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Kookaburra: the ultra-small-angle neutron scattering instrument at OPAL[1]

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The new double crystal ultra-small-angle neutron scattering instrument Kookaburra, currently under construction at the ANSTO OPAL reactor, will allow characterization of microstructures covering length scales in the range of 0.1 to 10 µm. Using the 002 and 004 reflections of a doubly curved mosaic highly oriented pyrolytic graphite premonochromator crystal at a fixed Bragg angle of 45° in conjunction with two pairs of Si(111) and Si(311) quintuple-reflection channel-cut crystals will allow operation of the instrument at two different wavelengths, thus optimally accommodating weakly and strongly scattering samples in one sample position. The versatility, the estimated neutron fluxes and the low background noise of Kookaburra suggest that this state-of-the-art instrument will have a major impact in the field of large-scale structure determination.

1. Introduction

An ultra-small-angle neutron scattering (USANS) instrument, named Kookaburra, is currently being installed at the cold-neutron guide CG3 at the ANSTO OPAL reactor. This instrument will extend the range of experimentally measurable length scales currently accessible through the already existing small-angle neutron scattering (SANS) instrument Quokka (Gilbert *et al.*, 2006) by two orders of magnitude into the micrometre regime. Thus, multilevel structures in solids and liquids, containing not only nanometre-sized molecules and particles but also micrometre-sized aggregates and agglomerates, can be probed *via* neutron scattering. USANS is useful for studies of pores and cracks in rocks, cement or engineering materials, very large biological or polymer molecules or macromolecular assemblies, and mesoscopic magnetic particles.

The Kookaburra USANS instrument is based on the classical Bonse–Hart method (Bonse & Hart, 1965), which consists of using two multiple-reflection crystal systems arranged in a nondispersive geometry to achieve a steep decrease in the tails of the perfect crystal diffraction curves. This technique permits the detection of very small angular deviations of the neutron beam after scattering from a sample placed between two channel-cut crystals (Agamalian *et al.*, 1997).

2. Instrument concept

The central parts of a Bonse–Hart USANS instrument are two identical multi-bounce channel-cut perfect single crystals (Schwahn *et al.*, 1985), labelled monochromator and analyser, mounted on a single optical bench. When the monochromator and analyser crystals are aligned (analyser rotation angle θ = 0°), the incident beam is totally reflected into the main detector. During the experiment, neutron scattering intensities are measured as a function of the momentum transfer Q (or scattering angle θ). In fact, Q is determined by rotation of the analyser crystal by an angle θ, at which data are collected at one value of Q (or θ) at a time, *i.e.* only the small-angle scattering corresponding to the same angle θ is reflected. Minute deviations in the neutron beam direction after scattering by the sample are observed as a broadening of the double-crystal-analyser rocking curve.

Because the channel-cut crystals are perfect, Bragg reflection of a particular wavelength λ is restricted to a very narrow angular range of neutron trajectories that fall within the so-called Darwin width, which is typically a few arcseconds (Agamalian *et al.*, 1997; Barker *et al.*, 2005). Within this uncertainty, neutron trajectories are completely correlated with neutron wavelength. The Bonse–Hart USANS technique can use a relatively wide range of wavelength/divergence-angle combinations simultaneously, which enhances the flux on the sample without sacrificing the Q resolution.

The high angular resolution required for USANS experiments is enhanced through multiple reflections of the neutron beam before and after the sample. Kookaburra utilizes quintuple bounces inside perfect silicon channel-cut crystals. Optimization of the instrument with regards to determining the most efficient type of premonochromator crystal, channel-cut crystals and scattering geometry has been discussed by Freund & Rehm (2011).

Note that in a traditional SANS experiment the optimum sample scatters between 5 and 20% of the incident neutron

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Table 1
Kookaburra instrument performances expected for operation at two different wavelengths.

Note that the precise values for the full width at half-height depend on whether the Darwin or the Ewald solution applies, see *e.g.* Agamalian *et al.* (2010).

	Weakly scattering samples	Strongly scattering samples
Wavelength λ (Å)	4.74	2.37
Premonochromator	HOPG (002) at $\theta_{\rm Bragg} = 45°$	HOPG (004) at $\theta_{\rm Bragg} = 45°$
Beryllium filter	In	Out
Channel-cut crystals	Si(111) at $\theta_{\rm Bragg} = 49.2°$	Si(311) at $\theta_{\rm Bragg} = 46.4°$
Full Darwin width $2\Delta\theta_{\rm D}$ (µrad)	21	5.4
Minimum momentum transfer, $Q_{\rm min}$ (Å)	2.8×10^{-5}	1.4×10^{-5}
Flux on sample (n cm^{-2} s^{-1})	190 000	23 000

beam. For the case of less than 5% scattering, the signal-to-noise ratio might be poor (note that the lower limit depends somewhat on the shape of scattering curve). For scattering of more than 20% of the beam, the SANS intensity would be strong enough to create noticeable multiple scattering, resulting in a shape-distorted scattering curve. For the strongly scattering samples, an adequate sample optimization (*i.e.* a reduction of the scattering) can often be achieved by manipulating the sample *via*, for example, dilution, reduction in sample thickness or contrast matching. Note that the scattering power of a sample is also proportional to λ^2.

In order to optimally accommodate samples which differ in scattering powers, the versatile Kookaburra USANS instrument is designed to allow individual operation at two different wavelengths. The incident neutron beams are reflected off a highly oriented pyrolytic graphite (HOPG) premonochromator crystal at a fixed angle of $\theta_{\rm Bragg}$ = 45°, either at λ = 4.74 Å using the HOPG 002 first-order reflection or at λ = 2.37 Å using the HOPG 004 second-order reflection. The expected performances of Kookaburra when operating at either wavelength are summarized in Table 1.

When operating the instrument with 4.74 Å neutrons, two Si(111) channel-cut crystals will be used at a Bragg angle of 49.2°. Also, a beryllium filter will be moved into the beam to remove by scattering neutrons with wavelengths below 4 Å, *i.e.* to suppress higher-order wavelengths emerging from the premonochromator. When operating the instrument with 2.37 Å neutrons, two Si(311) channel-cut crystals will be used at a Bragg angle of 46.4° and the beryllium filter will be moved out of the beam. The 4.74 Å wavelength high-intensity operation mode is most appropriate for weakly scattering samples, but it should be noted that the four times larger full Darwin width for the 4.74 Å operation of $2\Delta\theta_{\rm D}$ = 21 µrad (compared to a value of $2\Delta\theta_{\rm D}$ = 5.4 µrad for the 2.37 Å operation) results in a poorer resolution at small values of Q ($Q_{\rm min}$ for 4.74 Å is a factor of two larger than $Q_{\rm min}$ for 2.37 Å, see Table 1). Note that $Q_{\rm min}$ is estimated as $Q_{\rm min} \simeq 4\pi\Delta\theta_{\rm D}/\lambda$ (Agamalian, 2011). Concerns about multiple scattering for strongly scattering samples can be addressed by switching to the λ = 2.37 Å mode when necessary. Although both the first- and second-order neutrons will reach the channel-cut monochromator crystal when the beryllium filter is moved out of the beam when operating Kookaburra at 2.37 Å, the 4.74 Å neutrons will not be further transported through the Si(311) channel-cut crystal setup.

3. Instrument layout

The general assembly of the Kookaburra USANS instrument is shown in Fig. 1.

Kookaburra is located upstream of the Platypus neutron reflectometer (James *et al.*, 2006, 2011) at the cold-neutron guide CG3. A doubly focusing HOPG premonochromator device will extract a monochromatic neutron beam. A beryllium filter can be used to remove by scattering neutrons with wavelengths below 4 Å, if required. A sample shutter allows opening and closing of the neutron beam. The sample position is surrounded by the channel-cut monochromator and analyser stages. The distance between these two stages is variable in order to be able to accommodate any sample environment equipment. It follows that the location of the main detector (which needs to be in line with the channel-cut analyser crystal) is variable, too. For detailed descriptions of the main instrument components see sections below.

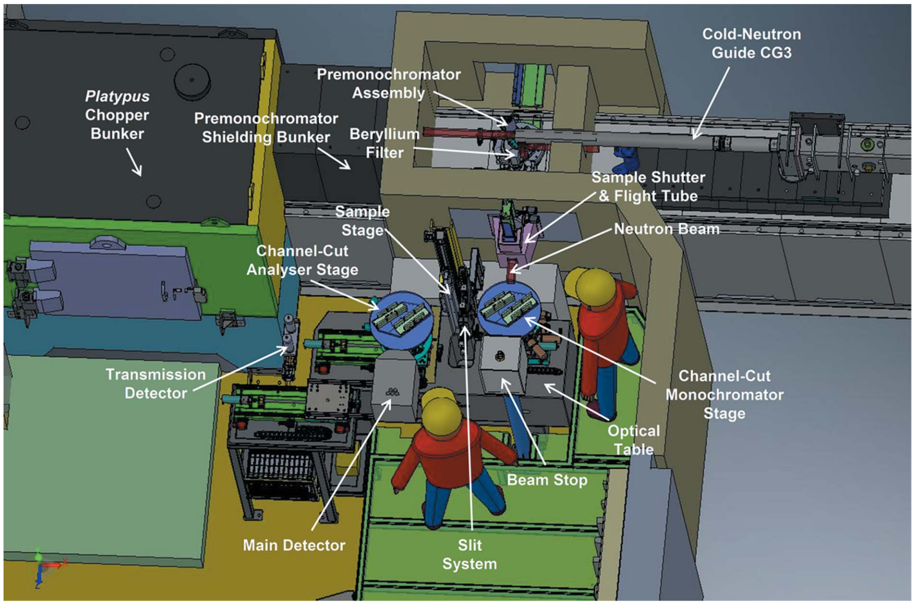

Figure 1
General assembly of the Kookaburra USANS instrument at the ANSTO OPAL cold-neutron guide CG3 with main components indicated.

4. Instrument components

4.1. Cold-neutron guide CG3

Kookaburra utilizes neutrons delivered by the OPAL cold-neutron guide CG3. This guide is fed by a hydrogen cold-neutron source 363 mm high × 108 mm wide with a total flux of 1.8×10^{13} n cm^{-2} s^{-1} at T = 25 K (Kennedy, 2006). CG3 includes straight neutron guide sections with curved neutron guide sections in between (with a radius of curvature of 1.3 km), supermirror coatings of m = 3 at the top and bottom of the neutron guides and m = 2.5 on the sides, a cut-off wavelength of about λ = 0.8 Å, and a beam cross section 200 mm high × 50 mm wide. The upper 20 mm of CG3 are exclusively used to provide neutrons to the Platypus neutron reflectometer installed downstream of Kookaburra. Therefore, the remaining effective beam cross section for Kookaburra is 170 mm high × 50 mm wide.

4.2. Premonochromator assembly

About 30 m away from the cold source, a premonochromator crystal will be installed in the white neutron beam exiting the cold-neutron guide CG3 of effective dimensions 170 mm high × 50 mm wide. Its function is to select a monochromatic neutron beam and to focus it onto the sample position, which is located between the channel-cut monochromator and the channel-cut analyser crystal at a nominal distance of 1600 mm from the premonochromator.

The premonochromator crystal material is highly oriented pyrolytic graphite with dimensions 82.5 mm (horizontal) × 170 mm (vertical) × 1.8 mm thick. Since the premonochromator is doubly focusing (*i.e.* it is curved both horizontally and vertically), it consists of a composite of five vertical arrays of 17 HOPG pieces, each piece with dimensions 16.5 mm (horizontal) × 10 mm (vertical). The pieces were mounted on a silicon support plate (SESO, http://www.seso.com) approaching the nominal curvatures with a sagittal (vertical) radius of curvature of 2.27 m and a meridional (horizontal) radius of curvature of 4.54 m, using indium metal as bonding material. The Bragg angle of the assembled premonochromator crystal is fixed at 45°.

The crystal assembly was characterized on the double-crystal test instrument T13C of the Institute Laue–Langevin (ILL) in Grenoble, France (Boeuf *et al.*, 1975), using the HOPG 004 reflection at a Bragg angle of 45° with a corresponding wavelength of 2.37 Å. The neutron mosaic spread was measured to be ∼0.6° at FWHM in both the horizontal and vertical directions, while the average peak reflectivities were determined as 44.7 ± 5.5% in the sagittal and 44.0 ± 2.6% in the meridional directions, respectively. These figures show that the HOPG material is of excellent quality and that the mosaic spread is isotropic.

The use of such a doubly focusing premonochromator leads to significantly enhanced neutron intensity at the sample position (Freund & Rehm, 2011). The premonochromator crystal is mounted on tilt and rotation stages to allow position control (ADC: Advanced Design Consulting, USA, http://www.adc9001.com).

4.3. Beryllium filter

Since Kookaburra will enable individual operation at either λ = 4.74 or 2.37 Å, a beryllium filter is required to absorb higher-order wavelengths when operating the instrument at λ = 4.74 Å. The beryllium filter will be moved out of the neutron beam when Kookaburra operates at λ = 2.37 Å *via* a linear stage (Huber, http://www.xhuber.com).

The beryllium (Be) material is made of the highly pure and isotropic S-200-F standard grade, which contains very low amounts of hydrogen (Materion, http://www.materion.com; formerly Brush Wellman company). One block of polycrystalline beryllium (where the average grain size is 8.6 µm and theoretical density is 99.99%) with dimensions 60 mm (horizontal) × 170 mm (vertical) × 150 mm thick along the neutron path is surrounded on four sides by 2.3 mm-thick BORTEC (Al-B_4C metal matrix composite with 16% B_4C) sheets plus an additional outer layer of 0.5 mm-thick cadmium. Placed inside a vacuum shroud made from aluminium (Janis, http://www.janis.com), the Be/BORTEC/Cd assembly is mounted onto a 10 mm-thick copper frame. Since cooling of the filter has a large effect on the transmission of the Be (the transmission of 4 Å neutrons through Be is increased by 125 ± 5% on cooling from 300 to 80 K), a cooling system (closed cycle refrigerator) and an associated vacuum system are also required.

4.4. Sample shutter and beam stop

A sample shutter (or beam shutter) allows the opening and closing of the neutron beam. The premonochromator and the beryllium filter are mounted inside a premonochromator shielding bunker. This bunker features 200 mm-thick lead walls lined with 5 mm-thick borated rubber in order to achieve radiological doses outside the bunker below the design limit of ≤3 µSv h^{-1}. The sample shutter is mounted on the outside of the bunker.

A re-entrant, fixed beam stop will stop the diffracted beam past the channel-cut monochromator crystal. Its opening is 155 mm (horizontal) × 275 mm (vertical) with a depth of 200 mm. The lead thickness is 50 mm with its inside lined with 5 mm-thick borated rubber.

4.5. Channel-cut monochromator/analyser stages

All four channel-cut crystals to be used at Kookaburra, *i.e.* two Si(111) channel-cut crystals and two Si(311) channel-cut crystals (manufactured by Holm, http://www.holm-silicon.de), were satisfactorily characterized on the neutron optics instrument S18 at the ILL (Kroupa *et al.*, 2000), which was configured as a high-resolution Bonse–Hart camera using the Si(333) reflection at a Bragg angle of 45° with a corresponding wavelength of 1.48 Å.

At the Kookaburra channel-cut monochromator (CC-M) stage (before the sample position) both an Si(111) and an Si(311) channel-cut monochromator crystal will be mounted on a base plate back-to-back with a sidewise offset, so that the beam centre is aligned on first reflection for both crystals with respect to the premonochromator crystal. The Bragg angle for the Si(111) crystal is 49.2° (for λ = 4.74 Å), while the Bragg angle for the Si(311) crystal is 46.4° (for λ = 2.37 Å). Therefore, the respective neutron beam paths between monochromator and analyser crystals will differ by a few degrees. The schematic layout of the channel-cut crystals is shown in Fig. 2. Fig. 2(*a*) depicts the setup used when operating Kookaburra at λ = 4.74 Å. The neutron beam (shown in red) enters and leaves the Si(111) monochromator crystal (shown in green) at 49.2° relative to the beam coming off the premonochromator, before entering and leaving the Si(111) analyser crystal (also shown in green). The paths of the neutron beams transmitted towards the transmission detector and beam stop, respectively, are indicated in blue. The minimum distance between the channel-cut monochromator CC-M and the channel-cut analyser CC-A of 600 mm can be variably increased to up to 1100 mm for accommodation of any sample environment equipment (note that in Fig. 2 the position of the channel-cut analyser stage for the maximum distance of 1100 mm is labelled CC-A′). Fig. 2(*b*) shows the setup used when operating Kookaburra at λ = 2.37 Å. The Si(311) crystals (shown in pink) are rotated into the neutron beam at an angle of 46.4° relative to the beam coming off the premonochromator.

The channel-cut monochromator and analyser crystals will be housed in two separate, shielded monochromator/analyser boxes made from aluminium, lined with boron plastic, and furnished with beam entrance and exit cut-outs. Alignment of the channel-cut monochromator crystal in use will be done *via* tilt, rotation and linear stages (ADC) beneath the base plate. When swapping between channel-cut monochromator crystals, *i.e.* between the two wavelengths, both shielded boxes will be rotated by 180°. The same arrangement as described above for the channel-cut monochromator stage is valid for the channel-cut analyser stage located after the sample position. Both channel-cut monochromator and analyser stages are mounted on a single optical table made from granite (ADC). The total weight of the granite table is about 1.3 tons; this will reduce the impact of external vibrations on the relative angle of the monochromator and analyser channel-cut crystals, which determines the angular resolution of the instrument.

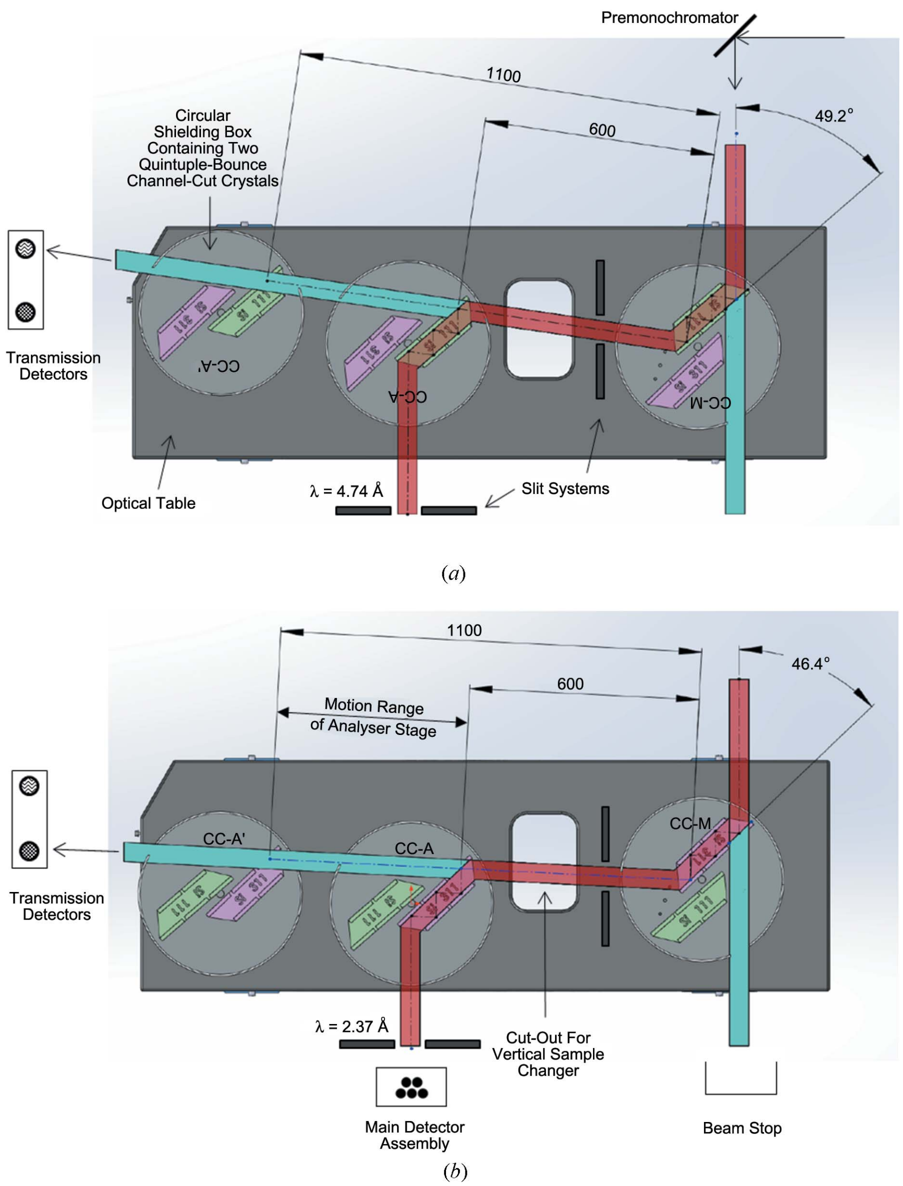

Figure 2
Schematic layout of two sets of channel-cut crystals mounted inside circular shielding boxes on top of the optical table. CC-M: channel-cut monochromator; CC-A: channel-cut analyser positioned 600 mm away from CC-M; CC-A′: channel-cut analyser positioned 1100 mm away from CC-M. Using (*a*) Si(111) at 49.2° and (*b*) Si(311) at 46.4°. For details see text.

4.6. Slit system

Two slit systems (ADC) will be used to define the dimensions of the neutron beam before and after the sample. The first slit system is placed between the channel-cut monochromator stage and the sample stage. Its opening will be 0–115 mm in horizontal direction and 0–70 mm in vertical direction. The wider opening in the horizontal direction takes into account the two different Bragg angles of the channel-cut crystals, or two different neutron beam paths, required when operating the instrument at two different wavelengths (see Fig. 2). A second slit system is placed between the channel-cut analyser stage and the main detector. Its opening is 0–70 mm in both the horizontal and vertical directions. The openings of both slit systems will be adjustable through four separate blades in the up/down/left/right directions, each controlled by motors and encoders. The material of the slit blades is sintered B_4C of 5 mm thickness.

During operation, the maximum opening of the slit systems corresponds to the maximum beam size transmitted through the channel-cut monochromator and the channel-cut analyser crystal of 50 × 50 mm. This value also determines the maximum sample size.

4.7. Sample stage

For routine experiments, a multi-position sample changer will be used to successively position various samples in the neutron beam centre using a linear translation stage. To this end the distance between channel-cut monochromator and analyser stage will be at its minimum of 600 mm (see Fig. 2). For experiments requiring any sample environment the distance between the two stages can be increased variably up to a maximum of 1100 mm by moving the channel-cut analyser stage away from the sample position. In the latter case the beam intensity will be reduced by a factor of 0.7 from the effect of defocusing. The sample changer as well as any sample environment equipment will be independently supported by an appropriate table, which must not be in contact with the optical table, in order not to transfer vibrations to the highly sensitive arrangement of the channel-cut crystals.

4.8. Detector system and data treatment/management

The detector system consists of a beam monitor required to record the intensity of the incident neutron beam, transmission detectors required to monitor the intensity of the neutron beam transmitted through the sample and a main detector required to measure the intensity of the neutron beam transmitted past the channel-cut analyser crystal.

The beam monitor is installed immediately behind the sample shutter as a neutron counter (ORDELA, http://www.ordela.com). It operates at low ^{3}He pressure with a neutron detection efficiency of ~5 × 10^{-6} at 15 meV (2.3 Å). Both transmission and main detectors consist of ^{3}He filled, 5 inch (1 inch = 25.4 mm) long position-sensitive detector tubes (GE, http://www.ge-energy.com). The operation of Kookaburra at two different wavelengths requires a set of two individual detectors to measure the transmitted beam (with efficiencies of ~1% for λ = 4.74 Å and ~10% for λ = 2.37 Å). The two transmission detector tubes (diameter = 2.5 inch) are mounted next to each other on a shielded support structure. The main detector consists of a hexagonal array of five ^{3}He detector tubes (diameter = 1 inch) with two tubes in the front and three tubes at the back inside a shielding box made from Cd-lined borated polyethylene to protect the detector tubes from stray neutrons.

Given that the distance between channel-cut monochromator and channel-cut analyser stages is variable, the main detector shielding box is mounted on a linear stage (ADC) which follows the position of the channel-cut analyser crystal (see Fig. 1). Besides the main detector shielding box and the linear stage, the detector support structure also holds the second slit system as well as a beam attenuator kit comprising plexiglass plates of different thicknesses on a small linear stage (HIWIN, http://www.hiwin.com).

Instrument control, data acquisition, experimental planning and data analysis of the Kookaburra USANS instrument is carried out by a computerized system consisting of the following: (i) a data acquisition computer directly interfaced with the USANS detector electronics complete with histogram memory software, (ii) *SICS* (SINQ Instrument Control System) computer server software (Herr *et al.*, 1997), (iii) *GumTree* user application software (Lam *et al.*, 2006). Reduced USANS data will then be analysed using model-independent methods or nonlinear fitting to one of a large and growing catalogue of structural models which has been developed at the NIST Center for Neutron Research (Kline, 2006).

5. Summary and outlook

A new Bonse–Hart type ultra-small-angle neutron scattering (USANS) instrument, named Kookaburra, is currently under construction at the ANSTO OPAL reactor. Kookaburra will extend the range of experimentally measurable length scales currently accessible through the already existing SANS instrument Quokka by two orders of magnitude into the micrometre regime. Therefore, the combined USANS/SANS utilization at ANSTO will allow the characterization of microstructure over four orders of magnitude in size (1 nm to 10 µm). Kookaburra utilizes an HOPG premonochromator in conjunction with two pairs of Si(111) and Si(311) quintuple-reflection channel-cut crystals, which will allow operation of the instrument at two different wavelengths and thus optimally accommodate weakly and strongly scattering samples in one sample position. It is expected that design, construction and commissioning of Kookaburra will be completed late in 2013, followed by a transition to first user experiments.

We wish to acknowledge the ANSTO Bragg Institute's mechanical, electrical, technical, data acquisition, electronics and computing teams for their invaluable contributions to the project, and the Kookaburra USANS Instrument Advisory Team with Duncan McGillivray as spokesperson. The mention of commercial products does not imply endorsement by ANSTO, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose.

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