantum computation. Polarised Neutron Reflectometry (PNR) is a technique well matched to the investigation of such systems, providing unique information in both absolute depth and in-plane magnetometry, as well as imaging. The predicted gain in performance of polREF (typically a factor **×15-23**) over the existing CRISP reflectometer on the first target station, coupled with the new capability for high-angle scattering, flexible sample environment and advanced beam polarisation conditioning, will provide a world-class facility, which will lead to new insights in the technologically significant area of thin film magnetism.

1 Science Case:

1.1 Introduction

It is now possible to grow magnetic multilayers with almost atomic-plane precision, potentially with tailor-made physical properties. The transition from scientific discovery to commercial exploitation has been dramatic in this field, and the panoply of practical applications includes magnetic devices for data storage, many types of sensor and, potentially, quantum computing. The revolutionary development is the creation of devices that exploit the spin rather than the charge of the electron. One such device made of magnetic multilayers, called a spin valve, is already employed as a read head for computer hard disks, and this has led to dramatic improvements in data storage densities. Arrays of spin valves may be used to create magnetic random access memory, which has the advantage of being non-volatile, yet faster, higher density and lower cost than the current dynamic random access memory devices. The ever-decreasing grain sizes mean that in the highest density recording media of today are approaching the limits of thermal stability and, as a consequence, patterned nanoparticle assemblies on surfaces are being considered for data storage. Multilayers also offer a new approach to some of the canonical problems in condensed matter research, such as the interplay between superconductivity and magnetism, and quantum confinement within thin layers.

Polarised Neutron Reflectivity (PNR) gives detailed information on the magnetic ordering through a surface or a buried interface and, as a consequence, it has played a pivotal role in the study of thin-film magnetism. For example, specular reflectivity determines the magnetic moment magnitude and direction as a function of depth, and it gives information on magnetic coupling in spin valves and flux penetration in superconductors. Off-specular reflectivity probes in-plane correlations providing unique insight into the magnetic disorder at interfaces and magnetic interactions in magnetic nanostructures on surfaces. The polREF reflectometer will offer dramatic improvements over existing instrumentation and will make a major contribution to all these areas of research.

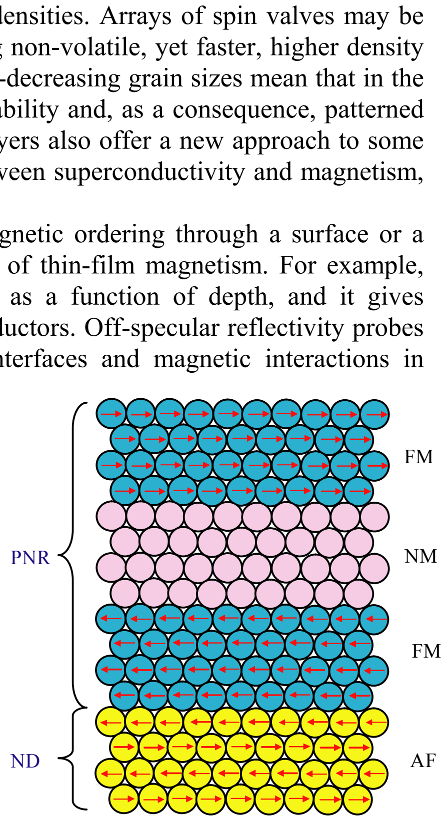

Figure 1 A Schematic of a spin valve structure. The combination of PNR and high angle diffraction will provide a complete description of the system

1.2 Spinelectronics

1.2.1 GMR Spin Valves

Magnetically coupled multilayers consisting of *3d* ferromagnetic (FM) layers interleaved with non-magnetic spacers (see Fig.1) exhibit giant magneto-resistance (GMR) [1] for appropriate thicknesses of the non-magnetic spacer layer. These are the regimes in which the oscillatory interlayer coupling has an antiferromagnetic (AF) ground state between the FM layers. The observed change in resistivity results from the spin dependent scattering of the conduction electrons, which depends on the magnetic moment alignment. It is clear that the GMR will be adversely affected if the formation of a perfect AF coupled structure is prevented by a vertically incoherent domain structure [2] or a

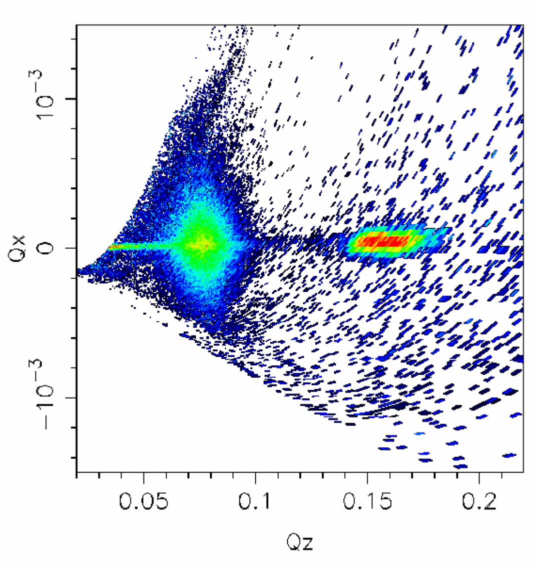

Figure 2 The diffuse scattering from a nominal [Co(9Å)/Cu(9Å)]25. The intensity centred at Q_Z~0.075$Å^{-1}$ corresponds to the AF ordering wave-vector and arises purely from the magnetic scattering. The diffuse scattering around the AF peak arises from domains.

noncolinear arrangement of FM blocks. The ability of the PNR technique, with polarisation analysis of the reflected beam, to make an *absolute* determination of the layer-by-layer magnetic structure allows very complicated structures to be studied. To optimise devices knowledge of the *magnetic* ordering and reversal mechanisms is of key importance. Real devices tend to be smaller than typical PNR samples, and the focussing capabilities of polREF will be of great benefit for these studies.

Recently, the role of the structural interface morphology has attracted considerable attention because of its important role in the transport properties [3]. For example, structural roughness on the length scale of the interlayer magnetic coupling reduces the GMR [4]. Given the *vectorial* nature of the magnetisation and the long-range exchange interaction it is clear that the magnetic disorder need not follow exactly the structural disorder. A detailed understanding of the interplay between structural and magnetic morphology is then clearly of real significance. The magnetic disorder may take the form of magnetic roughness: the correlation of the structural and magnetic interfaces or more conventional magnetic domains. Recently, PNR has made substantial contributions in relating the magnetic and interfacial disorder to the bulk magnetisation and transport measurements [5,6]. Combining the time of flight reflectometry technique with a linear detector efficiently collects not only the specular reflectivity but also a large region of reciprocal space (see Fig. 2) The polREF reflectometer has a much greater coverage of in-plane wave-vector transfer than CRISP and it will allow better surveys of reciprocal space to follow short-range correlations. The diffuse magnetic scattering is always weak, and the dramatic increase in signal will be a huge advantage.

1.2.2 TMR and CMR Devices

In principle, devices with much larger magneto-resistances and, therefore, greater sensitivity may be produced. For example, a tunnel junction may be obtained by replacing the non-magnetic spacer layer (as would be found in a GMR device) with an insulating layer. The tunnelling current through the insulating layer is very sensitive to the mutual orientation of adjacent FM layers, and large tunnelling magneto-resistances (TMR) have been reported. Unfortunately such results are strongly temperature dependent, possibly due to misaligned spins at the interface. If such TMR devices are to be commercially realised such difficulties need to be overcome. Colossal magneto-resistance (CMR) has generated enormous experimental interest due to the extremely large change in magneto-resistance associated with the metal-insulator transition in systems such as the manganites. Moreover, the conduction bands of such materials are fully spin polarised leading to exciting prospects for spin injection devices. At the moment CMR position and timing sensors are used to reduce emissions from car engines. The performance of such devices, as with tunnelling, is critically dependent on the magnetic properties of the interfacial region. We expect that polREF will make significant contributions to these new areas for the reasons outlined above for GMR devices.

1.2.3 Exchange Bias

Exchange bias, in which hysteresis loops are shifted from the zero-field axis, has been observed in a variety of systems comprising FM materials in contact with AF materials. The origin of this phenomenon is the interfacial exchange interaction between the FM and AF layers, but there are many unresolved issues. For example, the biasing fields are much smaller than expected, and it is a long-standing puzzle why exchange bias is observed at all for interfaces that are rough on an atomic scale. These issues are of enormous technological importance since GMR spin valves and tunnel junctions rely on the exchange biasing of one of the FM layers to magnetically pin this layer. High-angle neutron diffraction (ND) on polREF will allow the investigation of the AF ordering in epitaxial systems (See Fig. 1).

1.2.4 Spin-injection in Semiconductors

Although many hybrid FM/semiconductor device designs have been proposed, finding materials with which to implement them is a significant experimental challenge. Desirable properties include half-metallicity, and PNR allows the degree of spin polarisation to be directly determined in the interfacial region. Silicon-based spin transistors are being developed using different tunnel barriers in order to deal with the conductivity mismatch at the metal-semiconductor interface. The increased flux and dynamic range of polREF will enable the interfacial region to be studied with a new degree of statistical accuracy. For example, a non-uniform magnetisation profile, such as reduced magnetisation in the near interface region, would equate to a reduced polarisation and therefore a reduction in the spin injection.

1.3 Magnetic Nanostructures

1.3.1 Magnetic Dot Arrays

In the search for ever higher recording densities conventional recording media are approaching the so-called superparamagnetic limit as the size of the data bits is becoming increasingly small. As we approach this limit, the stored data becomes thermally unstable and it is unreliable. To increase storage densities to the order of Tera-bits/square inch (10^{12}bits/in^2) requires new approaches using nanotechnology. One promising idea is to pattern laterally magnetic films. Several techniques are currently under consideration for the self-assembly of regular arrays of 0D magnetic islands. Such patterned elements can only support two magnetic states, and the resulting small size features lend themselves to extremely high recording densities. To fully understand and optimise such systems requires a full knowledge of the magnetisation density within the magnetic islands and the nature of any magnetic interactions between the dots. polREF will provide such information on both the structural and magnetic order through the harmonics of the relectivity. It is increasingly recognised that the interplay between the physical and magnetic structure can significantly influence the device performance. Studying how the patterned magnetic structure responds to external influences (see sample environment) such as temperature and magnetic field and intrinsic parameters such as the size and shape of the elements will lead to greater insight into these challenging nanomaterials. Given the complexity of such experiments the literature contains few examples of such studies [7,8]: a scarcity that the capabilities of polREF will address. Such measurements will complement those eventually possible on the offSPEC beam line utilising the more difficult spin echo technique, which potentially gives full two-dimensional information on the in-plane correlations. Although the in-plane correlations are projected onto one dimension for polREF essential information on the relation between structure and magnetism and magnetic interactions will be available immediately.

1.3.2 Magnetic Stripes

Another aspect of nanotechnology where polREF will be able to make an immediate impact is the study of magnetic nanowires (see Fig. 3). One-dimensional magnetic stripes may readily be grown at the step edges on miscut substrates. In this case the geometry of polREF is ideally suited to measure the Bragg peaks from the wires, enabling a detailed comparison between structure and magnetism and a study of the development of magnetic interactions. [In principle, the magnetic stripes should not exhibit long-range magnetic order at all, but the magnetic interactions between the stripes are believed to be the key ingredient leading to ordering.]

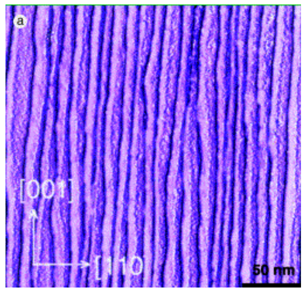

Figure 3 Fe stripes at the step edge of miscut Si.

1.4 Fundamental Magnetism

1.4.1 Superconductivity

There has recently been much interest in the interplay between magnetism and superconductivity. For example, knowledge of the magnetic induction profile at the surface of a superconductor allows one to test ideas in the Ginzburg-Landau theory. PNR has been demonstrated to provide valuable information on such parameters as the penetration depth [9], which are less readily accessible through other techniques. Furthermore, PNR has been used to study magnetic vortices, which form in type-II superconductors [10]. The current transport in such systems, which is crucial to technological applications, depends on the pinning of such vortices. Moreover, it is well known that the properties of thin film superconductors can be modified when they are in contact with a ferromagnetic material: the proximity effect. PNR is able to provide information on both the flux penetration and the magnetic structure. The coexistence of 3D superconductivity and magnetism has recently been discovered at ISIS [11]. The superconducting ordering temperature of blocks much thinner than the coherence length is comparable to the bulk for an antiferromagnetic alignment of ferromagnetic blocks, but the superconductivity is suppressed when the orientation is ferromagnetic, suggesting new possibilities to study the interplay between these usually mutually exclusive phenomena and the potential for a superconducting spin valve (See Fig. 4). The enhanced flux, coupled with a greater range of external parameters (temperature, magnetic field) on polREF will allow data collection of high statistical quality (essential for the study of such subtle phenomena) and will provide greater insight into the ordering mechanisms in such systems.

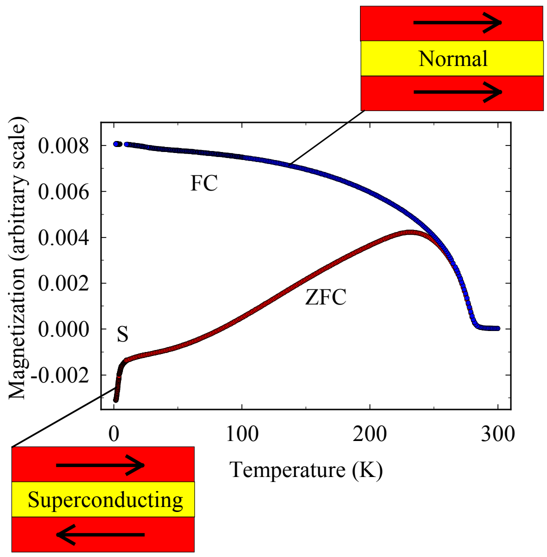

Figure 4 polREF will allow the study of proximity effects in superconducting/magnetic systems.

1.4.2 Model Magnetic Systems

The influence of the surface on phase transitions is a problem of fundamental importance in statistical physics. Despite considerable progress in theoretical studies of surface magnetic phase transitions, attempts to verify the predictions experimentally have been thwarted by the severe demands on surface quality. However, in magnetic multilayers one can study the first few blocks of magnetic material rather than the first few atomic planes. The surface spin-flop transition predicted for a chain of XY spins with AF interactions and uniaxial anisotropy was recently detected using PNR [12]. In a similar approach a ferromagnetically coupled multilayer grown on an antiferromagnetically coupled multilayer has been used to shed light on the mechanism of exchange bias [13] (Fig. 5). Many other problems can be approached in this manner, and polREF will provide the sensitivity required to study weak features such as domain walls.

1.4.3 Lanthanide and Actinide Research

The large magnetic moments and anisotropies in lanthanide systems make them of great interest. An AF neutron diffraction peak has been observed from a Ho film only 46Å thick using ADAM at the ILL [14]. The high-angle diffraction capability of polREF will give wave vector information on wavefunctions in thin films, providing new opportunities to study quantum size effects. Given the large anisotropy within these systems the provision of a high magnetic field (>10T) cryomagnet at low temperatures (<2K) will vastly extend the parameter space available for such studies. The actinide elements exhibit behaviour somewhere in between those of the transition metals and the rare earths. Surprisingly, uranium based multilayers have been considered as candidate technologies for high density recording media due to the enormous magneto-optical effect. The quantities of radioactive materials in such samples are very small (typically <100μg) thereby minimising the radioprotection requirements. Such studies are still difficult and limited due to sample preparation but these issues are being actively addressed and such topics as induced magnetism, hydrogen loading *etc.* in actinide systems will offer new research possibilities.

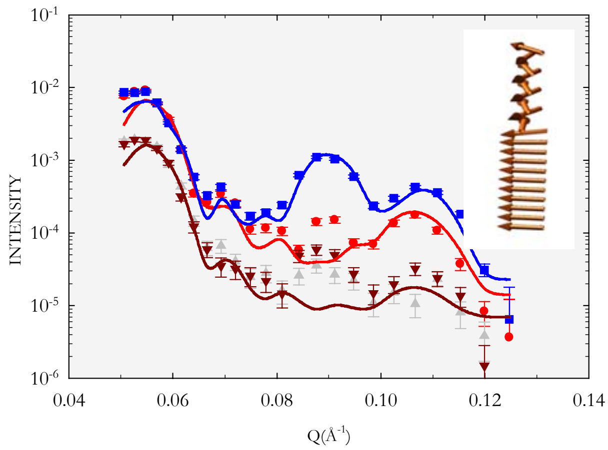

Figure 5 Direct observation of a spin flop phase from PNR data in a model exchange bias system.

1.5 Additional areas of research and future potential

1.5.1 Neutron Depolarisation

The ability to study the three dimensional structure of ferromagnetic domains is of relevance in high performance metallic alloys and in the optimisation of neutron polarising mirrors. The technique [15] involves measuring the 9 (XYZ) spin dependent cross-sections. The flexible beam polarisation on polREF will facilitate such measurements, which when coupled with the large gain in performance will allow more parametric and angular dependence studies.

1.5.2 The Inverse Phase Problem

In principle, a complete determination of the *complex* reflection coefficients in a reflectometry experiment is possible using magnetic reference layers [16]. This is commonly referred to as the inverse phase problem and is well known in both neutron and x-ray scattering communities. The addition of an additional scattering potential (the magnetic reference layer) allows one to solve uniquely the scattering potential profile, which gave rise to the observed reflectivity. Experimentally, such ideas are complicated through a finite wave-vector range, statistics and the need for a complete characterisation of the reference layer. Clearly, polREF will significantly contribute in wave-vector range and counting statistics. Such techniques offer promise for complicated *biological membrane* structures where a-priori knowledge is not readily available [17].

1.5.3 In-situ growth

An exciting possibility for polREF is the addition of a UHV deposition chamber at the sample position. Many of the recent advances in magnetic nanostructures have been driven by developments in deposition techniques such as sputtering and molecular beam epitaxy (MBE). The ability to study magnetic surfaces during *in-situ* growth with reflectometry would provide a new and unique insight into the growth and magnetic ordering mechanisms in magnetic thin film systems. The avoidance of surface contamination or capping layers can be crucial in, for example, understanding finite size effects [18].

2 Technical Case:

The design of polREF is such that it dramatically extends the measurements currently possible on existing polarised neutron reflectometers. It clearly complements the science case presented for the offSPEC proposal in the study of in-plane order/disorder and patterned substrates, and the design also allows surface chemistry experiments of the type discussed in the INTER case.

2.1 Outline Technical Specification:

The outline technical design of polREF has been developed to optimise the possibilities for PNR on the ISIS second target station in the following key areas:

- Enhanced Flux at the sample position
- Maximise use of the increased wavelength band
- Minimise the noise floor on the instrument through design and shielding
- Secondary shutter to reduce access time
- Flexible neutron polarisation control

- Optimised sample environment

These points are addressed in the following sections. The performance summary is presented in table 1.

Moderator	Coupled grooved cold H_2/CH_4
Incident Wavelength	0.9Å - 15Å
Resolution	~3-10%ΔQ/Q
Flux at the sample position	~10^7 n cm^{-2} s^{-1}
L_1	23m
L_2	3m
Beam size	60 mm x 30mm H x V
Performance	×15-23 over ISIS Target Station

Table 1 Outline design specification for the polREF reflectometer

2.2 Neutron collection and transport: optics

The beamline optics (see Fig. 6) will consist of a channelled beam bender and a tapered guide. The bender will bend the neutron beam down by 2.3° (ensuring compatibility with the *inter* reflectometer proposal for liquid surfaces, ferrofluids *etc.*) and minimising the fast neutron and γ-ray background from the direct line of sight with the moderator. The bender design is being optimised but is likely to consist of a long bender with multiple channels. An interesting possibility is that the bender may be mounted in the primary shutter although this represents an engineering challenge to ensure repeatable positioning. A secondary fast shutter will be used to control the neutron access to the blockhouse. Long wavelength neutrons will be removed through a frame overlap mirror.

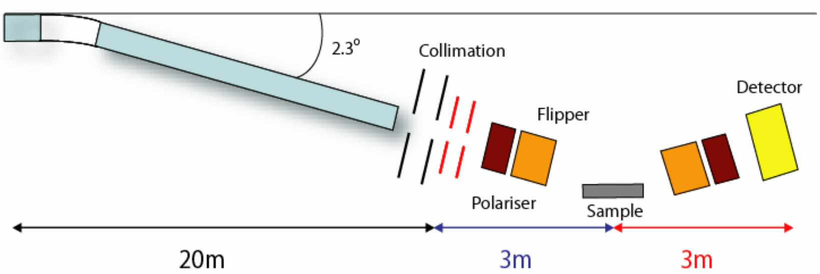

Figure 6 An Outline layout for polREF

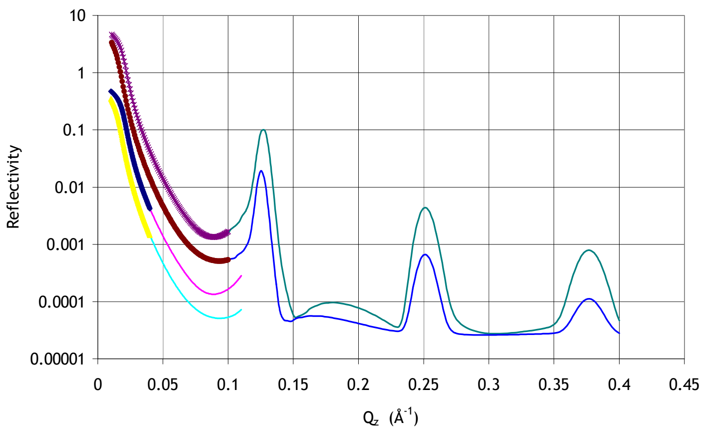

Figure 7 Comparison of the accessible wave-vector range on CRISP in two angular settings (bottom) against polREF (top) . The curves have been offset for clarity.

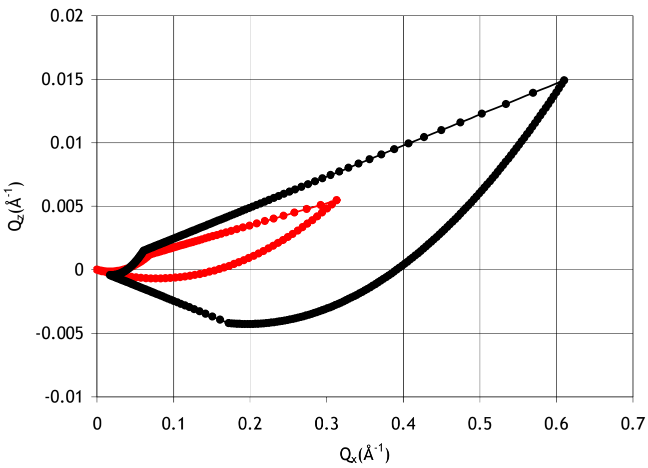

Figure 8 The accessible wave-vector range on CRISP (red) for a typical experimental geometry and polREF (black).

Samples for PNR experiments are typically much smaller (~1cm^2) than those encountered in surface chemistry (~40cm^2) for example. In addition, there is poor resolution in the horizontal plane (*cf. offspec)*. One can then use this to gain in neutron flux by tapering the guide in the horizontal direction at the expense of the wavelength dependent divergence. Monte Carlo simulations are currently underway to calculate the acceptable horizontal divergence without compromising the vertical resolution. To minimise scattering and absorption the system will be kept under vacuum and windows kept to a minimum. At the current state of design, the relative merits of background suppressing and wavelength defining choppers against the loss in short wavelength flux are being assessed. The large gains in wave-vector range both specular and diffuse coupled with the optimised flux will lead to a large increase in data collection efficiency. This will allow one to study smaller samples (an increasing requirement for patterned samples), more complex sample environment or to survey reciprocal space.

2.3 Polarising and Flipping Elements:

In the wavelength range (1-14Å) of interest on polREF, polarising supermirrors match the requirement for a high polarisation of >95%, high reflectivity >80% and transmission >90%. 80% reflectivity up to m=4 is now realisable. To simplify the subsequent beamline the polariser will be run in a transmission mode with the flexibility to run in reflection should there be new polariser designs, background minimisation issues to be addressed.

For the analysing assembly it is clear that the ability to analyse a diffuse beam is essential. A curved stacked supermirror assembly (oriented perpendicular to the incident polariser) provides a high degree of polarisation with good angular coverage. The design is finalised but may take advantage of the solid state polarisers developed at HMI [19].

The flipping elements are characterised by a high flipping efficiency (>99.9%) over the entire wavelength range and over the beam size (40mm). This can be realised by several technologies relying on either adiabatic or non-adiabatic rotation of the neutron spin. In addition, the flipper should not introduce significant background and be tolerant to stray magnetic field (associated with the cryomagnet for example). Technologies currently under investigation include Drabkin spinflippers and adiabatic rotators [20] both of which are in use on the CRISP reflectometer at ISIS. The flippers will also be used to control the incident polarisation onto the sample allowing us to access all the elements of the polarisation matrix. Permanent magnets will provide the neutron guide field. Design effort will be given to minimising the effects of stray magnetic fields on the polarising and guide field elements and the need for active compensation control.

2.4 Sample Environment:

The science case determines the required sample environment, which consists of primarily two regimes. For many transition metal systems room temperature and low magnetic fields are required. Of more importance is the accurate control of the applied magnetic field. This will be achieved through a computer controlled bipolar electromagnet ($\mu_o H<1.5T$). In parallel, *in-situ* Magneto-Optic Kerr microscopy will provide detailed information on the magnetic configuration complementing and maximising the efficiency of the neutron measurements. The rare earth and superconductivity measurements are characterised by low temperatures (<2K) and high magnetic fields (~10T). These can be realised using a superconducting magnet. In addition, the proposal requires a magnetic field (~0.5T), which can be applied in any direction preferably with a maximum vertical field to pull in-plane magnetism out of the plane. Such a magnet is matched to the controllable incident polarisation allowing access to the full elements of the polarisation matrix. All of the beamline sample environment will be mountable on heavy duty (100Kg load capability) high precision stages.

2.5 Neutron Detection:

For high count rate, high signal-to-noise ratio measurements a ^{3}He single detector will be required. For the off-specular and out of plane diffraction measurements a 200mm×200mm area detector with a resolution of <1mm. With a detector distance of 3m this represents an angular resolution of ~0.02° which is well matched to the upstream resolution. To study the structural and magnetic ordering on an atomic length scale polREF will require a high angle detector capability. To maximise the intensity from the broad reflections, typical of thin film samples relaxed collimation will be employed.

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