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ToF-Backscattering spectroscopy at the ISIS Facility: Status and Perspectives

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Abstract. The cold neutron spectrometers IRIS and OSIRIS at the ISIS Facility have evolved over the past decades continuously. Both spectrometers are inverted geometry instruments using time-of-flight to determine the incoming energy and Bragg reflection for final energy analysis. The scientific applications are diverse and range from quasielastic investigations of molecular diffusion in nanoporous materials to high-resolution spectroscopy of magnetic excitations. Here we will present upgrade opportunities toward improved energy resolution and increased flux which will keep the instruments competitive over the next decade and beyond.

1. Introduction
The backscattering technique with neutrons was developed about five decades ago in Garching, Germany [1]. This type of spectrometer promised the highest energy resolution of about 1 μeV at that time and hence access to 1 ns relaxation times. Most of the following developments focused on enhancing the flux and to this end the most modern instruments exploit phase space transformation [2, 3, 4].

At pulsed sources the idea of backscattering from an analyser crystal has also been applied and combined with an analysis of the incoming energy by measuring the time-of-flight (tof) to the detector. One of the first instruments of this type was IRIS at the ISIS Facility, UK [5, 6]. The chosen setup was different to the classical backscattering spectrometers as the cooled pyrolytic graphite analysers are placed in a near-backscattering geometry. The energy resolution was relaxed to about 15 μeV, but the dynamic range was significantly larger compared to classical reactor-based backscattering spectrometers. That combination of high resolution over a wide range in momentum transfers with a large dynamic range compared to classical backscattering spectrometers allowed novel applications in cold neutron spectroscopy. With the OSIRIS spectrometer a large gain in intensity was achieved by using a supermirror guide and an increased analyser unit [7, 8]. A decisive step toward the μeV resolution regime at spallation sources was undertaken at the spallation neutron source SNS at Oak Ridge, by combining a long flight path with a large silicon analyser (SA) [9]. As a natural further development tof backscattering spectrometers with a silicon analyser using a pulse-shaping chopper have been built or proposed [10, 11].

IRIS is a day-one instrument which opened in the 1980s and uses a Ni-coated guide terminated with 2.5 m of supermirror focusing guide. OSIRIS is the last of the first-generation of instruments
at ISIS and has a curved $m = 2$ supermirror guide. Both spectrometers use the reflections of cooled pyrolytic graphite crystals to define the final energy. To remove higher order reflections, a cooled beryllium filter is installed between sample and analyser [12]. More details about the energy resolution and performance have been obtained through extensive Monte Carlo simulations, see e.g. Refs. [8, 13, 14]. IRIS is additionally equipped with a Mica analyser bank to achieve higher energy resolution of e.g. 11 $\mu$eV with the 006 reflection.

The strength of both spectrometers lies in the combination of relative high energy resolution due to the near backscattering geometry with a large dynamic range (up to several meV in energy transfer) due to the tof-technique. That virtue enabled unique experiments in the field of correlated electrons [15], research on Energy related materials and catalysis [16]. The need to cover a wide range of relaxation dynamics over several decades in time in complex materials and limitations in studying ionic conductivity in novel battery materials are exemplary drivers for new capabilities. The IRIS Mica option is often not viable due to the loss in intensity and the restricted momentum transfer range for lower reflection orders.

These performance limitations triggered a proposal to upgrade the OSIRIS spectrometer with a SA bench [17]. It will improve the spectral resolution by a factor of 2.5 to $\Delta E \approx 10$ $\mu$eV. In the following the proposed SA upgrade will be discussed in more detail and at the end we will present further directions of development, which are under investigation.

2. New capabilities
From a previous study on the resolution function of the OSIRIS spectrometer it became clear that the dominating part for the resolution stems from the analyser geometry [14]. To improve the energy resolution, silicon is considered as analyser material which is the standard at classical backscattering spectrometers and nowadays also on high-resolution tof-backscattering instruments [18].

There are several advantages in using silicon compared to the present pyrolytic graphite. Because Si(111) has no second order reflection, there is no need for the installation of a cooled beryllium filter. Furthermore, silicon does not show strong thermal diffuse scattering as it was demonstrated recently by the DNA spectrometer with a very high signal to noise ratio, approaching $10^5$ [10]. However, there is a disadvantage with silicon crystals concerning its neutron reflectivity: the crystals are too perfect for the required resolution, in particular, for the more relaxed resolution in near-backscattering geometry. For the Si(111) reflection, the energy resolution contribution from the Darwin width is 0.077 $\mu$eV [18] and hence about a factor 100 too small for OSIRIS. That mismatch with the resolution of the primary spectrometer would result in a substantial intensity loss. Since many decades standard practice is to deform elastically the crystals, which increases the reflected intensity. The elastic bending of the wafers creates a gradient in lattice spacing which depends on the curvature radius of the analysers $R_A$ and the thickness of the wafers $D$ [2]: 

$$\Delta d = \mu_{eff} D R_A$$

with $\mu_{eff} = 0.44$ an average Poisson ratio and the negligible contribution from the Darwin width has been omitted. To calculate analytically the expected energy resolution for a silicon analyser setup on OSIRIS, we are using the same formalism and parameters as previously [14, 19]:

$$\frac{\Delta E}{E} = 2\left\{(\frac{\Delta f}{f})^2 + (\frac{\Delta L}{L})^2 + (\frac{\Delta d}{d})^2 + (\cot\theta\Delta\theta)^2\right\}^{1/2}$$

(1)

For monochromatic focusing conditions of a Bragg-reflecting spherically bent crystal the focusing distance $f$ is $f = \frac{R_A}{2} \sin\theta$ with $\theta$ the Bragg angle and the object and image distances are equal to $2f$ [20]. The angular term of the analyser resolution is determined by the near-backscattering Bragg angle $\theta = 83^0$, which is imposed by engineering constraints, and the angular spread $\Delta\theta$. For a crystal with incoming $\alpha_i = 2^0$ and outgoing $\alpha_f = 0.5^0$ divergences and negligible mosaic spread $\Delta\theta$ can be written in the following form [21]: $\Delta\theta = \frac{\alpha_i \alpha_f}{\sqrt{\alpha_i^2 + \alpha_f^2}}$. For the thickness of the
wafers, we assume $D = 0.8 \text{ mm}$ and a radius $R_A = 880 \text{ mm}$. With all these input parameters, we calculate the energy resolution around the final energy $E_f = 2.11 \text{ meV}$. Figure 1 shows the resulting energy resolution for energy transfers around the elastic line. The calculated resolution for the PG analyser agrees perfectly with the measured value from a vanadium measurement at the elastic line and with Monte Carlo simulations [14]. With the SA the energy resolution will approach $10 \mu \text{eV}$ at the elastic line, hence an improvement by a factor 2.5. That value is very similar to the resolution attainable with the Mica006 reflection on IRIS. Included is also the contribution solely from the primary spectrometer, which dominates the resolution. The part of the secondary spectrometer is $\Delta E_f = 4.6 \mu \text{eV}$. In the time domain, relaxation times up to around 400 ps will be accessible and in total about 3 decades in relaxation times will be covered over a wide range of wave vectors ($Q = 0.2 - 1.9 \text{ Å}^{-1}$) in a single setting.

In Figure 2 an engineering view of the new setup is shown inside the OSIRIS tank. Clearly visible are two SA benches and their respective detector arrays. The detectors are arranged on a circle around the 400 mm diameter sample bin. The detectors for the new setup will be linear position sensitive He3 tubes with a half inch diameter, a half inch position sensitivity and an active length of 6 inches. The position sensitivity will provide two advantages compared to the detector setup on the PG side. At first, the position sensitivity will allow to measure momentum transfers in the vertical direction, a quality that will benefit mostly the single-crystal community with now access to full four-dimensional $S(Q,\omega)$ spectra. Secondly, the arrival times of the neutrons from different vertical sections can be measured separately. To increase the solid angle the height of the analysers will reach about 600 mm and hence it will be necessary to implement two separate detector benches. For the expected intensity of this new high-resolution capability an estimate can be provided. The measured intensity from the Mica(006) reflection on IRIS is about an order of magnitude less than the intensity with the PG analyser. On OSIRIS, the height of the analyser will be about a factor 3 larger compared to the Mica installation on IRIS, the silicon crystals will be a factor 3-7 thicker and hence through elastic bending more
neutrons are reflected. The flux on OSIRIS is about a factor 1.5 – 2 larger at \( \lambda = 6 \text{ Å} \), therefore we expect a measured intensity on OSIRIS with the silicon analyser similar or even higher than IRIS provides with the PG analyser.

Beyond the increase of dynamic range, the need and opportunity for more intensity has been identified. The OSIRIS spectrometer utilizes a non-ballistic curved \( m = 2 \) supermirror guide with a straight focusing end and the older IRIS uses a Ni-coated guide. Just from the accepted phase space, one would expect a factor 3-4 larger intensity transported to the sample position on OSIRIS compared to IRIS. However, all measurements indicate only a gain factor of 1.5 – 2 at \( \lambda \approx 6 \text{ Å} \). Since some time now it is known that superior guide geometries exist, which provide a high transmittance of cold neutrons over long distances [22, 23, 24]. Monte Carlo simulations are now planned to determine the exact guide geometry along a previous study [25]. A further direction of development is providing a platform for high-throughput QENS research. IRIS would serve as a first port of call for new research groups and this high-throughput approach to QENS research would pave the way for spatially and temporally resolved maps of the dynamical response of complex materials as a function of temperature or other external stimuli.

3. Conclusions
We have presented the status of the near-backscattering spectrometers IRIS and OSIRIS at the ISIS Facility. Both are highly productive instruments and provide cold neutron spectroscopic capabilities for quasielastic and low-energy inelastic problems. In a changing neutron scattering landscape the proposed new capabilities will ensure that the instruments remain competitive over the coming decade and beyond.

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