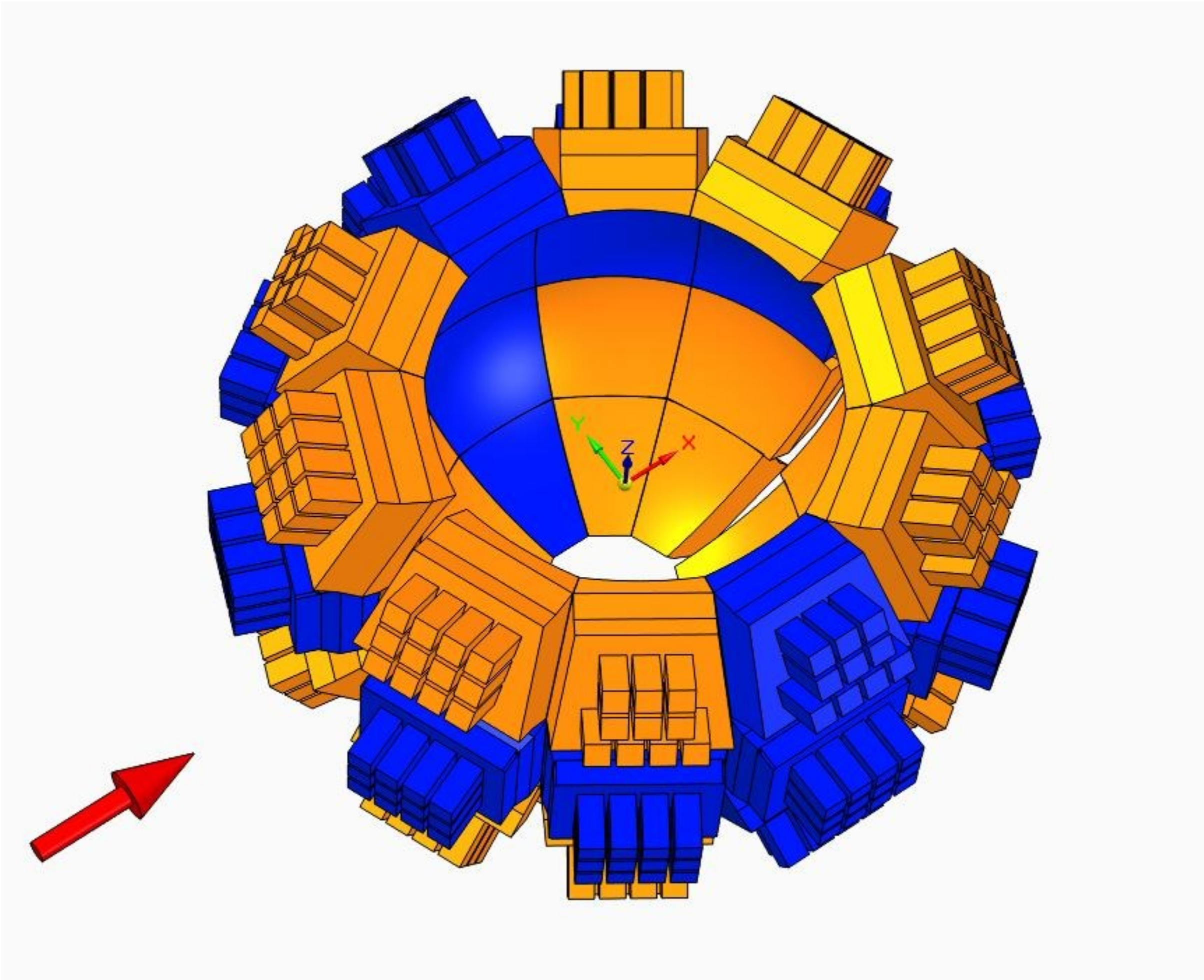
A new single crystal and thin film diffractometer WISH-II and a polarization upgrade on WISH



Project overview

The project is twofold: building a new single crystal and thin film cold neutron diffractometer WISH II with a large continuous detector coverage (approx. 2.5π) and an upgrade to the existing WISH beamline to implement polarized neutron measurement capability - which together promise transformative breakthroughs into Advanced Manufacturing, Materials of the Future and Clean Growth.

1. Background

Neutron scattering is still by far the easiest and most complete technique to provide the detailed microscopic understanding required to engineer and fine tune the properties of a whole variety of quantum materials capable of bringing transformative technologies on a par with the still ongoing semiconductor revolution. The magnetic diffractometer WISH, primarily designed for powder samples, is now a mature instrument and its excellence is recognized worldwide as demonstrated by its impressive oversubscription and scientific output. Over the years, the science programme has also evolved towards more single crystal experiments and now encompass topics such as thin films and Metal Organic Frameworks (MOFs). By combining advances in detector technology and innovative design, we propose a new instrument, called WISH-II as a working title, which will complement the existing one by focussing on single crystal and specialized high pressure powder experiments on these emerging (for neutrons) but extremely exciting topics. By absorbing part of the powder programme, the new instrument also provides an opportunity to upgrade the existing WISH with polarisation and tackle completely new science. In the coming few years, many single crystal instruments at other facilities are likely to close whilst only a few new instruments directly aiming at the strong WISH science case are planned at other facilities. For ISIS to remain competitive in this hugely strategic scientific portfolio, encompassing **Advanced Manufacturing, Materials of the Future and Clean Growth**, a state-of-the-art beamline with new capabilities is required as soon as possible.

The project will deliver a new cold neutron diffractometer on the second target station **optimized for the study of small single crystals and epitaxial thin films** with a tuneable horizontal and vertical divergence, whilst also allowing powder experiments. The proposed diffractometer will have a primary flight path of 40m with m=3 elliptic guides and will view the decoupled solid methane moderator (currently undergoing an upgrade to improve its line shape). The detectors will span a continuous spherical coverage from 175° to 5° in 2θ and from +45° to -75° out of plane using the new wave shifting fibres technology with a ~4x4mm^2 pixel size. A fully evacuated secondary flight path of 1.1 m and a series of compact 3D printed collimators tailored for each type of experiment will ensure a very high signal to noise ratio and low background. A proposal for the installation of a polarizer and an analyser on the WISH instrument is also put forward. A 1m long section of the existing elliptical guide on a carousel will host a V-cavity polarizer (the second half of the guide is "polarization ready") to achieve polarization over a wide wavelength range. The proposed analyser bank, which will sit on the current collimator oscillation mechanism, will employ an innovative log spiral (m=5.5) analyser, validated through Monte-Carlo simulations as well as by a recent prototype test on LARMOR, to provide excellent performance at λ > 2 Å. It will cover ~30° in plane and out of plane, and can be placed anywhere along one side of the instrument to maximize the q-coverage.

2. Science case

The excellent characteristics of the WISH beamline in terms of flux, high signal:noise ratio, resolution and its flexibility to easily switch between powder/single crystal modes made it extremely successful. Indeed, over the past 10 years, WISH has strengthened its high reputation with the international user community. Primarily designed as a magnetic powder diffractometer with a capability to do single crystal measurements, WISH quickly proved to be extremely efficient in a much broader scope. In particular, studies of **magnetic thin films and energy materials** based on **metal-organic frameworks** (MOFs) became a significant part of its scientific programme and led to many high impact works [1,2,3,4,5]. The permanently high oversubscription rate (2 to 5) and very impressive scientific output of WISH (see business case) are both natural drivers for the further development of a cold neutron diffraction instrument on the second target station. However, the present project is much more than simply expanding capacity.

The main drawback of the WISH beamline is the limited detector coverage dictated by its compatibility with the large 14T cryomagnet, an essential strategic piece of sample environment at ISIS to remain competitive with other sources on the extreme sample environment front. Avoiding this high magnetic field option (but retaining lower field capabilities), a further optimization is possible by a significant increase of the detector coverage, as illustrated in Figure 1, and this constitutes the main concept behind the WISH II project. Realization of this concept will be hugely beneficial for many parts of the broad scientific programme developed over the past ten years of operation of the WISH beamline.

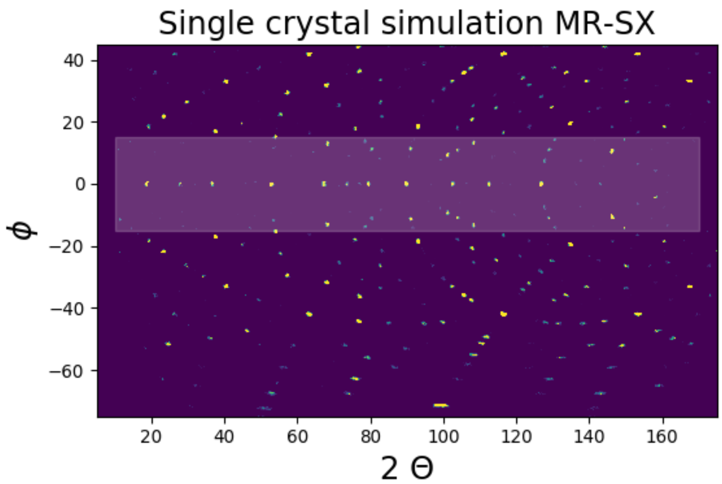


Fig 1: simulated diffraction pattern from a C_{60} crystal on half the detectors proposed on the WISH-II instrument showing the large number of accessible reflections compared to the existing WISH instrument (shaded region)

One key aspect of the proposed coverage increase is that it will **enable a quantitative characterisation of single crystal data**, taking full advantage of the wide wavelength range available, which itself is one of the main strengths of the instrument. More precisely, our data analysis work over the past 4 years has highlighted that the limited out-of-plane detector coverage does not allow the collection of a sufficient number of diffraction peaks to model extinction effects properly. This can be somewhat mitigated on WISH in some favourable circumstances (within a basic sample environment) by using a goniometer or by remounting the sample, but it severely restricts the possible external parameter range as well as being very time consuming and therefore not practical.

In addition, the enlarged coverage will be a great advantage for **thin film** experiments since several symmetry equivalent directions will be measured at the same time which is the crucial factor to **understand and reconstruct magnetic and structural domains**, largely affecting properties of these samples. This is illustrated in Figure 2 below where all interesting peaks could be acquired in one shot on WISH-II instead of 4 rotations on WISH.

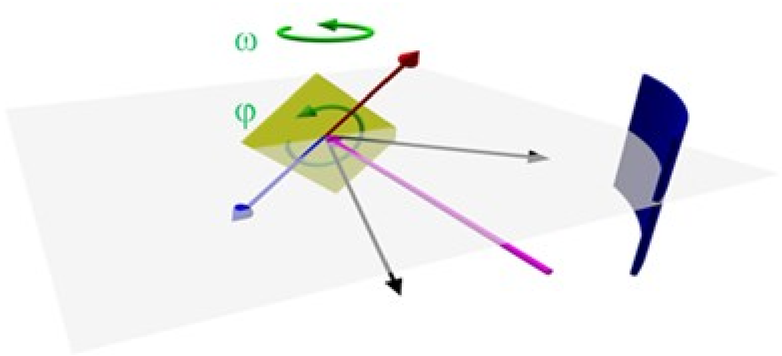


Figure 2: scattering geometry for a $BiFeO_3$ thin film. On WISH, several φ rotations are required to capture domain information due to detector coverage limitation.

This time constraint is currently a major obstacle in fully exploiting such experiments and prevent detailed studies as a function of external parameters. This stems from the fact that interesting thin films that retain the epitaxial growth are typically **5mm*5mm*30nm** i.e. less than 0.001mm^3 on top of a much larger signal emanating from the substrate and counting times of the order of days are not uncommon. The competition with powder measurements for which, for instance, the full temperature dependence of several samples can be measured in a single day means it is impossible for these finer detailed thin film experiments to take place. This is however an area for which the magnetism community has a huge and increasing appetite, driven by the ability to tailor the magnetic properties of a system and the direct application potential for making devices ranging from spintronics and the exquisite control of spin current polarisation to multi-caloric refrigeration. In these thin film and hetero-structure systems, understanding the changes to the magnetic structure of the nm-thick film driven by compressive or tensile strain stemming from the substrate and comparing with theoretical calculations from Density Functional Theory is absolutely key in designing the next generation devices. WISH-II will allow the community to increase the existing synergy with Diamond on thin film studies by allowing complementary neutron measurements and synchrotron magnetic diffraction and imaging experiments on identical samples [6]. **Topological materials** are another extremely active area of condensed matter physics, distinguished by the 2016 Nobel Prize in Physics, which is poised to benefit from the proposed instrument with many exotic phases predicted and some eventually observed in real materials. Common to all these exotic electronic states is their protection by some symmetry elements. Clearly, the introduction of magnetic order and associated breaking of time-reversal symmetry can provide another handle for experimentalists to play with. This is especially true for cases where different types of magnetic order (these changes in magnetic structure can be very subtle and require the best instrumentation), and therefore different symmetries can tune the electronic structure by removing/adding band degeneracies. A recent well cited WISH publication [7] on such a system demonstrates the impact of properly understanding the magnetic structure for comparison with ab-initio calculations. For these systems, high quality samples are difficult to synthesize in powder form but are more readily available as small single crystals. Therefore, the ability to collect as many magnetic reflections as possible on a single crystal to extract the magnetic structure from quantitative refinement is a key requirement for ISIS to cater for this highly dynamic community. This will nicely complement ARPES work done at Diamond.

Another drawback from the existing WISH is the "limited" q-range despite covering an already impressive 0.6-50 Å in d-spacing. For systems with very long-period magnetic structures, it is sometimes necessary to run the instrument in double frame mode (5Hz) thereby immediately increasing counting times by factor 2. This is compounded by the fact that the low angle scattering bank is noisier than the others. We propose to remedy these issues by extending the 2θ coverage from 10°-170° to 5°-175° in the scattering plane (and even 2.5°-177.5° out of plane) as well as placing the whole detector array in vacuum. This will at the same time allow the users to better exploit the instrument's **diffuse scattering** capabilities, especially for magnetism. Indeed, as shown in figure 3, the q-coverage at low q is rather "pointy" (as well as noisier due to the detectors not being in vacuum) if only one rotation is collected and many runs are needed to cover this low-q region properly. By extending the lower scattering angle by 5 degrees, the width of that small region is extended and fewer rotations will be needed. This significant gain in the low q region is of course additional to the huge increase in out of plane coverage which will allow many planes to be collected simultaneously. We also propose to add a moveable small angle bank within the get lost tube to achieve even lower q. Whilst we do not seek to compete with SANS instruments, having the opportunity to reach zero satellites reflections on **systems with very long modulation** (e.g. $BiFeO_3$) and **skyrmion** and collect diffraction data at the same time would clearly be a unique asset.

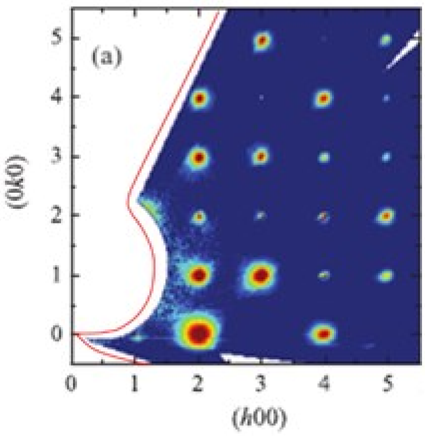


Figure 2: Example of diffuse scattering data in the horizontal scattering plane, with a sample aligned with the c-axis vertical, collected on WISH from a single shot (no merging of rotations) on one of the instrument side. The red lines delimit the extension of reciprocal space coverage due the increased 2ϑ range. The greatest improvement occurs at the lowest q and will consequently reduce the number of rotations needed to cover the lowest q.

WISH's existing vibrant MOFs research programme is also poised to greatly benefit from the proposed new instrumentation. In reality, this interest could be extended to any large molecule systems especially when considering the fact that experience on WISH has shown that it is possible to observe reflections from a sub-0.01mm^3 crystal all the way up to 55 Å in d-spacing [8] (in double frame mode on WISH, which would be accessible in single frame on WISH-II) and **comfortably collect data with good signal:noise ratio with crystals ~0.1mm^3**. The crystal structure of these materials is usually characterised by large unit cells with many independent atoms and precise determination of their positions and/or magnetic moments requires a large number of reflections. In recent years, the number of powder MOFs experiments has been quickly going up and WISH II will enable to significantly extend this programme by covering single crystal samples. Another emerging topic in condensed matter research is the ability to **tune electronic properties of various systems directly via strain**. This is however generally limited to bulk measurements or synchrotron due to the requirement having small single crystal samples to be able to apply significant strain. We have recently demonstrated that it is possible to do such experiments on WISH [9]. However, the geometry constraints (applied strain direction vs available reciprocal space) are severe and basically imply each experiment is bespoke requiring very careful planning in terms of strain direction and visible reflections in the horizontal plane. The increased coverage allowing many reflections out of plane to be collected will transform this type of experiments.

The powder option of WISH II will also offer a significant improvement compared to WISH especially for the 90-degree scattering geometry, which is the most appropriate for **high pressure experiments**. This has been clearly demonstrated by the success of the existing PEARL beamline due to shape of the Debye Scherrer cones and the fact that it is easier to achieve good collimation near 90°. WISH has already proven itself to be very good at these high-pressure experiments on large unit cells (such as magnetic systems) but the factor four in solid angle coverage will enable to deal with smaller signals coming from either **smaller samples (allowing extended pressure range) or smaller moment systems**. The latter is particularly important for **frustrated and low-dimensional quantum systems** with substantially suppressed ordered moments, a current hot topic in condensed matter research. The increased 90 degrees coverage and its associated high collimation is also appealing for **complex experiments on MOFs** such as **high pressure volumetric sorption measurements.** The extended q-range due to the increased coverage at low and high angles is also an attractive prospect for refining complex structures (in particular a factor 2 can be gained in cases where double frame data is needed). Overall, it is anticipated that, whilst not giving the straight factor 4 gain seen for single crystal experiments due to the impossibility of exploiting the full vertical divergence of the guide across the whole out-of-plane extension, WISH-II will be at least as good as WISH in all cases and a factor 2 in other cases, including the important area of magnetic systems under pressure.

Last but not least, the release of the current pressure on the WISH beamline will allow the installation of a **polarised neutron** option on it. This will enable new types of experiments available at other neutron facilities, to be performed at ISIS, but with an unprecedented flux/resolution combination. The nature of these experiments means that counting times are at least doubled and realistically can therefore only be viable in the context where the time pressure on the existing WISH is reduced. Particularly interesting opportunities include **drastically reduced incoherent scattering from hydrogen for looking at organic compounds** when deuteration is not (or only partially) possible as well as the precise reconstruction of **spin-density maps**. Indeed, polarized neutrons provide a means to separate the magnetic components from the other contributions to the total scattering. This will be extremely useful for **molecular magnets and frustrated systems** where the magnetic scattering is weak, and will therefore be very appealing for a broad range of materials relevant to **quantum computing**. Moreover, diffraction data collected using unpolarised neutrons are sometimes unable to provide a unique magnetic structure solution and using polarised neutrons is the only way to establish the right model. Obtaining such accurate information is key for unlocking the structure/property relationship and therefore future design of new **sensors and memory storage materials**. The ability to separate chiral components readily available with polarised neutrons is also very attractive in the light of the huge recent interest in **chiral systems**. Aside from its intrinsic fundamental interest spanning multiple branches of science, chirality and its coupling to other technologically relevant microscopic quantities, offers another tuning parameter which is only recently been exploited by the condensed matter community [10].

3. Business case

Historically, the work done on the WISH instrument was mainly related to magnetic powder diffraction and to a limited number of qualitative parametric studies on single crystals. In the last 5 years the appetite for more complex, complete, and quantitative single crystal experiments and for magnetic diffraction on thin film systems has grown, putting increased pressure on the demand for the beamline. This is reflected on the instrument oversubscription, which remains in the 3-4 times range, and it is predicted to increase since **single crystal and thin film experiments require ~5 times the average powder experiment time** due to the limited WISH detector coverage. The success of the WISH diffractometer during its 10 years of operation naturally leads to a call from the stakeholders for a new long-wavelength single crystal and thin film diffractometer. Figure 1 reports the peer-reviewed journal publications record of WISH showing a steady increase in the number of papers reaching ~50 articles in 2020. This metric reveals the astonishing productivity of the beamline which generates the greatest number of papers of all the ISIS beamlines. Indeed, the WISH publications account to roughly 7% of the total ISIS journal publications, a share which in fact rises to 11% if only papers related to beamtime are considered, with a substantial part (~20%) published in very high impact journals with impact factor (IF)>8. On the current WISH diffractometer, the single crystal and thin film experiments cover ~35% of the awarded beam time and produce in average 6 papers/year with an average IF of ~8, i.e. the outputs from **these experiments are high impact**. The proposed WISH-II instrument will allow ISIS to strongly increase the number of single crystal and thin film experiments on magnetic systems performed, while increasing the total time available for powder measurements, many of which are high quality but stay below the current acceptance threshold.

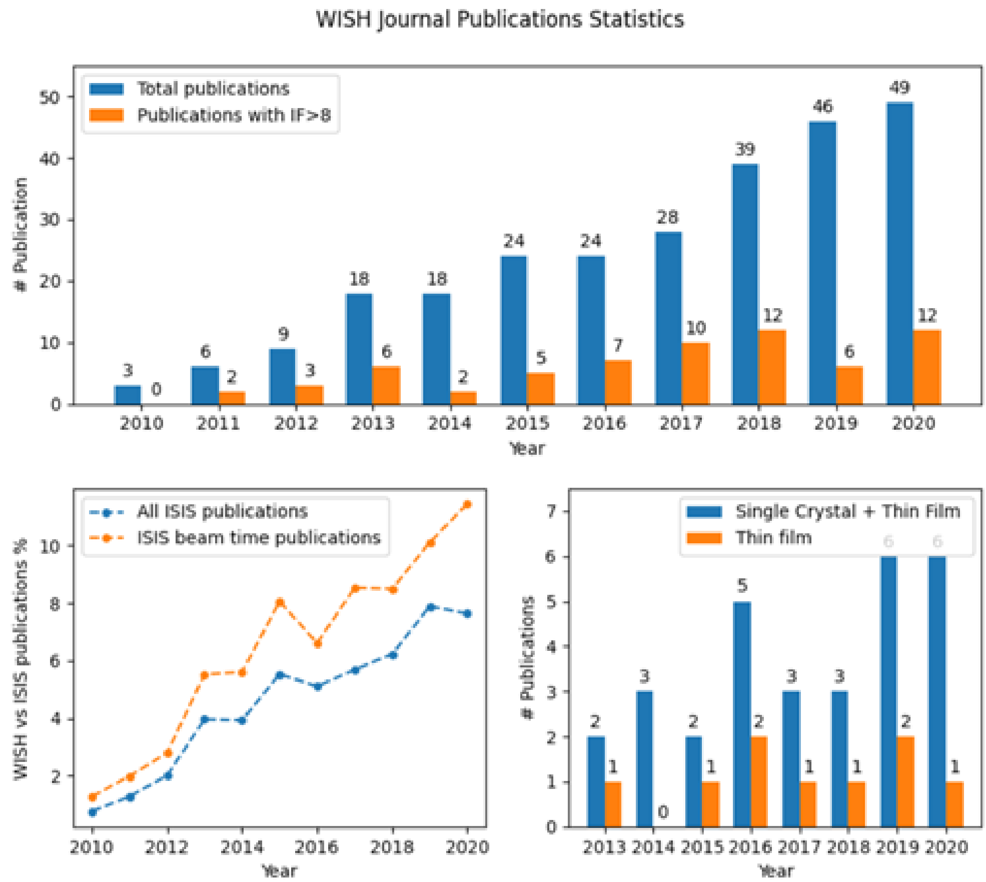


Figure 1 WISH publication record updated to January 2021 from the e-pubs archive. The top panel shows the total WISH publications per year and the relative papers published in journal with impact factor greater than 8. The bottom left panel shows the comparison with the ISIS publications and the bottom right panel shows publications linked to single crystal and thin film experiments.

Additionally, the proposed scattering geometry will allow us to perform **better high-pressure experiments**, due to the much larger 90 degrees coverage, thereby expanding the existing research programme on WISH which is also time-limited, as well as performing more routine powder experiments. This will reduce the pressure on the WISH instrument and enable the development of **the polarization option on WISH** (it is impossible for analysers to match the huge detector coverage of WISH-II) which was designed from the onset with a possible polarization upgrade that was never exploited. Without the availability of the WISH-II instrument, adding polarization on the current WISH would bring an unbearable user demand, due to the long duration of such experiments, which would be detrimental for the current science programme. **Magnetic neutron diffraction** exploiting a **polarized beam** is a **powerful technique that is currently absent at ISIS**, leaving the facility behind with respect to its competitors in Europe and worldwide. Building on the recent success in implementing polarized neutrons on LET at ISIS, the opportunity of using the in-house technical know-how should be seized so ISIS can expand its scientific capability to include magnetic diffraction. Indeed, the polarization design for the analyser part of WISH has now matured and recent Monte Carlo simulations on an innovative log-spiral wide analyser show it is possible to exploit most of the wavelength band of the moderator (above 2Å) over a solid angle covering a typical WISH detector bank. This promises to bring transformative experiments to the beamline and finally reach out to a **significant fraction of the UK magnetism community which, for the moment, has to perform its experiments at the ILL**.

4. References

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