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# First white beam on a von Hámos spectrometer at the PolyX beamline of SOLARIS

W. Błachucki [a,*], K.M. Sowa [b], T. Kołodziej [b], P. Wróbel [c], P. Korecki [b,d], J. Szlachetko [b]

[a] Insitute of Nuclear Physics, Polish Academy of Sciences, Kraków, Poland
[b] National Synchrotron Radiation Centre SOLARIS, Jagiellonian University, Kraków, Poland
[c] AGH University of Science and Technology, Kraków, Poland
[d] Marian Smoluchowski Institute of Physics, Jagiellonian University, Kraków, Poland

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ABSTRACT

Synchrotrons are brilliant sources of X-ray radiation used in a variety of methods to study the structure of matter and dynamics of processes on the atomic scale. X-ray spectroscopy methods at synchrotrons are typically combined with the beam monochromatization which guarantees better energy resolution but also reduces orders of magnitude the photon flux incident on the sample and thus wastes the vast majority of the photons produced. Here we report on the commissioning of an X-ray spectrometer at the PolyX beamline of SOLARIS specialized in application of white, broadband and monochromatic X-ray beam in multimodal microimaging, microtomography and microscpectroscopic studies. The spectrometer was used to acquire good quality Fe K-edge X-ray absorption spectrum over about 120 eV-range within seconds. In this work we present the first X-ray absorption spectrum measured using a synchrotron white beam in combination with a von Hámos geometry-based spectrometer.

## 1. Introduction

Synchrotrons are very bright X-ray sources and provide users with a diverse X-ray spectroscopy instrumentation and methods to study the structure of matter and dynamics of processes on the atomic scale [1]. The spectroscopic studies can be carried out to the highest precision using wavelength-dispersive spectrometry [2] and/or monochromatic synchrotron beam. Improving the energy resolution is, however, at the cost of beam intensity. The synchrotron beam monochromatization reduces the photon flux incident on the sample thousands of times, dissipating the vast majority of the photons produced [3]. Hence, certain steps have been taken to profit from the high energy resolution of wavelength-dispersive spectrometry together with the high brightness of polychromatic synchrotron beam, however, mainly in non-resonant X-ray emission spectroscopy (XES) with a pink beam [4–6].

Recently, different laboratory-based X-ray spectrometers have been developed for high resolution X-ray absorption spectroscopy (XAS) and X-ray emission spectroscopy (XES) measurements with an X-ray tube as the photon source [7–11]. Profiting by the recent advances in laboratory-based X-ray spectrometry, we have undertaken activity to equip the PolyX beamline of SOLARIS [12,13] with white/broadband beam-based high energy resolution XAS capability by means of a von Hámos X-ray spectrometer. PolyX, a compact bending magnet beamline under construction at National Synchrotron Radiation Centre SOLARIS, will be able to deliver polychromatic and monochromatic beams under one of three modes available and the X-ray beam will be focused by capillary optics. The PolyX beamline has already undergone a series of test measurements with the white beam and in this work we report on a von Hámos spectrometer commissioned during a short beamtime available in September 2022. The spectrometer was used to measure Fe K-edge X-ray absorption spectrum and, aware of other beamlines specialized in polychromatic beams [14–16], we believe it is the first time to irradiate a von Hámos spectrometer with a white beam.

The presented von Hámos X-ray spectrometer is a very flexible and versatile tool. At the PolyX beamline it will be used for fast high energy resolution XAS measurements with a white (no monochromator) or broadband (with double multilayer monochromator, DMM) beam, also in multimodal measurements in parallel with microimaging scans [12, 13]. At the beamline a double crystal monochromator (DCM) will be also available, enabling XAS studies with a highly monochromatic beam. As standard DCM-based XAS requires scanning across a given absorption edge to record a XAS spectrum, a white/broadband beam-based XAS with the presented spectrometer is expected to be faster. The spectrometer will be also very useful in cases where a white/broadband

* Corresponding author.
*E-mail address:* wojciech.blachucki@ifj.edu.pl (W. Błachucki).

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beam will be preferential, e.g. to increase the brightness for micro-maging scans, in which case the spectrometer will enable parallel high energy resolution XAS for each scan point.

At the current stage the spectrometer does not allow the extremely fast XAS available at the dedicated time-resolved XAS beamlines [14, 17] but performance improvements are planned. The time resolution of X-ray spectra collection is currently targeted at the level of the exposure time of a single point in the microimaging measurement, namely a tenth or a hundredth of a second. The von Hámos geometry [18], similar to the energy dispersive XAS setup [14], guarantees stable energy scale and focal spot position since it does not require any moving components during acquisition. In the von Hámos geometry, however, the polychromator crystal is placed downstream of the sample which is fully compatible with the multimodal equipment of the compact PolyX beamline [12,13]. The spectrometer will be used for element-specific determination of the sample chemical condition (oxidation state and local electronic structure at the site of atoms of interest) and will find application in biomedical sciences, material science, physics and chemistry, environmental science as well as in cultural heritage science.

## 2. Experimental

PolyX is a compact beamline under development at SOLARIS to be available for users in 2023 [12,13]. PolyX is specialized in X-ray imaging and X-ray spectroscopic methods at energies 4 keV–15 keV with 2–200 μm-spot size achieved with capillary optics. The beamline will provide three different modes of operation:

- white beam mode without any optical elements,
- "high flux" mode with a double multilayer monochromator (bandwidth of 1-2 %),
- "high resolution" mode with a double Si(111) crystal monochromator.

The present work was performed with the white beam filtered using a 10 μm aluminum absorber. The simulated spectrum of the filtered white beam used in the measurements is shown in Fig. 1. The synchrotron beam was focused with a cutted polycapillary optics from Helmut Fischer (symbol 177mkl30-1). The focused beam divergence was about 5.4 ° and at the output focal distance of 25 mm the spot size was about 210 μm for the photon energy of 7112 eV (the Fe K-edge binding energy [19]).

The Fe K-edge X-ray absorption spectrum was measured for a 5 μm metallic iron foil (purity: 99.85%, supplier: Goodfellow) using a wavelength-dispersive setup arranged in the von Hámos geometry [18], the setup outline is presented in Fig. 2. In this setup, the polychromatic and divergent X-ray beam is pointed at a cylindrically bent crystal which leads to the beam diffraction through the Bragg's law. The diffraction has two phenomenological effects on the beam: reflection and sorting of the beam with respect to the photon energy and incidence angle. In result, the diffracted beam propagates away from the crystal and is organized such that photons of different energies propagate along different directions. The diffracted beam reaches a position-sensitive detector (PSD) oriented along the axis of dispersion and fixed at the distance from crystal equal to the crystal radius of curvature, i.e. in the crystal focal plane. As different points of the PSD detector detect photons propagating along different directions (thus having different energies), the count rate registered along the detector is the spectrum of the analyzed beam. Noteworthy is the difference of the focusing direction in the von Hámos geometry-based XAS setups [18] and in energy dispersive XAS setups [14], defined by the crystal curvature. In the von Hámos geometry the excitation beam is focused in the non-dispersive plane and the sample is placed before the crystal in the beam focal spot, as shown in Fig. 2. In energy dispersive XAS geometry, the excitation beam is focused in the dispersive plane and the sample is between crystal and detector in the beam focal spot imaged by the crystal. A von Hámos spectrometer does not require

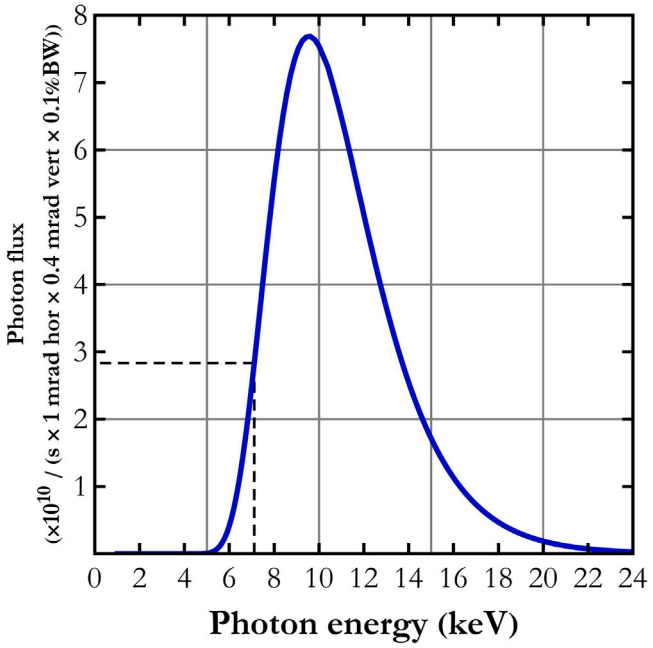


**Fig. 1.** The PolyX beamline spectrum calculated for 1 mrad (H) × 0.4 mrad (V) aperture, storage ring energy of 1.5 GeV and the electron current of 400 mA. The calculation takes into account absorption in the following: 250 μm berylium window, 10 μm aluminum attenuator, 120 cm air path, 300 μm kapton foil and 400 μm chemical vapor deposition (CVD) diamond. The dashed line marks the photon flux available within the domain of the Fe K-edge binding energy (7112 eV [19]).

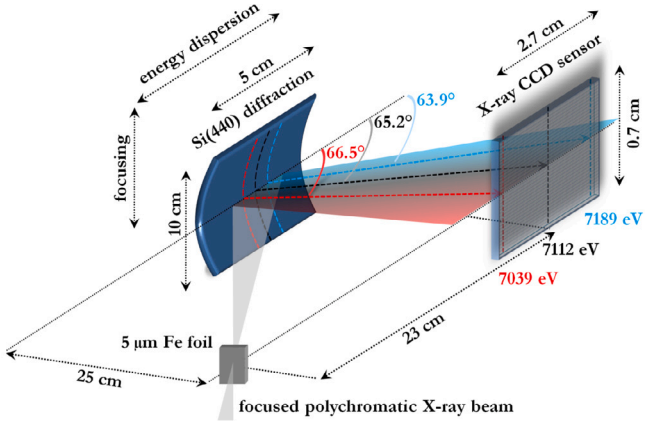


**Fig. 2.** Outline of the X-ray spectrometer used (figure not to scale). The Bragg angles and distances were calculated for the central energy of 7112 eV being the Fe K-edge binding energy [19]. The energy span of 120 eV in the spectra shown in this work is narrower than the total energy range of 150 eV covered by the detector. It is caused by a shade cast by the device case on one of the detector ends which was removed in the data analysis.

any scanning or moving components characteristic of Johann [20] and Johansson [21] geometries and the measurement is accomplished with a static setup in single shot within an energy window from tens to over 200 eV. The setup can be aligned manually, although automation with stepper motors is possible, too.

The spectrometer used in this work was composed of a segmented-type cylindrically bent Si(110) crystal of 25 cm radius of curvature [22] and Andor Newton DO920P camera equipped with 250 μm-thick Be window. The crystal was used at the diffraction order of 4. The camera heart was a front-illuminated charge-coupled device (CCD) of 1024 × 256 26 μm-sized pixels. The sensor was sealed under high vacuum which allowed to safely cool it down to −40 °C by means

of the built-in thermoelectric cooler. The readout rate was set to the highest available 3 MHz and exposure time per frame was set to quite small value of 0.05 s to ensure single photon count per pixel at the large photon flux resulting from the white beam. The setup was operated in air at room conditions. The relative energy resolution was at the level of about $1 \times 10^{-4}$ to $2 \times 10^{-4}$. The setup itself was developed previously in a laboratory with an X-ray tube as the source of photons for routine X-ray spectroscopy measurements and methodology advancements [11,23–28]. Thanks to the simplicity, compactness and modularity guaranteed by the von Hámos geometry, the spectrometer could be easily transported and adapted to the PolyX beamline.

## 3. Data analysis and results

The measured spectra of the incident beam $I_0$ (measured without the sample in the beam) and the beam transmitted through a 5 μm metallic iron foil $I_1$ (with the sample moved into the beam) are shown in Fig. 3(a). The measurement of $I_0$ was done over a sequence of 1000 CCD frames and $I_1$ was measured over different numbers of frames: 1, 10, 100 and 1000. The spectra were obtained through conversion of the raw CCD images from analog-to-digital (ADU) units to photon counts assuming a narrow discrimination ADU window allowing only photons of energy close to the Fe K-edge binding energy. The converted images were summed, projected onto the dispersive axis (horizontal edge of the sensor) and normalized to the measurement time — product of the number of frames and the acquisition time of one frame equal to 0.1102 s (comprises 0.05 s-exposure, readout, transfer). The numbers of frames: 1, 10, 100 and 1000 correspond to the total measurement times (0.05 s-exposure, readout, transfer) of: 0.1 s, 1.1 s, 11.0 s and 110.2 s, respectively. The average registered spectral intensity was $785 \pm 55$ $(\mathrm{s} \times \mathrm{eV})^{-1}$ and, as shown, good counting statistics was obtained within as few as 11 s. Worth noting is also the central position of the $I_0$ maximum resulting from the very precise alignment of the spectrometer achieved manually with merely a ruler and a compact laser pointer. The absorbance $A$ for Fe K-edge X-ray absorption spectra was calculated from the normalized $I_0$ and $I_1$ data using the relation $A = -\log_{10}(I_1/I_0)$, the obtained spectra are presented in Fig. 3(b). The obtainment of an X-ray absorption spectrum requires both $I_0$ and $I_1$ data, however, for a given absorption edge of interest $I_0$ is typically measured once and the speed of XAS spectrum acquisition is limited only by the $I_1$ spectrum counting speed. As shown, the rising edge was well resolved within already seconds of acquisition and the resolution of X-ray absorption near edge structure (XANES) features required as few as 11 s. The Fe K XANES features arise from electron photoexcitations from the core K (1s) energy level to the valence levels $N_{2,3}$ (4p) and the unfilled $M_{4,5}$ (3p) and to the continuum of states. Fig. 3(c) presents the measured Fe K-edge X-ray absorption spectrum juxtaposed with one measured previously at a synchrotron [29]. The reference spectrum was measured in standard transmission mode using ionization chambers filled with nitrogen at 1 atmosphere pressure and with the synchrotron beam monochromatized by a double crystal monochromator equipped with Ge(220) crystals. As shown, the same resolution of XAS measurement is obtained in the present work with the combination of a white beam and the presented X-ray spectrometer. The reference spectrum was used to calibrate our raw absorption spectrum assuming a linear pixel-to-energy conversion function.

## 4. Conclusions

The commissioning of an originally laboratory von Hámos spectrometer with the PolyX white beam required most of all the tuning of CCD camera to a very large readout speed. The short beamtime available was enough to install the spectrometer, tune the detector and measure good quality Fe K-edge X-ray absorption spectrum. The registered count rate was several hundreds times larger than that achieved in laboratory with the XOS X-Beam Superflux PF X-ray tube operated at 50 kV and 1 mA [11,23]. The white beam required considerable reduction of a single frame exposure time which increased the camera dead time to 55 %. Although there are ways to further optimize the

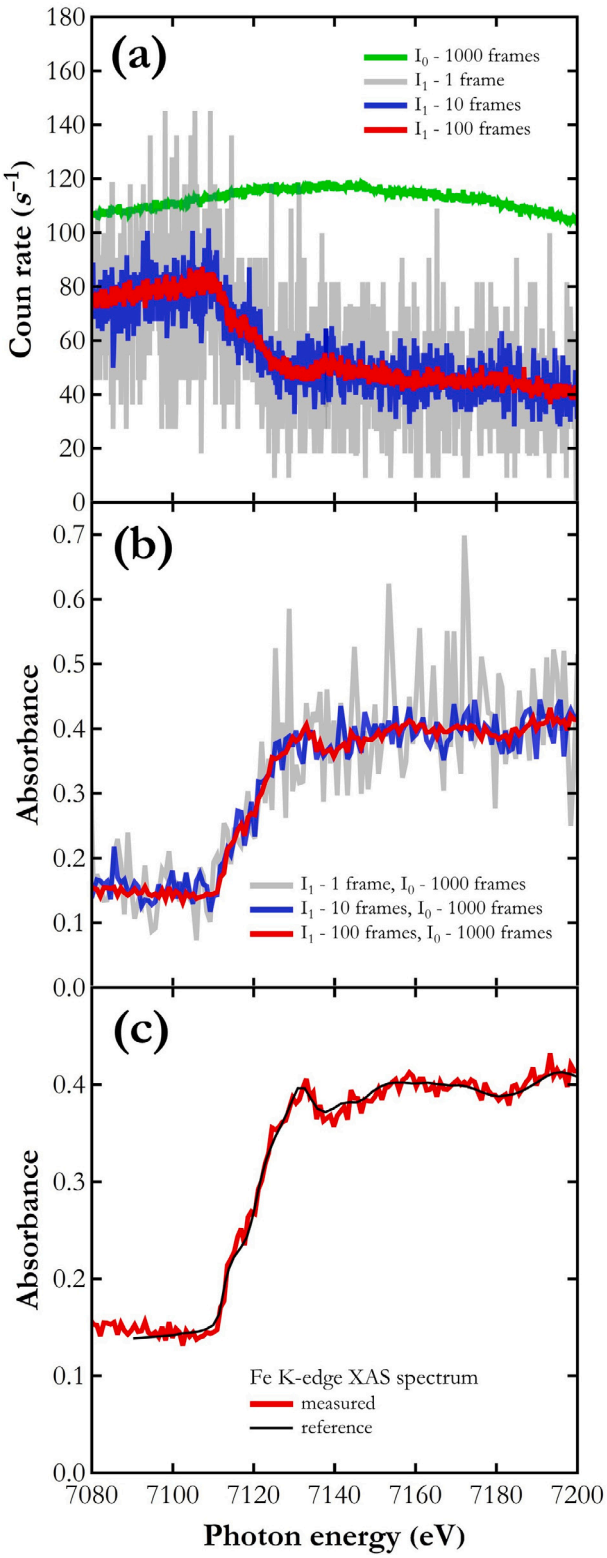


**Fig. 3.** (a) The incident beam spectrum $I_0$ and transmitted beam spectra $I_1$ measured for different numbers of frames. The numbers of frames: 1, 10, 100 and 1000 correspond to the total measurement times (0.05 s-exposure, readout, transfer) of: 0.1 s, 1.1 s, 11.0 s and 110.2 s, respectively. (b) The absorption spectra calculated from the data in (a). (c) The measured Fe K-edge X-ray absorption spectrum (red line from (b)) overlapped with a reference spectrum from Ref. [29].

camera used, a scientific complementary metal–oxide–semiconductor (sCMOS) sensor, a hybrid pixel detector or a strip detector would allow much faster spectra acquisition. The advancements on the part of the PSD detector are desired also in the view of the fact that about 20 times increase in photon flux is expected after installing a He pipe and removing the temporary 400 μm CVD diamond window, to come in 2023.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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