PA20 : A new SANS and GISANS project for soft matter, materials and magnetism

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Abstract. This article presents the new Small Angle Neutron Scattering (SANS) instrument PA20 which will replace the PAXE instrument at LLB-Orphée. SANS is well-known to be especially well adapted to research in soft matter, materials and nanosciences and SANS is particularly powerful in the studies of complex systems, with isotopic labeling and contrast variation method, but also for large-scale structures (magnetic or not). PA20 is part of the LLB instrumental upgrade program "CAP2015". PA20 will not only maintain LLB’s capabilities in SANS, but also considerably extend them in terms of SANS for magnetism with a polarized neutron option and Grazing Incidence SANS (GISANS), with an improved dynamical Q-range. The total length of PA20 will be 40 m, including a 19 m collimation length, a 20 m detector tank containing high-resolution/high-efficiency \(XY\) detectors, and a casemate containing a monochromator (velocity selector \(\lambda = 0.3 – 2 \text{ nm}\)), a chopper system for Time-of-Flight (TOF) mode, a polarizer and an RF spin flipper. PA20 will allow faster measurements, with "single-shot" access to a wider range of scattering vectors, on possibly small samples (few mm in size). In addition, polarized neutrons will enable magnetic studies in both SANS and GISANS configurations. Studies of nanostructured surfaces and interfaces (deposited or embedded nano-objects), magnetic domain formation, multilayered materials or magnetic thin films through specular and off-specular signals will be possible through GISANS setups. The versatility of PA20 should contribute to both enlarge the neutron user community, especially in expanding areas like nanosciences, and offer improved services for users in the years to come.

1. Motivations and Objectives

At the heart of the LLB instrumental upgrade program CAP2015 lies the need to improve and expand the instrumental suite to meet users demands in the next 10-15 years. The LLB is equipped with 4 SANS instruments (PAXE, PAXY, PACE and TPA [1]) and 2 reflectometers (PRISM and EROS) [2]. This instrumental suite covers small and very small neutron scattering, polarized neutron reflectometry and liquid reflectometry. As SANS instruments represent over a third of the total beam time requests at LLB, it is imperative to maintain top-level instruments in this area. On the other hand, increased interests in nanostructured materials with surface or interface issues [3, 4, 5] have brought about the need for LLB to offer a complete new GISANS instrument. In this context, a new Small Angle Neutron Scattering (SANS) and Grazing Incidence Small Angle Neutron Scattering (GISANS) instrument, PA20, is under construction in place of the PAXE spectrometer located at the end of the G5 neutron guide. PA20 aims
at providing a better service to users, with more versatility, and at answering the needs of the expanding nanosciences community. The length of the instrument, approximately 40 meters, will make it possible to exploit both the flux and the resolution. PA20 will cover a broad dynamical range, from very small scattering vectors, lower than $0.01 \text{ nm}^{-1}$ (at $\lambda = 8 \text{ Å}$ and 19 m) with reasonable flux, up to $7 \text{ nm}^{-1}$ (at $\lambda = 4 \text{ Å}$ and 1.0 m). The increased performances (flux, resolution and Q-range, versatility, polarized neutrons) will also broaden the span of experiments that can be performed with SANS and GISANS experiments. The global progress compared to existing SANS at LLB will be particularly appreciable to study the objects from a few nanometers up to several hundreds nanometers [24].

1.1. SANS

SANS is a particularly relevant technique to investigate inhomogeneities of size in the range between 1 and 500 nm [6, 7], presenting a neutron scattering contrast being structural, chemical or magnetic in origin. Information like density or size distributions, shape or interface effects, and magnetic structurations [8, 9] can be addressed using SANS instruments. The fact that several SANS instruments are currently under construction in Europe highlights the importance and relevance of SANS studies in these wide range of topics [10, 11, 12]. This is especially true for soft matter (polymers, polyelectrolytes, micelles, vesicles, biological systems like proteins) [13, 14, 15, 16], materials science (chemical demixing, precipitates, porous materials, alloys of industrial interest) [17] and magnetic materials (critical magnetic fluctuations near transitions, magnetic particles of ferrofluids [18], vortices in superconductors, nanomagnets, long-range helicoidal orders).

SANS is without doubt one of the main experimental technique for soft matter and, to a lesser extent, biosystems. Its success originates obviously from H/D isotopic substitution of hydrogendeous materials allowing labeling and contrast matching experiments but also to the capacities of neutron scattering to probe form and structure factors of large objects. Increased performances in terms of flux and efficiencies will help to investigate small volume samples like thin polymer films, confined systems, grafted objects, finely tuned molecular assemblies where isotopic labeling is possible on small fraction of the materials. This is particularly relevant to biological systems [25, 26, 27] which, until now, are mostly studied with X-ray diffraction, but do not profit to the powerful insight offered by contrast variation methods. Other examples are the studies of materials based on nano-particles inserted in polymeric matrices [28, 29, 30], which could be done at lower concentration of particles, and rheological studies of complex fluids under strain, which could be done in-situ with greater ease. The polarized neutron mode is also of prime interest in the case of magnetic nanostructured materials like inserted magnetic objects [31]. Polarization will also be extremely helpful to eliminate the incoherent background of hydrogendeous systems, from small molecules to proteins.

PA20 will improve SANS experiments in a variety of ways in terms of neutron flux, scattering angle coverage, Q-resolution and neutron polarization. For instance, faster data acquisition rates enables kinetics measurements or systematic studies as a function of temperature near phase transitions where better resolution are critical to differentiate between models or theories. The increased Q-range coverage allowed by the 2D front and rear detectors will permit a more rigorous analysis of interface effects, their fractal aspects (Porod regime) compared to volume effects (Guinier regime). Pushing the limits in terms of flux and resolution is also critical when dealing with samples that are difficult to synthesize or which can be obtained only in modest quantity. The new instrument PA20 will make it possible to tackle studies of such samples.

Regarding magnetic studies, PA20 will be the first SANS with multi-wavelength neutron polarization at LLB, fulfilling the needs to study magnetization processes in nano-objects and thin films. Another range of topics which will benefit from SANS with polarized neutron option
relates to magnetic systems and correlated electron systems [32, 33]. The existence of a magnetic contrast in SANS and GISANS allows to study magnetic structures. Cluster formation in spin glasses, long-range helicoidal orders, vortices in superconductors [34], phase segregation in CMR materials like manganites [35] have been widely and successfully investigated with SANS. This technique has provided decisive contributions because the response function can be measured across a broad range of scattering vectors (10$^{-2}$ – 5 nm$^{-1}$). Weak Bragg peaks, even with some disorder, are accessible providing sufficient neutron flux. This also applies to study the role played by long-range modulations (several tens of nm) in multiferroic materials. With PA20, it will also be possible to study thin magnetic films in both SANS and GISANS modes to address questions related to surfaces and interfaces effects, alteration of the properties when going from bulk to layers, etc. The prospect of having a dedicated upgraded SANS instrument to magnetic studies at the nanometer scale opens the way to more detailed approaches.

Finally, PA20 will be especially helpful in materials sciences [17, 36] as SANS proved to be an indispensable tool to study microstructural heterogeneities on a nanometric scale, in particular precipitation or segregation in metal alloys. In comparison with the usual techniques used in metallurgy (TEM, SAXS), SANS provides quantitative data at the sub-micron scale on large volumes (several mm$^3$). This is a key asset for industrial materials which present heterogeneities or multiples phases. Irradiated materials, exposed to high-energy neutrons and/or elevated temperatures over very long periods of time, often present microstructural changes like swelling, cavities, hardening, etc. SANS can address these issues and provides invaluable information to engineers and scientists in the nuclear industry.

1.2. GISANS

GISANS is less developed worldwide but, as a surface and interface probe, complements SANS and neutron reflectivity techniques [19] as well as Xrays GISAXS. The 2D detectors allow measurements of off-specular signal, characteristic of structural and magnetic surface inhomogeneities. PA20 will have the capacity to probe structural or magnetic inhomogeneities in the range of 1 to 500 nm on surfaces and buried interfaces along both orientations - vertical or horizontal. Combining high-precision slits collimations and high angular coverage by both rear and front detectors will permit off-specular scattering studies in the most versatile possible way. Particularly relevant issues are related to magnetic layered materials, magnetic domain formation, organic or polymeric layered films, etc. [20, 21, 22, 23]. If GISANS is known to be complementary to GISAXS X-Rays experiments, neutron sources cannot compete with modern synchrotrons in terms of flux. Therefore, GISANS within PA20 will focus essentially to the study of networks of magnetic nano-objects (wires, ribbons), magnetic correlations or domains formed at the surface or at interfaces. Important issues related to emergent materials such as thermoelectrics or multiferroics could be addressed.

2. Instrument description

This section describes the technical key components of PA20 whose general layout is shown in Figure 1.

2.1. Casemate: monochromator, chopper, polarizer, RF flipper

The 3 m long casemate, composed of lead and heavy concrete, will house a Dornier-type velocity selector that can be replaced by a chopper when PA20 is operating in Time of Flight (TOF) mode, an escapable polarizer placed in vacuum, and a RF spin flipper (see Figure 2). The multi-blade velocity selector (Astrium-EADS) has a transmission larger than 95% (for a theoretical
Figure 1. General layout of the PA20 spectrometer (in orange) in LLB-Orphée Guide Hall. From left to right: Casemate housing monochromator, polarizer and flipper; collimation line; sample area; detectors tank. The 20 m evacuated tube contains a $64\times64 \text{ cm}^2 \text{ } ^3\text{He}$ rear detector ($5 \text{ mm vertical and horizontal pixel size}$) which can be moved from 1 m to 19 m from the sample position and a front detector consisting of two sets of 10 mm diameter tubes placed perpendicular one to another. Each set will be 20 cm wide and 64 cm high. The front detectors can be located between 1 and 10 m from the sample.

non diverging neutron beam) and a wavelength distribution of about 10% .

Beam polarization will be ensured by a 1.4 m long cavity polarizer [43] with a tapering angle of 1.6 degrees and with a double-V configuration (with transmission $\approx 80\%$ ) [44, 37, 38, 39] using 0.9 m long silicon substrates, double-side coated with $m = 3.4$. A 450 Gauss magnetic field will be generated to ensure magnetization. This setup will provide a polarization ratio superior to 98 % from 4 Å upwards. A classical radio-frequency spin flipper located after the polarizer can reverse the polarization direction. PA20 will also offer the possibility to perform high Q-resolution experiments with high-Q dynamical range (typically $50 \times \lambda_{\text{max}}/\lambda_{\text{min}}$). This is best done using a chopper in TOF mode where wavelength spread could be limited to less than 1 %. This setup complements the velocity selector mode and will find applications in various areas where very high resolution is needed.

2.2. Collimation System

The collimation system has been designed to offer maximum flexibility in terms of collimated beam shapes and accuracy. The maximum collimation length, in polarized mode, will be 16 m from the exit point of the casemate to the sample position. In non polarized mode an additional 3 m of collimation is available with an extra slit system located at the entrance of the casemate. From the casemate exit point to the end-point of the collimation, there will be 7 collimation slits separating 5 super-mirror guide elements (Ni/Ti, $m = 2$) of $85 \times 25 \text{ mm}^2$ internal section. The available collimation lengths will therefore approximately be 19, 16, 12, 8, 4, 2.5 and 1 m. A free space is booked inside the collimator and over all its length for future implementation of modern optics such as new guide shapes or multi-beam mode [45, 47]. At the end of the collimator, an under vacuum variable-length extension (50-80 cm) will optimize neutron transmission to the sample.

A system of 4 independent slits, 2 horizontal and 2 vertical, has been chosen to provide square shaped collimation for SANS and vertical or horizontal rectangular shape for GISANS. These slits have a positioning accuracy of 50 µm and offer openings up to 80 mm along the vertical
Figure 2. Casemate plan (roof removed): Velocity selector monochromator (left) and polarizing setup including transmission polarizer and RF spin flipper (right).

direction and 30 mm along the horizontal direction. To increase the scattered intensity at small scattering vectors or to be able to get even smaller scattering vectors, two motorized systems of MgF$_2$ focusing lenses [40, 46, 41] can be inserted inside the collimator, just before the sample, with a 25 mm diameter, a 25 mm radius of curvature and a 1 mm thickness at the centre. The number of lenses for each device will be 19 and 37 respectively.

This option is well adapted to long wavelengths and large sample-to-detector distances, a situation where counting times are usually the longest. Neutron polarization after the casemate is maintained by a vertical 30 Gauss guiding magnetic field, generated by vertical columns of permanent cylindrical magnets (NbFeB, $\Omega = 25$ mm, 1 Tesla) and two iron horizontal plates situated below and above the neutron guides and separated by approximately 200 mm. At the very end of the collimation, right before the extension, the polarization is switched to horizontal to ease the use of polarized neutrons with horizontal magnetic fields at the sample position (2 Tesla electromagnets and 10 Tesla cryomagnets currently available at LLB). Both the extension of the collimator and the entrance part of the detectors tank will be non magnetic.

2.3. Detectors

In order to achieve high angular resolution and large angular coverage, two sets of 2D XY $^{3}$He detectors will be placed inside the detector tank (see Figure 1). The rear detector is a 64x64 cm$^2$ $^{3}$He single-bloc Aluminum multi-tube detector, with 5 mm vertical and horizontal pixel size and square section tubes, which can move 30 cm sideways from its central position to access larger scattering vectors still with a good pixel definition. Its expected efficiency of 85% at 5 Å and 15 bar pressure is among the best in the world. As high resolution at large scattering angle is not absolutely necessary, the front detector, also with $^{3}$He, with tube sections in the order of 10 mm, consists of two sets of tubes placed perpendicular one to another. Each set will be 20 cm wide and 64 cm high. The front detector will allow covering an extended Q-range and its distance...
to sample (between 1 and 10 m) will be adapted according to the position of the rear detector [42] (between 1 and 19 m). The simultaneous use of the detectors will allow the collection, in a single measurement, of the scattered intensity over a dynamic Q-range of $Q_{\text{max}}/Q_{\text{min}} \approx 50$ and could even be pushed further by shifting the rear detector and the vertical front detector sideways. These high-resolution and high-efficiency $XY$ detectors will greatly increase both the counting rate and the $Q$-range covered in a single measurement compared to our current SANS. A total gain factor of 15 is anticipated in a standard configuration, which will go at least to 50 with the focusing lenses in place (for $Q_{\text{min}} = 10^{-2}$ nm$^{-1}$, $\lambda = 8$ Å, $L = 12$ m detector distance).

3. Conclusion

The new Small Angle Neutron Scattering instrument, PA20, has been presented. PA20 will be located at the LLB-Orphée reactor in Saclay and will replace the actual spectrometer PAXE. The total length of PA20 will provide a 19 m collimation length and a 20 m detector tank. Among its specifications, PA20 will offer an extended dynamical $Q$-range of 50, and an intensity gain of 15 compared to PAXE, in monochromatic mode through the simultaneous use of high resolution and high efficiency rear and front 2D detectors. The collimator will offer the possibility to use either square or slit shape apertures for GISANS or specific SANS measurements, two sets of MgF$_2$ neutron lenses, and also leave room for future implementation of modern techniques in neutron optics such as new guide shapes or multibeam devices. PA20 will also enable the use of polarized neutrons with a polarization ratio larger than 98 % from 4 Å upwards. Finally, TOF option is envisaged in the future with a resolution close to 1 % and a dynamical scattering range of 500. Therefore, due to its design and versatility, PA20 will contribute to enlarge the neutron user community, in particular in nanosciences.

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5. References
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