Preliminary Overview of the Extreme Condition Beamline (EMA) at the new Brazilian Synchrotron Source (Sirius)

R D dos Reis, U F Kaneko, B A Francisco, J Fonseca Jr., M A S Eleoterio and N M Souza-Neto

Brazilian Synchrotron Light Laboratory, Brazilian Center for Research in Energy and Materials, Campinas, Sao Paulo 13083-970

E-mail: narcizo.souza@lnls.br

Abstract. Experimentally achieving extreme thermodynamical conditions of temperature, pressure and magnetic field such as the ones found in the interior of planets and stars has been a dream to many scientists seeking to reproduce those conditions on earth to study and produce unconventional materials. The advent of the 4th generation Brazilian synchrotron source (named after the “Sirius” star) allows us to get closer to this dream by implementing a state-of-the-art beamline facility to study samples under extreme thermodynamical conditions by means of a multitude of synchrotron x-ray techniques. The EMA Beamline (Extreme condition Methods of Analysis) will be able to do this by coupling both microfocus (1x1 µm$^2$) and nanofocus (100x100 nm$^2$) beam sizes to x-ray magnetic spectroscopy, x-ray diffraction and x-ray coherent imaging in multiple experimental instruments, placed along the beam path for optimization. Support laboratories (thermodynamical conditions, nuclear materials, laser and optics) were also planned to fulfil all requirements for the experiments under extreme. The EMA beamline, as overviewed here, should open a plethora of opportunities for diverse studies of materials at extreme conditions with synchrotron x-ray techniques.

1. Introduction

Recently high-pressure techniques have become more common around the world allowing to compress materials to the point where the inter-atomic spacing are reduced by a factor of two and the densities increase more than one order of magnitude. This is especially true when we consider the recent developments of two-stage diamond anvil cells or the specially machined toroidal shaped diamond anvils, both of which intended to reach static pressures as high as 1 TPa. To understand the implication of such huge contraction, small focused X-ray beams, smaller than 1 micrometer, are essential to allow in-situ investigations of crystalline and electronic structure under high-pressure. Today most of the state-of-the-art beamlines dedicated beamlines for high pressure research have x-ray beam sizes as small as 3x3 µm$^2$ with the beam tail extending to several microns, which limits the application of these methods for extreme pressures. At the Extreme condition Methods of Analysis beamline (EMA) we aim at having both ~1x1 µm$^2$ focused beamsize with high photon flux (10$^{12}$ photons/s @ 20 keV) and ~100x100 nm$^2$ focused beamsize (with ~10$^{10}$ photons/s @ 20 keV), both with well-defined gaussian beam shape, which will allow the realization of X-ray absorption (XAS), X-ray diffraction (XRD), coherent diffraction image (CDI) and X-ray Raman experiments at extreme pressure with a good spatial selectivity and avoiding pressure gradients on the diamond culet area. The extreme thermodynamic conditions aimed at the EMA beamline are not limited to high pressures, as most complex scientific problems do need a combination of conditions to
explore yet unreached points of the phase diagram, we aim at coupling the high pressure capabilities to low and high temperatures (as low as 100 mK and as high as 8000 K) and high magnetic fields (up to 11T).

With these capabilities, EMA is designed to allow exploring several areas of science such as magnetism, superconductivity, spintronics, carbon-based materials, geoscience and outer planets by exploring the combination of several x-ray techniques with extreme conditions of pressure, temperature and magnetic field. In this manuscript, we describe the main characteristics of the proposed EMA beamline, as well outline all the possibilities for experiments with x-rays techniques combined with extreme conditions of temperature, pressure and magnetic field will be available once EMA begins their operation.

2. Beamline layout

The source of EMA beamline is based on a delta undulator which has the ability to deliver circular/linear polarized photons. Due to the low divergence of the source, it allows the first optical element of the beamline to be a vertical bounce monochromator [6] which allows stable and fast energy scanning. An x-ray $\frac{1}{4}$ wave plate is also planned to allow fast polarization switching, which is placed right after the monochromator Error! Fonte de referência não encontrada.. Harmonic rejection mirrors, with three stripes (Si, Rh, Pt) will also be implemented in the optics hutch. The focusing optical design of the EMA beamline was certified by ray-tracing and wave propagation simulation results. This demonstrated the need for very low slope error mirrors (<100nrad) to deliver a gaussian shaped focused beam. In addition, this first K-B mirror pair can be dynamically bended to allow a focused beam at several working distances to allow multiple experimental setups in tandem, as shown in the schematic optical layout of the beamline in figure 1 and summarized on table 1. This first K-B mirror will deliver beamsizes ranging from 1 µm to 10 µm (at focus) depending on the working distance used, as described below. This will also be used to generate a secondary source to a additional fixed curvature K-B mirror pair in a second experimental hutch of EMA intended to provide round beamsizes as small as 100x100 nm$^2$ well compatible with x-ray experiments under high pressure, low temperature and applied magnetic.

![Figure 1: Optical layout of the EMA beamline](image-url)
Table 1 – Experimental conditions at multiple experimental setups at the EMA beamline. The last setup is planned for the nanofocus experimental station, in addition to the first three setups at the microfocus experimental station.

| Energy range (keV) | Multipurpose setup | SC Magnet setup | XRS diffractometer setup | Nano station setup |
|-------------------|--------------------|-----------------|--------------------------|--------------------|
| 3 – 30            | 3 – 30             | 3 – 30          | 3 – 30                   | 5 – 20             |

| Beam sizes (@ 10 keV) | Beam divergence (@ 10 keV) | Photon flux (ph/s @ 10 keV, 100mA) | Experimental techniques | Pressure (Max) | Temperature | Magnetic Field (Max.) |
|-----------------------|-----------------------------|------------------------------------|------------------------|---------------|-------------|----------------------|
| 1 x 0.4 µm²           | 1 x 0.6 mrad²               | ~5x10^{12}                         | XAS, XRD, CDI         | ~600 GPa      | 0.5 K to 8000 K | 3 T (1 T w/ pressure) |
| 6 x 2 µm²             | 0.2 x 0.1 mrad²             | ~5x10^{12}                         | XAS, XRD, CDI         | ~300 GPa      | 0.1 K to 300 K  | 11 T                 |
| 13 x 3 µm²            | 1.6 x 0.05 mrad²            | ~5x10^{12}                         | XRS, XRD              | ~300 GPa      | 5 K to 300 K   | 6 T                  |
| 1.5 x 0.1 µm²         | 1.7 x 1.4 mrad²             | ~1x10^{11}                         | XAS, XRD, CDI         | ~ 1 TPa       | 5 K to 8000 K  | 0.5 T                |

3. Experimental capabilities at the microfocus station

The EMA beamline is planned to have two experimental stations to cover both micrometer and nanometer focused beamsizes with high photon flux. While the expected parameter and capabilities of the nanofocus station are shown in table 1, here we focus on first experimental station, the microfocus station, as drawn in figure 2. This station aims to combine x-ray techniques (XAS, XRD, XRS and CDI) with a beam size from 1 to 10 µm, depending of the experiment position, with several extreme thermodynamic conditions of pressure, temperature and magnetic field. This combination should allow to explore yet unreached points of the P-T-H phase diagram to tackle complex scientific problems on the intersection between such thermodynamical conditions.

The three experimental setups displaced along the beam path in the microfocus station are named here as: multipurpose setup, superconducting (SC) magnet setup and x-ray Raman (XRS) diffractometer setup, as summarized on table 1, drawn on figure 2 and described below.
Figure 2: Top view drawings of the three experimental setups at the microfocus experimental station of EMA beamline.

The multipurpose setup, located at the beginning of hutch, has the smallest beamsize possible at the microfocus station (~1x0.4 µm²) which should allow X-ray spectroscopy, diffraction and imaging experiments to be performed up to pressures as high as 600 GPa, low and high temperatures (0.5 K to 8000 K) and under a moderate magnetic field up to 3 T at ambient pressure or 1 T under applied pressure. This multipurpose setup is illustrated in Figure 3. A resistive electromagnet (GMW 3474-140) is used to apply magnetic fields of up 3 T, which is essential for X-ray magnetic circular dichroism experiments (XMCD). A custom LHe flow cryostat will cool down the samples by contact with a cold-finger tip and/or in contact cold Helium exchange gas to cover all shapes and kinds of samples and sample holders. Both of these modes of operation should allow to reach temperatures as low as 1.8 K. In addition, a He3 insert was designed to couple to the cryostat and allow to reach temperatures as low as 0.5 K. In any of these configurations the cool down time from room temperature should be on the order of 25 minutes. In order to keep the cold sample aligned to the x-ray spot position (within 10 nm) an interferometer-based feedback was planned. At this same setup it will also be possible to perform XRD experiments of the sample under low temperature by sliding the electromagnet to the side and fitting in an x-ray area detector with sample-detector distance between 0.1 to 4 meters. The area detector used for XRD experiments will be fixed in a robot arm allowing us to cover a large solid angle and sample-detector distance to allow mapping the reciprocal space. This possibility to easily move from XAS to XRD configuration will optimize the need to perform experiments at the same pressure and temperature conditions to determine both crystalline and electronic structures. Still in this multipurpose setup there will be the possibility to perform XRD and XAS experiments at temperatures as high as 8000 K by employing a dedicated double side laser heating [8] optical apparatus. This is placed on the two level optical platforms on top of the multipurpose setup, as shown in Figure 3, including all optics needed to also perform visible Raman spectroscopy (with 532 nm green excitation Laser), Ruby luminescence as needed for all high pressure experiments.
Figure 3: Drawing of the first experimental setup of EMA beamline. The details of the setup are described on the main text.

At the middle of the experimental hutch, an 11 T superconducting magnet manufactured by Oxford Instruments will allow both XMCD/XANES and XRD experiments, with a focused x-ray beamsize of about 6x2 $\mu$m$^2$. The sample space of this magnet will be compatible with up to 2 inch standard symmetric diamond anvil cells as well as strain cells from Razorbill instruments, opening the possibility of combine the high magnetic field with high pressure and low temperatures (down to 1.8 K). Also, two optical windows will be disposed perpendicular to the beam direction for in situ pressure calibration. This optical window will be also used to a laser interferometer system that will guarantee the stability of the sample position during the measurements. A dilution He$^3$ insert is also planned to be added at this magnet allowing the experiments XAS and XRD be performed in temperatures as low as 100 mK, however with a more limited sample space.

At the end of the experimental hutch, we will implement a X-ray Raman setup [9], which open the possibility to probe light elements absorption edges (Carbon K-edge, in particular) with hard x-rays, which is fundamental to allow extreme conditions requirements such as diamond anvil cell or extreme low temperatures. On this setup, a 36 spherically bent crystal analyzers array is employed in order to increase solid angle acceptance and decrease collection time, which will be mounted on the detector arm of the 6+2 circle diffractometer, as shown in Figure 4. This setup will work with both 0.5m and 1m radius of curvature analyzers depending on the need for higher collection efficiency or higher resolution.
Figure 4. Drawings of a setup with 36 spherical analyzers for X-ray Raman scattering experiments mounted in a 6+2 circle diffractometer.

4. Support laboratories

The research under extreme thermodynamics conditions will be supported by three thematic laboratories which will be installed close to the EMA experimental stations. These will deliver all conditions to perform experiments under high pressure conditions, in nuclear related materials (actinides, radioactive isotopes, etc), as well as provide a laser infrastructure to the couple to the x-ray experiments at the nanofocus station. The capabilities of these laboratories are listed below.

4.1. Extreme thermodynamic conditions

Since most of techniques available at EMA will require the use of Diamond Anvil Cell (DAC) to perform experiments under high applied pressure, we must provide the conditions needed to prepare and load the samples into these devices. With the need for extremely small samples if we intend to dramatically reduce the size of the diamond tip to reach high pressures, this task becomes quite difficult. With this aim, this laboratory should allow:

1) Preparation of specially shaped diamond anvils, either commercially available or by FIB machining;

2) Indentation and perforation of the metallic gasket used to hold the samples. For perforations down to 50 µm we can use an Electro discharge machine (EDM), while for smaller samples spaces laser drilled holes (or FIB drilled holes) will be available;

3) Samples loading into the DAC. For samples bigger than 50 µm the loading can be manually done in stereomicroscopes with high magnification. For smaller samples a computer controlled micro manipulator shall be used. For the case of air sensitive samples, an inert atmosphere environment (glove box) shall also be used for the loading.

4) Loading of a hydrostatic medium. While liquid media are used to provide hydrostaticity at low pressure (<10GPa), noble gases are normally used at high pressures. These gases loading into the DACs will be possibly using a compressor-based gas loading apparatus [9]. In addition, a cryogenic loading apparatus to allow H$_2$ and H$_2$S is also being implemented [11], which may also serve as a backup to load noble gases (Ne, Ar, etc).

4.2. Nuclear materials
Experiments on materials containing nuclear elements require special attention due to the inherent difficulties to handle radioactive or toxic materials. To allow these materials to be handled safely and efficiently for high pressure experiments we planned a support lab specifically for this aim. This lab will work at a pressure lower than 1 atmosphere and will require an isolated ventilation system. The access to the lab will be done in separate antechambers equipped with radioactive contaminant detectors for the user to check radiation content before and after using the lab. Inside the lab, the ambient pressure, temperature and radioactive levels will be constantly monitored. The samples manipulation will be performed inside gloveboxes. For loading diamond anvil cells inside the glovebox, we plan to use a micromanipulator which will allow a more efficient manipulation of the sample while avoiding direct contact. A stereomicroscope outside the glovebox will be available for optical adjustments of the DAC before and after the sample loading.

4.3. Laser and optics
A support lab dedicated to optics instrumentation is planned to be installed attached to the nanofocus hutch at the end of the beamline. This lab is meant to allow the implementation of dedicated laser and optics setups to be used either independently of the x-ray, in order to preliminary tests on the samples, or simultaneously to x-ray experiments.

5. Summary and Conclusion

The Extreme condition Methods of Analysis beamline (EMA) is one of the hard x-ray undulator beamlines within the first phase of the new Brazilian synchrotron source (Sirius). This beamline is designed to allow the study of materials under extreme thermodynamic conditions (pressure, temperature and magnetic field), covering several areas of knowledge in both applied and fundamental science. In particular, the microfocus station was designed to allow several extreme condition techniques today employed at synchrotron laboratories worldwide. In addition to the experimental stations, support laboratories will be strongly linked to the experiments at the beamline, covering the high-pressure instrumentation using diamond anvil cells and uniaxial pressure cells, handling nuclear samples and one laboratory dedicated to visible optics instrumentation for in-situ experiments simultaneously to the x-ray techniques. In summary, at EMA multiple X-ray techniques, such as XRD, XMCD and XRS, are aimed to be combined. This should optimize the extreme thermodynamic sample environments coupled to x-ray techniques in order to fully describe a material’s properties.

ACKNOWLEDGMENTS

The Sirius project is funded by the Brazilian Ministry of Science, Technology, Innovations and Communications (MCTIC). Technical support from all LNLS technical and engineering groups are acknowledged. The EMA team acknowledges financial support from the Brazilian agencies FAPESP (Grants 2013/22436-5, 2014/05480-3, 2018/00823-0 and 2018/19497-6) and Serrapilheira Institute (Grant G-1709-17301).

References

[1] Dubrovinsky L, Dubrovinskaia N, Prakapenka V et al. 2012 Nat. Commun 3 1163 https://doi.org/10.1038/ncomms2160
[2] Jenei Z, O’Bannon E F, Weir S T et al. 2018 Nat. Commun 9 3563 https://doi.org/10.1038/s41467-018-06071-x
[3] Dewaele A, Loubeyre P, Occelli F et al. 2018 Nat. Commun 9 2913
https://doi.org/10.1038/s41467-018-05294-2

[4] Lima F A, Saleta M E, Pagliuca R J S, Eleoterio M A, Reis R D, Fonseca Junior J, Meyer B, Bittar E M, Souza-Neto N M and Granado E 2016 J. Synchrotron Rad. 23 1538

[5] Tolentino H C N, Cezar J C, Watanabe N, Piamonteze C, Souza-Neto N M, Tamura E, Ramos A Y and Neueschwander R 2005 Physica Scripta T115

[6] Geraldes R R et al., 2018 The design of Sirius X-ray mirror systems Proc. MEDI SI

[7] Poldi E H T, Escanhoela Jr C A, Fonseca Junior J, Eleoterio M A S, Reis R D, Lang J C, Haskel D and Souza-Neto N M “A versatile x-ray phase retarder for lock-in XMCD measurements”. To be published

[8] Prakapenka V B, Kubo A, Kuznetsov A, Laskin A, Shkurikhin O, Dera P, Rivers M L and Sutton S R 2008 High Pressure Research 28 225 10.1080/08957950802050718

[9] Kaneko U F, Eleoterio M A S, Fonseca Junior J, Ceppi S, Casa D, Souza-Neto N M et al. To be published

[10] Rivers M, Prakapenka V B, Kubo A, Pullins C, Holl C M and Jacobsen S D 2008 High Pressure Research 28 273

[11] Chi Z, Nguyen H, Matsuoka T, Kagayama T, Hirao N, Ohishi Y and Shimizu K 2011 Review of Scientific Instruments 82 105109