



MULTIXS : A New Scanning Multianalyzer X-ray Emission Spectrometer at the GALAXIES Beamline at Synchrotron SOLEIL

J. Ablett, Anthony Berlioux, Dominique Prieur, Jack Harrison, Lars Heller,
Sebastian Gliga, Jean-Pascal Rueff

► To cite this version:

J. Ablett, Anthony Berlioux, Dominique Prieur, Jack Harrison, Lars Heller, et al.. MULTIXS : A New Scanning Multianalyzer X-ray Emission Spectrometer at the GALAXIES Beamline at Synchrotron SOLEIL. 2025. hal-05018895

HAL Id: hal-05018895

<https://hal.science/hal-05018895v1>

Preprint submitted on 3 Apr 2025

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

MULTIXS : A New Scanning Multianalyzer X-ray Emission Spectrometer at the GALAXIES Beamline at Synchrotron SOLEIL

James. M. Ablett,¹ Anthony Berlioux,¹ Dominique Prieur,¹ Jack Harrison,² Lars Heller,² Sebastian Gliga,² and Jean-Pascal Rueff^{1,3}

¹⁾*Synchrotron SOLEIL, L'Orme des Merisiers, Départementale 128,
91190 Saint-Aubin, France*

²⁾*Swiss Light Source, Paul Scherrer Institute PSI, 5232 Villigen PSI,
Switzerland*

³⁾*Sorbonne Université, CNRS, Paris 75005, France*

(Dated: 3 April 2025)

We present the design and performance of a new multi-crystal x-ray emission spectrometer installed at the GALAXIES beamline at Synchrotron SOLEIL. The new instrument, which we name 'MULTIXS', can host up to 5 analyzer crystals and supersedes our previous XES spectrometer design, providing a compact, simple design with all the analyzer crystals contained in the horizontal sample plane. This feature provides a direct view to the sample area and avoids the potential masking of the sample for constrained sample environments. This new design allows for the use of both 0.5m as well as 1m radius spherical analyzer crystals. In addition, the ability of continuously scanning the spectrometer energy provides relatively fast scanning with high quality emission data and minimum dead-time overhead.

I. INTRODUCTION

Inelastic x-ray scattering (IXS) is a widely used technique to probe the physico-chemical properties of materials. As an all-photon, orbital and element sensitive approach, IXS has found interest in a wide range of applications including correlated materials¹, high-pressure research² and chemistry or catalysis^{3,4} both in the soft and hard x-ray regions.

The detection of the scattered (or emitted) x-rays is normally realized with a dedicated spectrometer using dispersive or focusing optics. In the tender to hard x-ray region, which is the main interest here, these spectrometers all use analyzer crystals as energy selective optical elements which can be operated in focusing (Johann or Johansson) or dispersive (Von H  mos) geometries. There are numerous IXS spectrometers which operate both in the soft x-ray and hard x-ray range on different synchrotrons, which we cannot cite extensively. A recently published primer on Resonant IXS lists the synchrotron radiation facilities around the world where these spectrometers can be found⁵, in addition to a new five-analyzer Johann spectrometer at the inner shell spectroscopy beamline at NSLS-II⁶ and an off Rowland circle dispersive refocusing XES spectrometer⁷. The spectrometers efficiency depends mostly on collection solid angle as well as the analyzer crystals being employed. As the analyzer size cannot be arbitrarily increased due to optical aberrations, a large-solid angle spectrometer is normally achieved by decreasing the sample-analyzers distance and/or increasing the number of analyzers.

In this article, we report on the development of a new multi-analyzer x-ray emission spectrometer (MULTIXS) for x-ray emission spectroscopy (XES) and high-energy resolution fluorescence detected x-ray absorption spectroscopy (HERFD-XAS) at the GALAXIES beamline at Synchrotron SOLEIL. As explained below, the novelty of the instrument resides in the combination of a flexible design which allows to fit both $R = 1\text{m}$ and $R = 0.5\text{m}$ analyzers, the in-line scanning geometry where the analyzers are kept in the scattering plane and the possibility to perform continuous, dead-time free scanning.

Details of the GALAXIES beamline are described elsewhere⁸. Briefly, the beamline is dedicated to high-resolution x-ray spectroscopy techniques in the tender to hard x-ray range with a photon energy ranging from 2.3 to 12 keV. The beamline comprises of two endstations for hard x-ray (high kinetic energy) photoelectron spectroscopy and resonant inelastic x-ray scattering (RIXS) experiments. The x-rays produced by the GALAXIES U20 in-vacuum

undulator source (20mm period, 98 periods) are monochromatized by a liquid nitrogen cooled 2-bounce Si(111) monochromator. A four-bounce post-monochromator equipped with Si(110) crystals can be inserted in the beam to achieve energy higher resolution.⁹ The beam is then focused onto the endstations using toroidal mirrors into a $30[\text{vertical}] \times 80[\text{horizontal}] \mu\text{m}^2$ FWHM spot. For the RIXS endstation, a smaller spot size can be achieved by using Kirkpatrick-Baez mirrors leading to a spot size of $15 \times 15 \mu\text{m}^2$ FWHM. An in-vacuum quarter-wave plate can also be used to convert the beam polarization from nominal linear horizontal to linear vertical or circular/elliptical polarizations.

The RIXS endstation is equipped with a variety of spectrometers, all operating in the vertical scattering plane, which are specialized for the various flavors of RIXS: XES, HERFD-XAS or non-resonant inelastic x-ray scattering / x-ray Raman scattering. Among the spectrometers at GALAXIES, we developed in the early days of the beamline a 4-crystal XES Rowland circle spectrometer in the Johann focusing geometry¹⁰. This multi-analyzer instrument was designed to select the spectrometer energy by simply moving the analyzer crystals vertically, in order to change the Bragg angle, and translating the detector vertically to intercept the Bragg-reflected x-rays. In addition, the focal distance was maintained by a lateral translation of the analyzers. While this configuration provided for a very simple motorization and scanning procedure and avoided the need for high-precision analyzer and detector rotation stages, it had some drawbacks. The main problem was that the vertical position of the analyzers crystals, which always lie above the sample horizontal plane, provided a limited view to the sample area and the x-ray path length exiting through the sample is Bragg-angle dependent. Sample environments with restricted opening angles, such as high-pressure cells, cryostats, furnaces or liquid cells could pose a problem especially at low Bragg angles, where the analyzers height above the horizontal sample plane becomes significant, and could potentially mask the sample. This primary reason motivated us to reconsider our multi-analyzer design with the aim to keep the analyzers always in the horizontal sample plane.

II. SPECTROMETER DETAILS

A. Design

The MULTIXS spectrometer can accommodate $R = 1\text{m}$ and $R = 0.5\text{m}$ analyzers, and there are currently four analyzer modules available to use for both set-ups. The $R = 1\text{m}$ configuration has space for an additional analyzer module which is planned to be procured in 2025. The gain in terms of solid angle for $R = 0.5\text{m}$ comes at the cost of energy resolution depending on the Bragg angle as the Johann error increases significantly for small R . The 0.5m configuration nevertheless appears well adapted to experiments having intrinsically low count rates such as in diluted samples or dichroic measurements or for fast acquisitions. The development of 0.5m spherical bent analyzers is described in Rovezzi *et al.*¹¹

1. Geometry

The spectrometer is laid out in the Rowland circle Johann configuration in the vertical scattering plane. In this geometry, the sample, the analyzer operated at a Bragg angle θ_B and the detector are positioned on a circle whose diameter equals the analyzer bending radius R as shown in Figure 1 (for a single analyzer). To place the analyzer in the sample plane, the Rowland circle is rotated around the sample by $90 - \theta_B$ so that the analyzer - sample direction is horizontal.

In the Rowland circle geometry, the analyzer-sample distance (d_{AS}) is defined by :

$$d_{AS} = R \sin(\theta_B) \quad (1)$$

and the sample-detector (d_{SD}) distance by :

$$d_{SD} = R \sin(2\theta_B) \quad (2)$$

The multi-analyzer configuration requires an additional geometrical transformation. To keep the Bragg angle constant on each analyzer, the analyzers are positioned on different Rowland circles which are rotated by an angle ψ around the sample - detector direction. This leads to a fan-like configuration with the analyzers equally distributed in angle around the sample as illustrated in Figure 2. For 1m radius analyzers, we chose $\psi = [-15, -5, +5, +15]$ and $\psi = [-20, -10, 0, +10, +20]$ degrees for four- and five crystal set-ups respectively. For

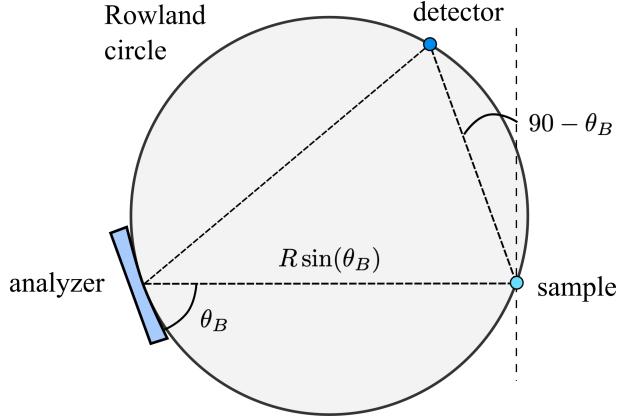


FIG. 1. Rowland circle geometry for a spherically bent crystal analyzer with bending radius R operated at a Bragg angle θ_B . In this geometry, the circle diameter equals the analyzer bending radius. To keep the analyzer-sample direction horizontal, the circle is rotated around the sample by $90 - \theta_B$.

0.5 m radius analyzers, $\psi = [-33, -11, +11, +33]$ degrees. These arrangements provide adequate space between the analyzers to avoid collisions based on the Bragg angular range of between 70 and 90 degrees while keeping a compact configuration.

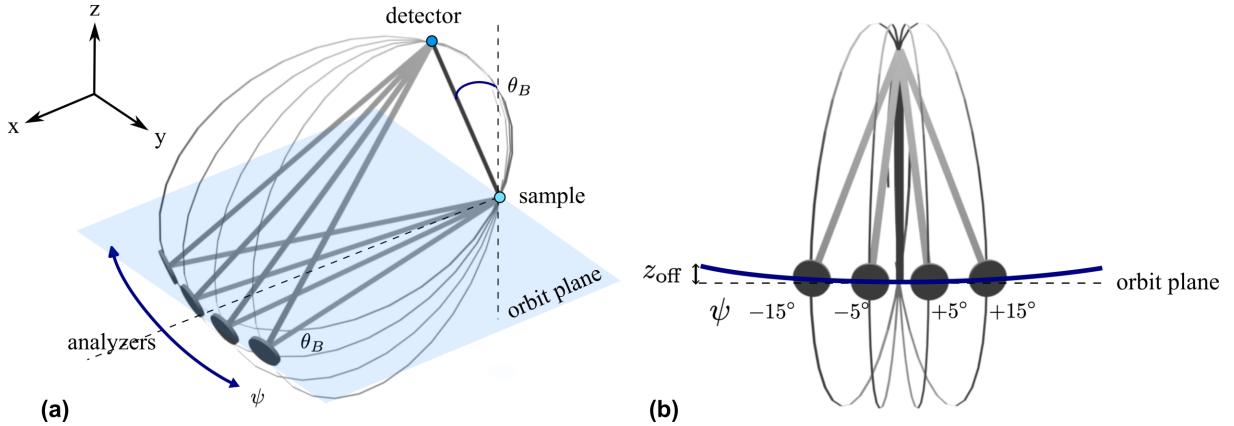


FIG. 2. (a) Configuration of the 4-crystal spectrometer for analyzers with a 1m radius. The analyzers are contained in the horizontal plane and positioned on individual Rowland circles which are obtained by a rotation ψ around the sample-detector direction. (b) The angular position of the different analyzers ψ is indicated on the right ; z_{off} is the change of analyzer height due to the ψ rotation.

2. Z offset

As the detector-sample direction is not vertical, the horizontal fan-like transformation of the analyzers imposes a small (millimeter) change of the analyzers height with respect to the horizontal plane, as shown in Figure 2(b), which depends on θ_B , ψ and R according to Eq. 3. The height offset z_{off} is graphically illustrated in Figure 3: the lower the Bragg angle, the larger the correction. In the worse case scenario for the 1 m bending radius configuration ($\theta_B = 70$ deg, $\psi = 15$ deg), z_{off} reaches a value of 10.3 mm which remains small with respect to the typical analyzer diameter (100 mm). Even for the 0.5 m radius analyzers, this height offset is still less than half of the analyzer diameter ($\theta_B = 70$ deg, $\psi = 30$ deg).

$$z_{\text{off}} = 2R \sin(\theta_B)^2 \cos(\theta_B) \sin(\psi/2)^2 \quad (3)$$

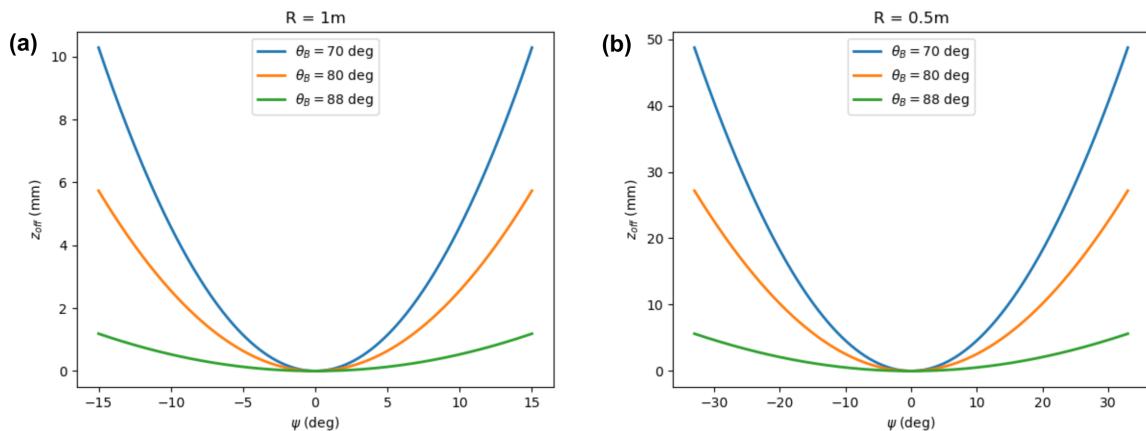


FIG. 3. Analyzer height z_{off} in mm as a function of the angular position ψ for different Bragg angles θ_B and a bending radius of $R = 1\text{m}$ (a) and 0.5m (b)

B. Realization

The spectrometer modules and assembly design were realized principally by the technical support group at Synchrotron SOLEIL. The motorization is based on a commercial solution using encoded motor stages and control from MKS Newport. The interface plate, allowing for the simple incorporation of both 0.5 m and 1 m radius analyzer crystals, was realized by the Swiss Light Source. Each analyzer module is secured to this interface plate by screws from

below the plate. Changing the analyzer modules between 0.5 m and 1 m radius crystals is a straightforward procedure and can be performed within a half hour. In addition, the ability to add a 5th analyzer to the 1 m radius crystal array in the future has been incorporated.

1. Analyzer module

Each analyzer is mounted on an independent module composed of four motorized stages to adjust precisely the analyzer surface to the Rowland circle (see Figure 4(a) inset). Two rotation stages serve to move the analyzer Bragg angle (θ_B stage) and in-plane angular correction (χ stage) while the linear stages serve to correct for the analyzer height (z stage) and position along the analyzer-sample direction (x stage), both depending on the Bragg angle and analyzer radius R (see Figures 1 and 3).

2. Full Assembly

The complete MULTIXS spectrometer assembly is shown in Figure 4 with a maximum of five analyzer modules. These modules are mounted on a base plate made of stainless steel with an angular spread respecting the distribution in ψ . The base plate with both $R = 1$ m and $R = 0.5$ m radius analyzers is shown in Figure 4(b). The plate itself is attached to a L-shape heavy-duty foot which is bolted onto the RIXS endstation at the GALAXIES beamline when required, as illustrated in Figure 4(a).

3. Implementation

The spectrometer is installed on the RIXS endstation of the GALAXIES beamline at Synchrotron SOLEIL. The endstation has been described elsewhere and consists of a long 2 m arm which can rotate both in the vertical and horizontal planes according the T_{γ_1} and δ_1 rotations and a short 1 m horizontal arm rotation in the horizontal plane around δ_2 ^{8,12}. The implementation of the MULTIXS spectrometer on this endstation is shown in Figure 5. The spectrometer bracket is fixed onto the δ_2 short-arm. The spectrometer can then rotate around the sample position in the horizontal plane to define the experimental scattering angle. During experiments, a helium filled bag is usually placed in the space between the

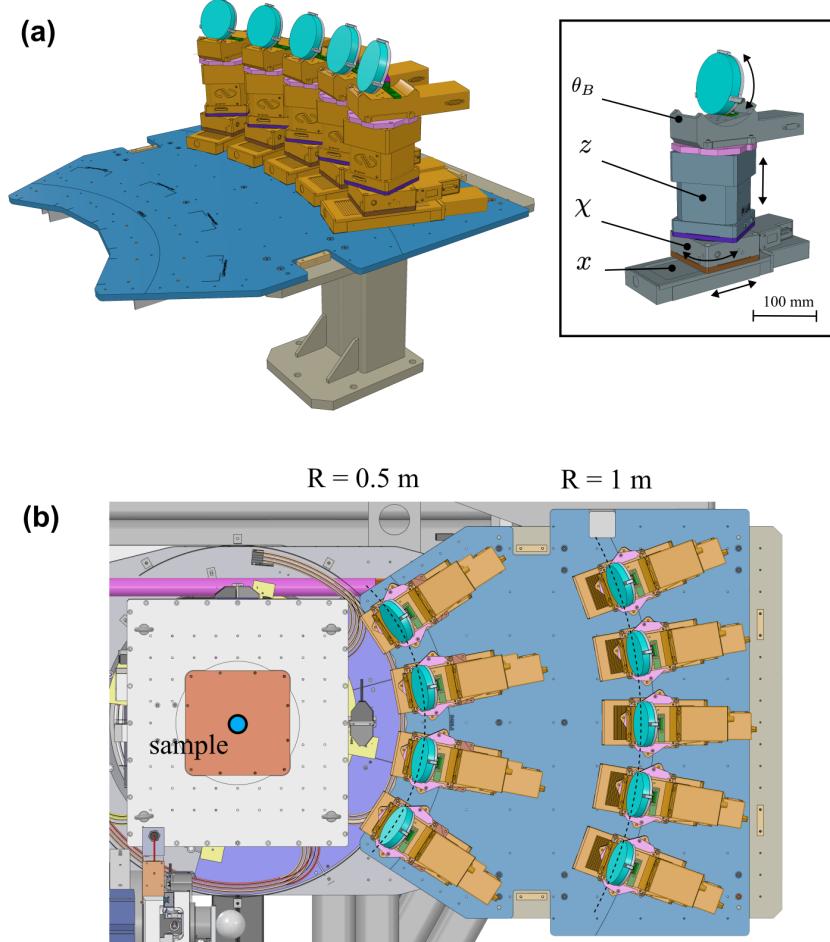


FIG. 4. Overview of the crystal analyzers assembly (a) and top view (b) showing both $R = 1\text{m}$ and 0.5m configurations. The motorization of a single analyzer module is shown in the inset. It comprises of two rotations (θ_B , χ) and two translations (x and z). The spectrometer base plate is mounted on a L-bracket

sample, analyzer and detector to limit the absorption of x-rays by air.

The detector is mounted on the spectrometer vertical long arm. The long arm main translation T_{γ_1} and rotation γ_1 are used to position the detector precisely on the Rowland circle as the Bragg angle is varied. For the detector, both energy-dispersive silicon drift detectors (SDDs) or hybrid pixelated 2D imagers are available. In general, we use a 2D imager for alignment purposes and an SDD for experimental data collection, due to its excellent energy resolution, which can be used to remove unwanted background contributions from the spectra.

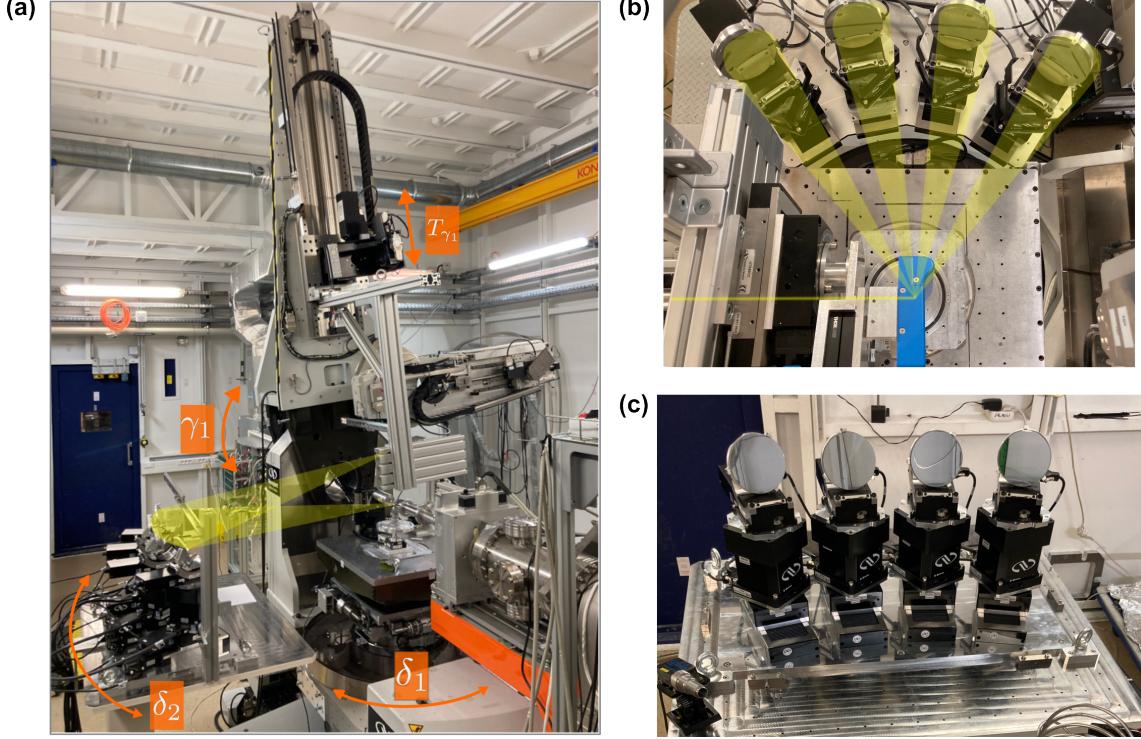


FIG. 5. Views of the MULTIXS spectrometer on the RIXS endstation at the GALAXIES beamline. The spectrometer is placed on the δ_2 horizontal arm with the detector attached to the long-arm placed on the δ_1 rotation (a). The detector position is adjusted by the T_{γ_1} translation and γ_1 rotation. (b) and (c) are views of the $R = 0.5\text{m}$ and 1m configuration. Yellow lines and areas indicate the incident X-ray beam and scattered beams. Note that (a) and (c) show the base plate dedicated to solely $R = 1\text{m}$ analyzer modules, whereas b) shows the new base plate with 0.5m radius crystals installed.

III. PERFORMANCES

A. Analyzers and Resolution

The beamline can provide a large variety of Si or Ge crystal analyzers which are adapted to the energies of many emission lines of interest. A complete list of analyzers can be found on the beamline website. The accessible emitted energy is approximately 3.5-12 keV which covers numerous emission lines from Ca to Bi including many $3d$, $4f$ or $5d$ elements.

The spectrometer resolution depends on the analyzer type and quality and the Bragg angle. A detailed description of the different factors which contribute to the spectrometer

resolution can be found in Refs. 13 and 14. For MULTIXS, the typical energy resolution is of the order of 0.3 to 1eV.

B. Scanning

The spectrometer scanning procedure is performed by a series of Python scripts that has been developed at the GALAXIES beamline and based on the TANGO control infrastructure. The scanning workflow is described in Fig. 6 for step by step a) or continuous modes b).

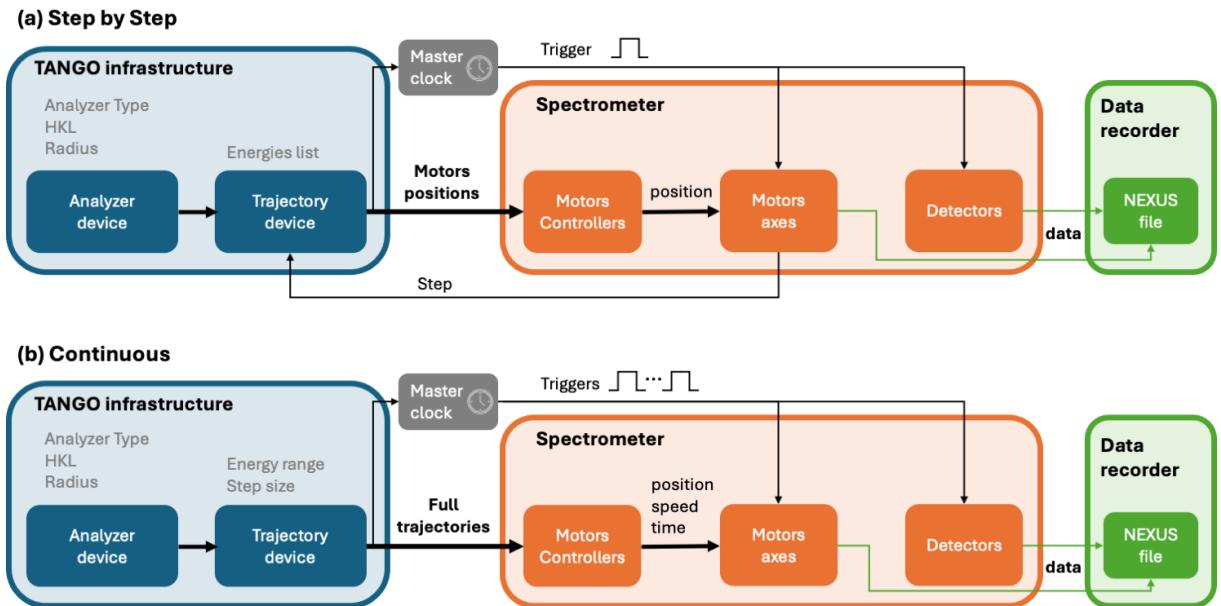


FIG. 6. Scanning workflows for the step by step (top) and continuous (bottom) mode: the analyzers and scan parameters are entered as inputs. Motors positions or full trajectories are sent to the motors controllers and motors axes. Synchronization between the detectors and motor axes positions is ensured by duplicate trigger signals from the master clock. The motors positions and data are saved in a hierarchical data format (HDF) NEXUS file.

Firstly, the analyzer is defined by providing the material type (Si or Ge), hkl Miller indices of the considered Bragg reflection and analyzer bending radius. The spectrometer scanning parameters are configured by providing the starting energy, final energy and energy step size, which are used as inputs to the TANGO devices that calculate the position of the MULTIXS motors. In total, there are 18 motors to scan to acquire an x-ray emission spectra

(four motors for each analyzer module and two motors for the detector arm). Each of the analyzer modules can be configured separately, depending on the analyzer being used, and the number of analyzer modules to be scanned can be selected. For example, 2 analyzer modules may be fitted with Ge 440 analyzers for Fe K α emission and the remaining modules equipped with Ge 620 for Fe K β emission. Based on the analyzer and scan parameters, the scan can be performed by either stepping the position or through continuous motion. In the latter case, the full trajectories are uploaded to the controllers prior to the scan, in arrays containing the positions of the motors as a function of time. The controllers then convert these arrays into position, velocity and time for each of the motor axes according to their individual trajectories. Synchronization between the various detectors and motor axes positions is ensured by trigger signals from the master clock. If needed, the scan function itself can be inserted in a Python script to automatically measure the emitted spectra as function of a varying external parameter such as the incident energy, the sample position or sample temperature. The complete experimental data is saved in a hierarchical data format (HDF) NEXUS file including the MULTIXS detector data, scan parameters, intensity monitors, beamline metadata and all motors positions.

C. Results

1. XES, HERFD-XAS

Figure 7 illustrates the use of MULTIXS for X-ray emission spectroscopy here in Mn₂O₃ powder. These XES measurements were performed with the MULTIXS spectrometer equipped with a single 0.5m radius Ge(440) crystal analyzer both in step-by-step ('step') and continuous ('zap') modes using a SDD detector. The incident x-ray energy was fixed above the Mn K-edge at 7000eV (using the GALAXIES Si 111 monochromator) and the x-ray emission energy was scanned between 6450eV and 6520eV with a step size of 0.25eV. Energy calibration was ensured by measuring a standard Mn foil. Each data set is comprised of a single x-ray emission scan. As shown, the quality and correspondence between the step and zap scans with 1s integration time are identical. For the zap scan, the total scan time was 4mn 41s which is 3.3x faster compared to that using the step scan. Faster zap scans down to 0.15s integration time, with this particular energy range and step size, were possible

while maintaining excellent data quality. Total scan times for zap scans with 0.5s, 0.25s and 0.15s were 2mn 21s, 1m 10s and 42s respectively.

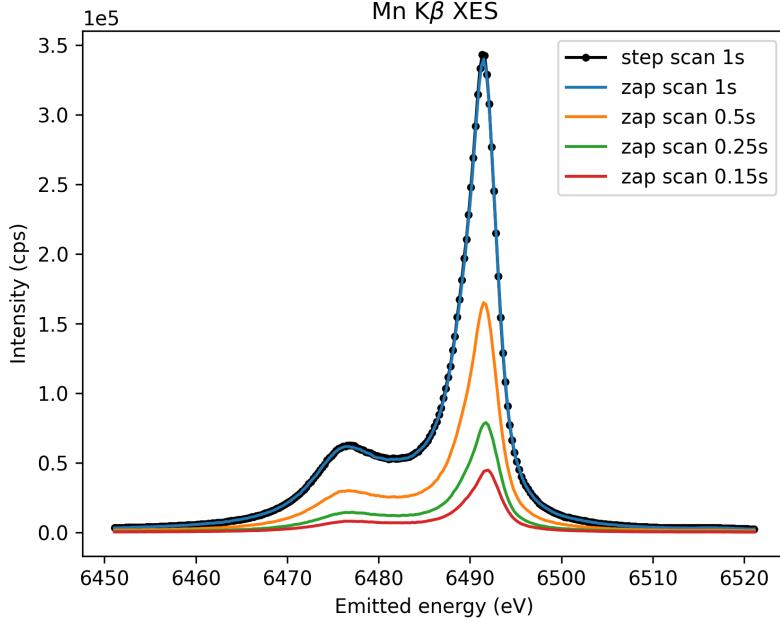


FIG. 7. Mn K β XES measured for Mn₂O₃ powder with MULTIXS equipped with a single Ge(440) 0.5m radius crystal analyzer. Comparison with XES acquired in 'step' mode with an integration time of 1s per point, are compared to those performed using continuous 'zap' scans with 1s, 0.5s, 0.25s and 0.15s. Excellent data quality is maintained using the faster zap scans, down to a total scan time of 42s, for this particular energy range and energy step size.

2. RXES map

An example of a resonant XES (RXES) intensity map in Mn₂O₃ powder is shown in Figure 8 using the same Ge(440) 0.5m radius analyzer crystal and a SDD detector. The measurements were carried out by continuously scanning the MULTIXS emission energy, which included the Mn K β_1 and valence band emission, for each incident energy, using the Si 111 monochromator, across the Mn K-edge. The x-ray emission energy was scanned between 6450eV and 6520eV with a step size of 0.2eV and an integration time of 0.25s per interval. Each of these 'zap' scans took 2mn 06s to complete. The incident energy range was varied across the Mn K-edge from 6532eV to 6546eV in 0.2 eV steps. The total scanning

time for the RXES map was 2hr 27mn, with a total effective 'dead time' of 15mn which primarily consisted of the time taken to move the MULTIXS motors back to their starting positions as well as moving the incident monochromator energy. The total time required for the RXES map was subsequently 2hr 42mn.

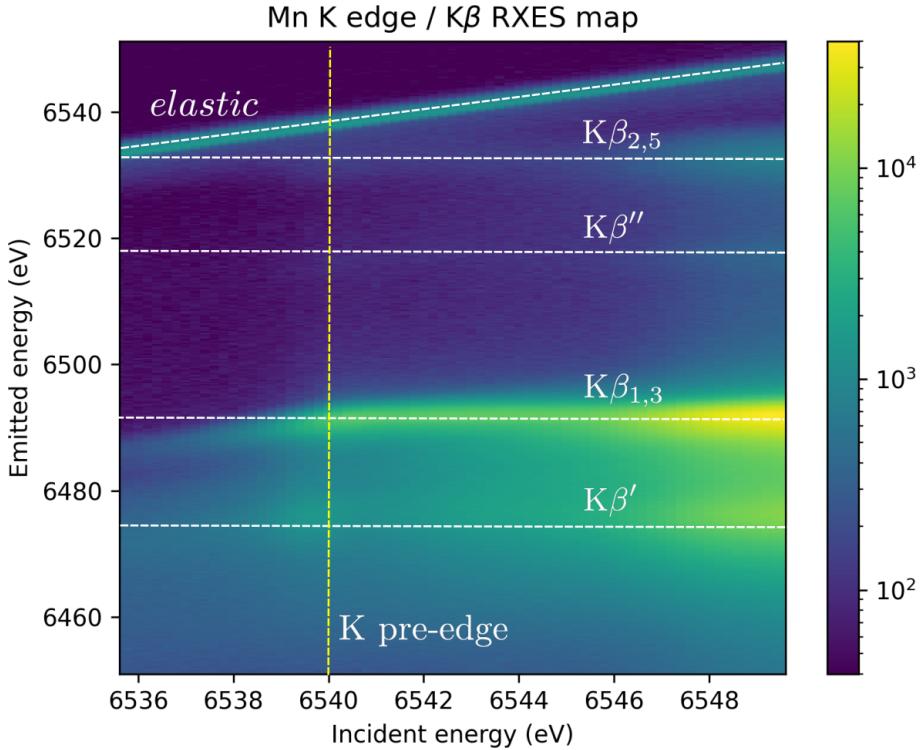


FIG. 8. Resonant Mn 'K β ' XES intensity maps in Mn_2O_3 at the Mn K-edge. The x-ray emission energy was scanned continuously for each incident energy, with an integration time of 0.25s per interval. The total time required for this RXES map was 2hr 42mn. The x-ray emission and incident energies were subsequently calibrated using a standard Mn foil.

IV. CONCLUSIONS AND PERSPECTIVES

MULTIXS is a new multi-analyzer spectrometer at the GALAXIES beamline dedicated to XES and HERFD-XAS in the 3.5 to 12keV range. The spectrometer is designed for using up to five spherically bent crystal analyzers with 1 m or four with 0.5 m bending radius, all lying in the sample plane, offering a large view to the sample area. The change between these analyzer modules is relatively quick and can be performed easily within an experimental run. The implementation of continuous scanning for the MULTIXS spectrometer provides

extremely fast and high-quality x-ray emission spectra with minimal dead-time overhead during an experiment. The instrument is operational and open to the users community through the SOLEIL standard proposal application.

ACKNOWLEDGMENTS

We acknowledge SOLEIL for the provision of synchrotron radiation facilities and the SOLEIL technical support group for the design of the MULTIXS spectrometer. S.G., J.H. and L.H. acknowledge funding by an Innosuisse & Swiss National Science Foundation BRIDGE Discovery grant, number 211517.

REFERENCES

- ¹L. J. P. Ament, M. van Veenendaal, T. P. Devereaux, J. P. Hill, and J. van den Brink, Rev. Mod. Phys. **83** (2010).
- ²J.-P. Rueff and A. Shukla, Rev. Mod. Phys. **82**, 847 (2010).
- ³U. Bergmann and P. Glatzel, Photosynthesis Research **102**, 255 (2009).
- ⁴J. K. Kowalska, F. A. Lima, C. J. Pollock, J. A. Rees, and S. deBeer, Israel Journal of Chemistry **56**, 803 (2016).
- ⁵F. de Groot, M. Haverkort, H. Elnagger, A. Juhin, K.-J. Zhou, and P. Glatzel, Nature Reviews methods primers , 1 (2024).
- ⁶A. Tayal, D. Coburn, D. Abel, M. Rakitin, O. Ivashkevych, J. Wlodek, D. Wierzbicki, W. Xu, E. Nazaretski, E. Stavitski, and D. Leshchhev, Journal of Synchrotron Radiation , 1609 (2024).
- ⁷W. M. Holden, O. R. Hoidn, A. S. Ditter, G. T. Seidler, J. Kas, J. L. Stein, B. M. Cossairt, S. A. Kozimor, J. Guo, Y. Ye, M. A. Marcus, and S. Fakra, Review of Scientific Instruments **88**, 073904 (2017).
- ⁸J.-P. Rueff, J. M. Ablett, D. Céolin, D. Prieur, T. Moreno, V. Balédent, B. Lassalle, J. E. Rault, M. Simon, and A. Shukla, J. Synchrotron Rad. **22**, 175 (2015).
- ⁹J. M. Ablett, J. Dubuisson, T. Moreno, D. Céolin, D. Raimon, D. Prieur, D. Corruble, A. Lestrade, C. Bourgoin, and J.-P. Rueff, Journal of Physics Conference Series **425**, 1 (2013).

¹⁰F. Gélebart, M. Morand, Q. Dermigny, P. Giura, J.-P. Rueff, and A. Shukla, AIP Conference Proceedings **879**, 1837 (2007).

¹¹M. Rovezzi, C. Lapras, A. Manceau, P. Glatzel, and R. Verbeni, Review of Scientific Instruments **88**, 013108 (2017).

¹²J. M. Ablett, D. Prieur, D. Céolin, B. Lassalle-Kaiser, B. Lebert, M. Sauvage, T. Moreno, S. Bac, V. Balédent, A. Ovono, M. Morand, F. Gélebart, A. Shukla, and J.-P. Rueff, Journal of Synchrotron Radiation **26**, 263 (2019).

¹³U. Bergmann and S. P. Cramer, SPIE-Proceedings **3448**, 198 (1998).

¹⁴P. Glatzel, A. Harris, P. Marion, M. Sikora, T.-C. Weng, C. Guilloud, S. Lafuerza, M. Rovezzi, B. Detlefs, and L. Ducotté, Journal of Synchrotron Radiation **28**, 362 (2021).