Journal of Physics: Conference Series **528** (2014) 012027 doi:10.1088/1742-6596/528/1/012027

exTAS: a new concept in three axis spectroscopy for small samples

A Piovano, S Roux and J Kulda

Institut Laue-Langevin, 6 rue Jules Horowitz, 38042 Grenoble Cedex 9, France

piovano@ill.fr

Abstract. The progress in neutron delivery systems and in neutron focusing techniques has made possible neutron studies of excitations in sub-cm^3-sized single crystals, which are still much larger than crystal sizes needed for standard laboratory characterization techniques. In an effort to further reduce this gap we are proposing the *exTAS* project, which intends to stimulate a paradigm shift towards the use of mm^3-sized samples in neutron spectroscopy. The *exTAS* project aims to boost the TAS sensitivity limits by combining sharp mm-sized focal spots, minimizing penumbra effects in sample environment illumination, with a spectrometer layout downscaled to a tabletop size and enclosed in a shielding casemate. The reduced spectrometer dimensions will provide enhanced flexibility in adapting the momentum resolution to the problem being studied and will facilitate the sub-millimetre positioning accuracy of its components, matching the reduced focal spot size.

1. Introduction

For many materials of current interest single crystal samples cannot be grown large enough to reach present standards of 0.1 – 1 cm^3. Even if large single crystals can be grown, this may introduce significant delays before neutron spectroscopy experiments can be performed. Moreover in many materials the relevant parameters are ‘fine-tuned’ by doping, whose chances for homogeneity increase with decreasing sample volumes. It should be noted that all laboratory measurements, such as specific heat and susceptibility, are performed on well-characterized sub-millimeter sized samples. In this respect neutron results from large volumes or multi-crystal mounts can exhibit sample-dependent features and often remain disputed for a while.

Therefore it is of utmost importance to maximize the amount of neutrons, which can contribute to the measured signal, at the sample location. The intensity of the real-space focal spot can only be augmented by increasing the density of incident neutron flux and by optimizing its momentum distribution [1]. The first way, involving the neutron source brilliance, moderator efficiency, crystal reflectivity and detector efficiency is close to being exhausted by technological progress accumulated since the early days of the technique. The second way has received an increased attention in the last two decades and has lead to quite spectacular gains by a combined implementation of horizontal and vertical focusing optics in three-axis spectrometers (TAS) [2-4], which has made possible efficient studies of excitations in sub-cm^3-sized single crystals [5,6]. After completion of the *ThALES* project [7] (end of 2014) the portfolio of the ILL TAS instruments will represent the state of the art in this respect worldwide. Nevertheless, due to much larger extent of possible parameter combinations, this way still offers a scope for new developments and the purpose of the *exTAS* project is to explore them.

2. The *exTAS* concept

The angular resolution (beam divergence) of the present state-of-the-art TAS is determined by their primary spectrometer focusing distances (usually about 2 m) in combination with lateral dimensions of their monochromators (about 250 x 200 mm^2), resulting in divergence angles of 2 - 3º and in momentum resolution components of about 0.05 to 0.15 Å^{-1} in the thermal energy range. For many practical purposes this resolution still could be relaxed by a factor of 2 – 3 within the framework of a compromise solution permitting to investigate an otherwise inaccessible system, thanks to a gain in count-rate proportional to the square or cube of the linear resolution scale factor.

This gain can be even more impressive in the cases of cold neutron TAS, which use the same construction elements as the thermal ones and where the same beam divergences lead to even finer momentum resolution because of the shorter (2 – 4 times) neutron wave-vectors. With the present design of the cold and thermal neutron TAS the divergence increase into the range of 5 - 10º is impractical due to huge lateral sizes of the crystal devices imposed and due to the desperate background levels resulting from opening up the windows of primary and secondary spectrometers.

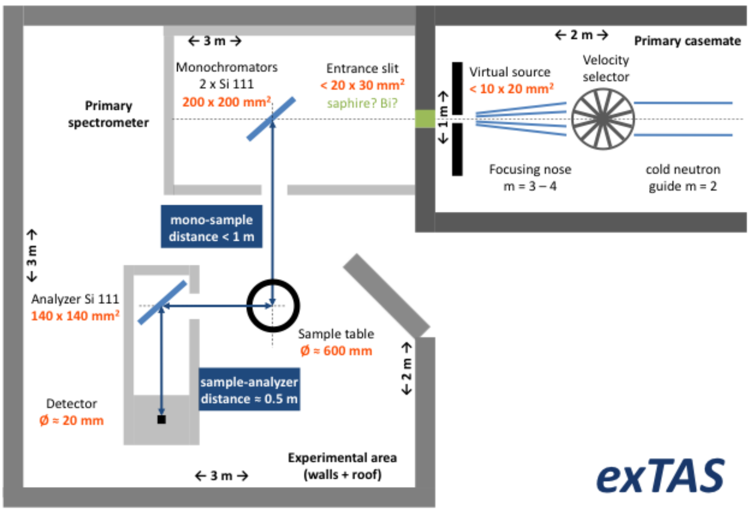

Figure 1: Schematic drawing of the *exTAS* set-up, including the most important dimensions and distances.

We believe, however, that such a step can be explored on a newly built and dedicated instrument, strictly following a number of assumptions and precautions:

- neutron beam from a cold guide, carefully filtered by a velocity selector, and possibly also by transmission filters, in order to minimize the need of shielding on the instrument parts;
- compact instrument design with minimum focusing distances (0.5 - 1 m), permitting to match standard optical devices with high beam divergence;
- sharply focusing optics (perfect bent Si, Ge, diamond crystals) to avoid smearing of focal spots and resolution volumes;
- general layout minimizing exposure of the analyzer/detector chain to parasitically scattered neutrons from the instrument neighbourhood as well as from its own beam.

The combined implementation of these principles should provide an increase in signal count rate of at least one order of magnitude, whenever a relaxed resolution could be sensibly used, and a largely improved signal-to-noise ratio in all cases.

3. Technical outline

The essential idea of *exTAS* is to provide an as intense sample illumination as possible while reducing to the strict minimum the intensity of parasitically scattered neutrons and of all other sources contributing to the signal background. These goals can be achieved by precise real- and reciprocal-space focusing combined with careful filtering of neutron beams and shielding of the whole instrument, implemented in a compact and highly flexible *exTAS* setup schematically represented in Figs. 1 and 2.

The primary casemate of *exTAS* will be best placed at an end position of a high-flux cold neutron guide, whose $m = 2$ super-mirror coating would allow for a significant sub-thermal (warmish) spectral component. A focusing nose will transform the large-area low-divergence beam of the guide into a high-flux beam spot of maximum section 10 x 20 mm^2 (w x h) at the virtual-source slit position with a divergence optimized to illuminate a 200 x 200 mm^2 active monochromator area over a distance of 1 meter. It is essential to filter the neutron beam by a velocity selector before it leaves the primary casemate to minimize radiation load on the monochromator and on its environment and to reduce background in the experimental area. In order to further reduce the neutron- and γ-background, the output window of the primary casemate may be filled with a crystal filter (Bi, sapphire). This will permit to keep the incoming neutron flux in the primary spectrometer close to the level of 1×10^{10} ns^{-1} so that its shielding would not need to contain lead or any other heavy element components.

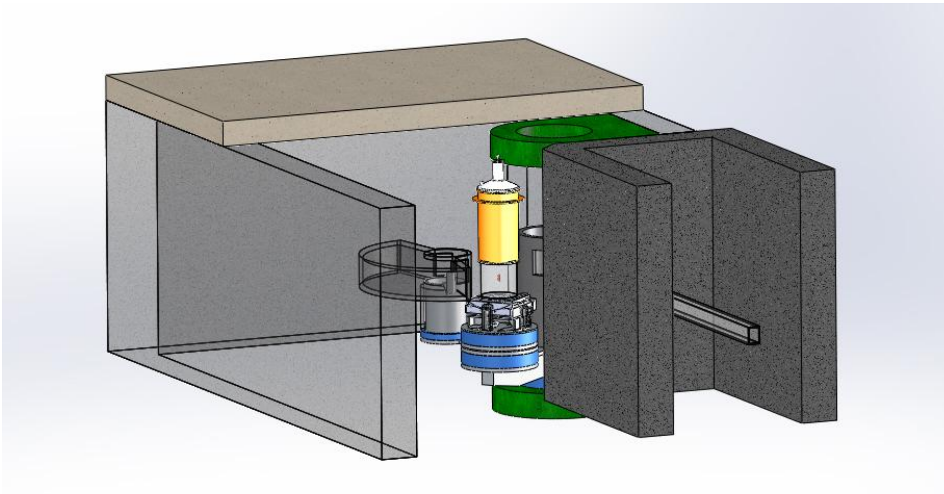

Figure 2: Prototype sketch of exTAS pointing out the compact instrument layout together with its most important shielding parts.

The monochromator will consist of two double-focusing faces (on a rotational changer), both using elastically bent Si 111 crystals but differing in the thickness of the crystal blade stacks, typically 1 – 2 and 10 – 15 mm, to permit quick switching between the low- and high-resolution modes. Its active area will remain about unchanged as compared to the design adopted for ILL TAS instruments, permitting to increase its solid angle acceptance by a factor of four thanks to the two times shorter focusing distance. A mobile slit system will provide a choice of an adequate momentum resolution by limiting its active area according to the needs of a given experiment.

The sample table will be kept as compact as possible but still designed to support equipment mass of up to 120 kg with maximum outer diameter of 600 mm to accommodate standard ILL sample environment equipment as well as the dedicated TAS *VacBox* (vacuum box). The whole secondary spectrometer can be sustained by means of robust arms connected to the primary spectrometer or move on air pads on a Tanzboden platform, elevated with respect to the experimental hall floor.

Thanks to the more compact layout and reduced weight a support plane quality closer to optical benches will be achieved, making possible sub-millimeter positioning accuracy.

The secondary spectrometer in its baseline version is an analogy of a classic single channel analyzer-detector system, employing a double-face silicon analyzer of the same design as the monochromator. Nevertheless sample-analyzer and analyzer-detector distances are reduced to about 50 cm to draw benefits from an increased solid angle. Precise focusing in combination with small sample sizes will permit to minimize the detector size so that its active volume can be kept below 20 x 50 mm^2 (Ø x h), reducing the requirements on the shielding volume of the secondary spectrometer.

The whole experimental area, hosting the primary and secondary spectrometers, will be enclosed in a shielding hutch (walls and roof) built of standard materials used for this purpose at the ILL (light concrete, Pb, B_4C) and minimizing the interaction with neighboring experimental areas in terms of radiation background. The access to experimental zone would be from

- ground level for maintenance and experimental equipment installation,
- top level (roof) for inspection, sample changing, He and N_2 filling.

The top access, through an opening just over the sample table area will further reduce chances of γ-exposure in the presence of lighter monochromator shielding.

4. Ray-tracing simulations

A complete ray-tracing optimization of a new instrument is a vast and complex task, having to take into account correlations of many design parameters. Here we present some preliminary results, obtained with the SimRes code [8], concerning the two optical elements playing a key role in the performance of the proposed instrument: the focusing supermirror nose, providing interface between the neutron guide and the virtual source, and the doubly focusing silicon monochromator, projecting a monochromatic image of the virtual source on the sample.

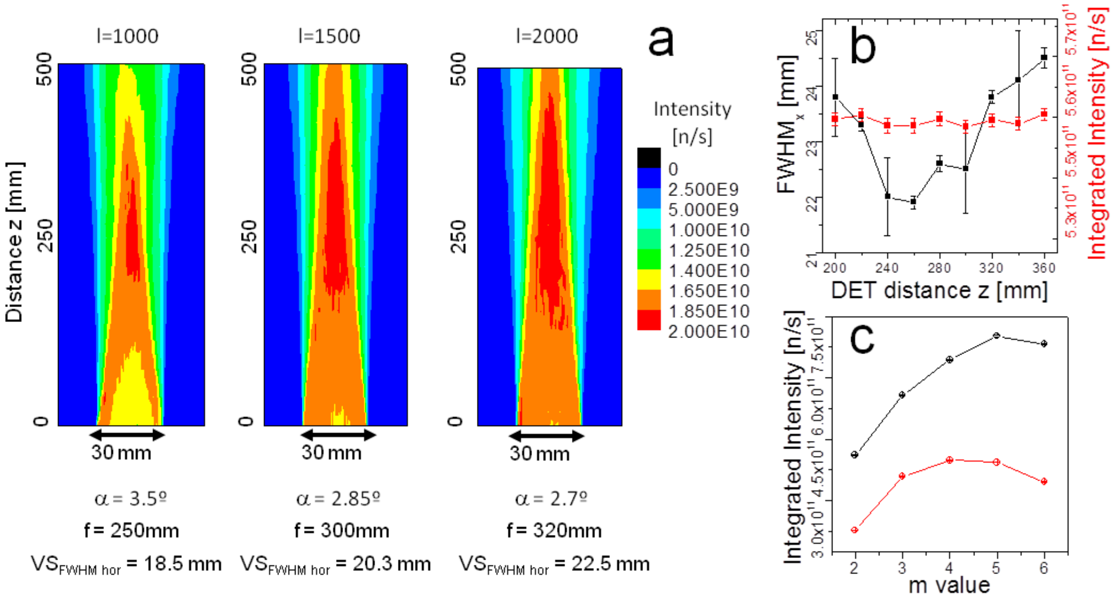

Figure 3: (a) 2D neutron intensity maps in horizontal plane in function of distance *z* from the exit of an *m* = 3 tapered parabolic focusing guides of different lengths *l* with no collimating blades inside; *α* is the calculated divergence, *f* the focusing distance and VS the horizontal width (FWHM) of the focal spot; (b) variation of the focal spot width and of the integrated intensity with distance *z* from the guide exit; (c) integrated intensity in function of *m* value for a guide with *l* = 1000 mm and no collimating blades (black) and *l* = 1000 m and 8 x 8 double side coated collimating blades (red).

The neutron guide system providing neutrons for *exTAS* is catered by the ILL horizontal cold source (cf. the *ThALES* project [4]) and has the following configuration:

- a cold neutron guide 90 m long with a cross-section of 60 x 120 mm^2 (width x height) with $m = 2$ and curvature of 4 km;
- a horizontally and vertically tapered parabolic focusing guide of length l, covered by supermirrors with an index M and consisting of h x k blades (horizontal x vertical); the blades have a thickness of 0.5 mm and may be coated on both sides;
- a heavy input slit acting as a virtual source (VS), placed at the optimum focusing position of the guide nose and having a maximum opening of 10 x 20 mm^2.

Figure 3 collects part of the results of the simulations of the tapered parabolic focusing guide to optimize the m value of the coating, the length l, the focusing distance f and the divergence angle α. From the simulations of focusing noses with different length l, we can infer that a length around 1000 mm is the optimum in terms of sharpness of the beam profile at the focal point (cf. Figure 3a), while keeping the focal distance above the technical minimum required by the *exTAS* layout.

To find out the position of focal point f we made use of a combined intensity and profile width plot as presented in Figure 3b (only horizontal for clarity). The integrated intensity does not vary significantly in the range of some centimeters around the focal point, while the profile width has a minimum at around 250 mm from the end of the guide of length 1000 mm.

Table 1: Comparison between the white beam integrated neutron flux from a straight guide and from a focusing nose with 8x8 blades of $m = 4$, length l = 1000mm and an exit window of 30x60 mm^2, at the guide exit and just after the VS. Integrated intensity calculated over the entire exit window of the guide in line 1 and over the 10 x 20 mm^2 area of the VS in line 2.

Integrated flux	Straight guide	Parabolic l = 1000 mm	ratio
Guide exit	1.00 x 10^{12}	0.40 x 10^{12}	0.40
VS 10x20 (w x h)	0.28 x 10^{11}	0.13 x 10^{12}	4.5

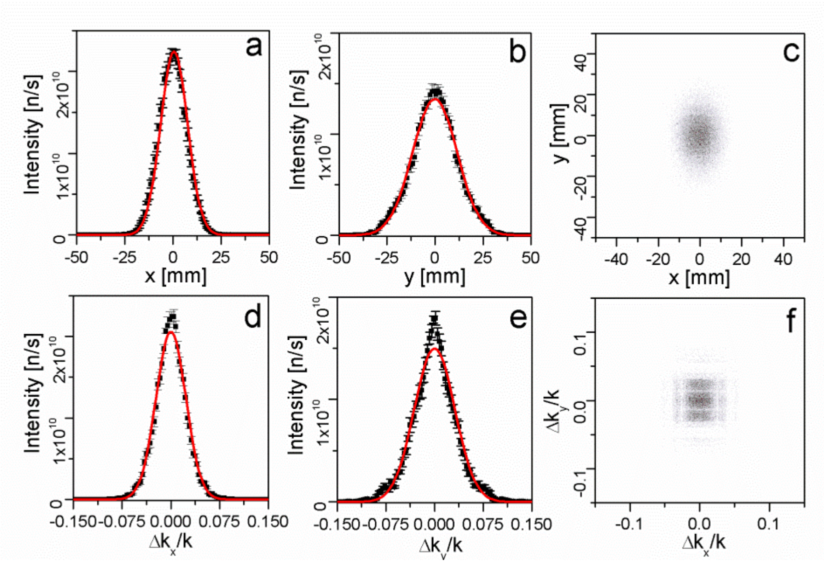

Figure 4: Real space (a-b) and momentum space (d-e) beam profiles together with a neutron density map (c) of the real space focal point and with the momentum density map (f) obtained from the central area 5 x 5 mm^2 of (c). The beam profiles have been fitted with Gaussian curves (red lines) and corresponding calculated FWHM are: (a) 16.5 mm, (b) 27.4 mm, (d) 0.054, (e) 0.073. x axis represents the horizontal direction perpendicular to beam path, y axis represents the vertical direction perpendicular to beam path.

The plot of the variation of integrated intensity versus m value is reported in Figure 3c. Black points refer to simulations without collimating blades while red points refer to simulations with 8x8 blades. In the first case the $m = 5$ value is the most performing solution, while the inclusion of collimating blades makes m = 4 the best compromise due to absorption effects. Even if these effects should be prevented in general, a compromise solution to obtain also a decent real and reciprocal space profiles is necessary in reality.

The profile of the beam at the VS position in real and reciprocal spaces is reported in Figure 4 for a 2D multichannel focusing guide with 8 x 8 blades of $m = 4$ and length $l = 1000$ mm. The profiles have been fitted with a Gaussian distribution to verify the level of homogeneity obtained. The results appear satisfactory and also in the 2D maps of neutron events in Figure 4 c,d display smooth and almost featureless intensity distributions.

Integrated intensity has been calculated and compared with a similar setup where the last 1000 mm of the guide have been kept straight in Table 1. Even if the absorption in the internal blades of the focusing nose halves the output beam flux with respect to the straight guide, the focusing effect allows to concentrate useful neutrons at the VS position and to obtain a gain factor close to 5 in brilliance. This increase is accompanied by an increase in divergence to about 3.5º.

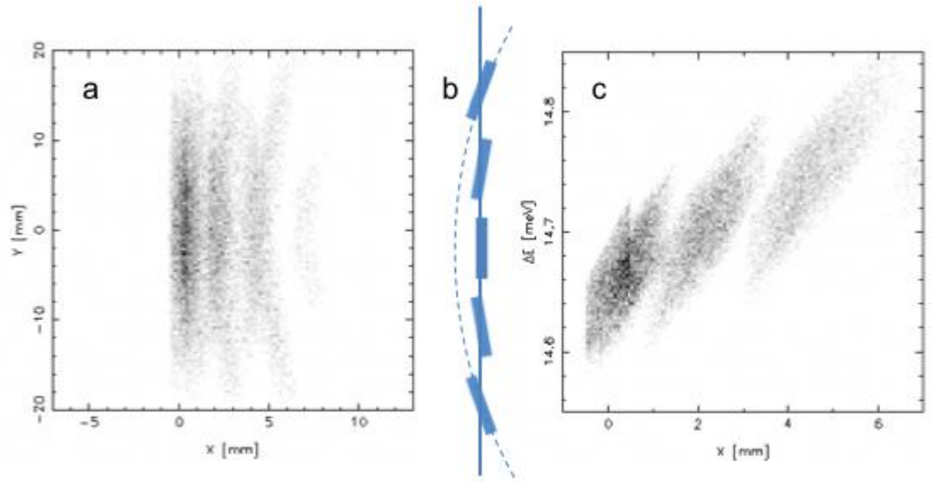

Figure 5: (a) Real space image of an *exTAS* VS slit 1 mm wide at the sample position by a doubly focusing Si 111 monochromator consisting of a vertical array of 9 blades 200 x 20 x 1 mm^3 set for optimum horizontal and vertical curvatures; the fringes are due to departures from perfect sagital focusing (cf. scheme (b)); (c) displays the corresponding shifts in neutron energy on an enlarged scale. x axis represents the horizontal direction perpendicular to beam path, y axis represents the vertical direction perpendicular to beam path.

Beam divergences of several degrees can be efficiently handled and refocused on the sample by large area horizontally and vertically focusing monochromators. In Figure 5 we present SimRes results for an image of a VS slit 1 mm wide at the sample position, projected by a conventional doubly focusing Si 111 monochromator using a vertical array of 9 elastically bent perfect silicon crystals. Thanks to the absence of mosaic structure the image is sharp in the horizontal plane (cf. the millimeter-scale in Figure 5). However, the vertical focusing of the present systems is only approximate, because for practical reasons it uses tilts of the vertical monochromator segments around axes lying in the vertical plane of the central segment. The lateral displacements with respect to the cylindrical surface, which would provide ideal sagital focusing, give rise to lateral aberrations of the focal spots and to slight deviations from ideal monochromatic focusing (cf. right-hand pane in Figure 5). This defect can be neglected when using mosaic crystals (PG, Cu), but cannot be tolerated in high-resolution conditions as those required for *exTAS*.

5. Concluding remarks

The combined implementation of advanced focusing techniques presented in this paper should provide a platform for boosting the TAS performance, when studying mm-sized samples, by at least an order of magnitude. To achieve this goal, the challenges in the development and realization of two key *exTAS* components have to be addressed. The first one is the multi-blade configuration of the two-dimensional tapered guide nose [8], up to now mostly considered only in its 1D form [9] and never implemented in an instrument, even at a prototype level. The other one is the mechanical realization of the true sagital focusing geometry in a 2D variable focusing monochromator/analyzer system required in case of mm-sized focal spots. Our preliminary ray-tracing simulations confirm the key role of these components in terms of optical accuracy and of increased luminosity of the instrument and motivate further development work along these directions. Most of the remaining ideas and elements of the *exTAS* project, outlined in this paper, represent relatively straightforward extrapolations of the present neutron instrumentation and neutron optics technologies, based on experience with design and construction of existing instruments.

Acknowledgement

We would like to mention the creative input of A. Hiess in the early stages of the project, notably concerning the sample environment part of *exTAS*, in the meantime materialized in the *VacBox* sample chamber, which become available to the ILL TAS users recently.

References

[1] Popovici M, 1975 *Acta Cryst.* A **31** 507-513
[2] Komarek A C, Boeni P, Braden M, 2011 *Nuclear Instruments and Methods in Physics Research* A **647** 63-72
[3] Janoschek M, Boeni P, Braden M, 2010 *Nuclear Instruments and Methods in Physics Research* A **613** 119-126
[4] Schneider M, Stahn J, Boeni P, 2009 *Nuclear Instruments and Methods in Physics Research* A **610** 530-533
[5] Kulda J, Šaroun J 1996 *Nuclear Instruments and Methods in Physics Research* A **379** 155-166
[6] Hiess A, Jiménez-Ruiz M, Courtois P, Currat R, Kulda J and Bermejo F J 2006 *Physica* B **385-386** 1077-1079
[7] Boehm M, Hiess A, Kulda J, Roux S and Šaroun J 2008 *Meas. Sci. Technol.* **19** 34024
[8] Šaroun J and Kulda J 2006 *Physica* B **385-386** 1250-1252
[9] Čermák P, Boehm M, Kulda J, Roux S, Hiess A, Steffens P and Saroun J 2013 *J. Phys. Soc. Japan Suppl. A* **82** SA026