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Abstract. A new undulator beamline (P22) for hard X-ray photoelectron spectroscopy (HAXPES) was built at PETRA III (DESY, Hamburg) to meet the increasing demand for HAXPES-based techniques. It provides four special instruments for high-resolution studies of the electronic and chemical structure of functional nano-materials and catalytic interfaces, with a focus on measurements under operando and/or ambient conditions: (i) a versatile solid-state spectroscopy setup with optional wide-angle lens and in-situ electrical characterization, (ii) a HAXPEEM instrument for sub-µm spectro-microscopy applications, (iii) an ambient pressure system (> 1 bar) for operando studies of catalytic reactions and (iv) a time-of-flight spectrometer as a full-field k-microscope for measurements of the 4D spectral function $\rho(E_B,k)$. The X-ray optics were designed to deliver high brightness photon flux within the HAXPES energy range 2.4 – 15 keV. An LN_2-cooled double-crystal monochromator with interchangeable pairs of Si(111) and (311) crystals is optionally combined with a double channel-cut post-monochromator to generate X-rays with variable energy bandpass adapted to the needs of the experiment. Additionally, the beam polarization can be varied using a diamond phase plate integrated into the beamline. Adaptive beam focusing is realized by Be compound refractive lenses and/or horizontally deflecting mirrors down to a spot size of ~20x17 μm^2 with a flux of up to $1.1x10^{13}$ ph/s (for Si(111) at 6 keV).

INTRODUCTION

The application of photoelectron spectroscopy using hard X-rays (HAXPES) is becoming increasingly popular at synchrotron sources as a powerful spectroscopic tool, because probing depths of several tens of nanometers are achieved routinely. Hence, the established strengths of conventional photoelectron spectroscopy for electronic and chemical structure investigations can be applied to real (functional) materials. Advances in the development of dedicated experimental setups combined with the X-ray beam parameters provided by state-of-the-art undulator beamlines, such as brilliance, energy tunability, polarization, time-structure etc. open up a wealth of opportunities for very specific and unique applications of HAXPES in materials science [1-2].

At DESY (Hamburg, Germany), high-energy photoelectron spectroscopy activities date back to the mid 1990´s. Work started at DORIS III (BW2 wiggler) [3] which was continued at the undulator source (P09) sharing the beamline with other techniques [4, 5]. Recently, a new X-ray undulator beamline (P22) dedicated to HAXPES techniques has been built as part of a major facility extension at PETRA III [6]. The design of the beamline was specially adapted to the needs of the growing HAXPES user community [7]. A unique selection of specialized experimental end stations, all making use of photoelectrons excited by hard X-rays is hosted at P22. The main instrument is used for "classic" HAXPES techniques with an optional wide-angle lens as well as sample cooling and *in-situ* voltage biasing for *operando* studies of functional materials. A separate instrument

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provides full-field, energy-filtered electron microscopy in the hard X-ray regime (HAXPEEM). Another specialized setup is targeting high-pressure HAXPES applications beyond 1 bar (POLARIS). In addition, a time-of-flight spectrometer for *k*-space microscopy will be tested and commissioned for experiments in July 2018. All these instruments are implemented and operated in strong collaboration with external user groups and as such reflect the wide range of scientific fields currently covered by HAXPES. User mode operation of the beamline will begin in September 2018.

HAXPES BEAMLINE P22

Beamline optics

The photon source is a 2m long U33 spectroscopy undulator (k_{max}=2.7) [8]. It shares a 5m high-β straight section (β_x=20m) with the insertion device of the neighboring beamline (P23 nano-diffraction) at a canting angle of 20 mrad. The undulator covers the main HAXPES working range from 2.4 keV to 15 keV on the 1st and 3rd harmonic with a cross-over around 8 keV (Fig. 1, inset). The source brilliance exceeds 10^{20} ph / (s · mrad^2 · mm^2 · 0.1%BW) at 6 keV with a source size of 140 · 6 µm^2 (σ rms). During the initial beamline commissioning, a flux of ~1.1x10^{13} ph/s (Si (111) at 6 keV) was measured in the focused beam (20· 16 µm^2) at 84m from the source.

Figure 1 shows a rendered image of the beamline optics, control and experimental hutches. The frontend components in the ring tunnel are not shown [9]. The primary high-heat load, LN_2-cooled double-crystal monochromator (DCM) [10] is located in the optics hutch and comprises two remotely interchangeable pairs of Si(111) and (311) crystals. For higher energy resolution requirements, a double channel-cut (4-bounce, zero offset) post-monochromator (PM) employing a Si(111) and a Si(220) channel-cut crystal pair can be remotely added. The PM is an in-house design consisting of two towers that hold a Huber goniometer circle and independent height and lateral translations. Fine tuning around the reflection condition is facilitated by a flex pivot and a piezo actuator [11]. All motions are monitored in vacuum with high resolution encoders. Combinations of DCM and PM allow working with an energy bandpass below 200 meV in the entire HAXPES working range.

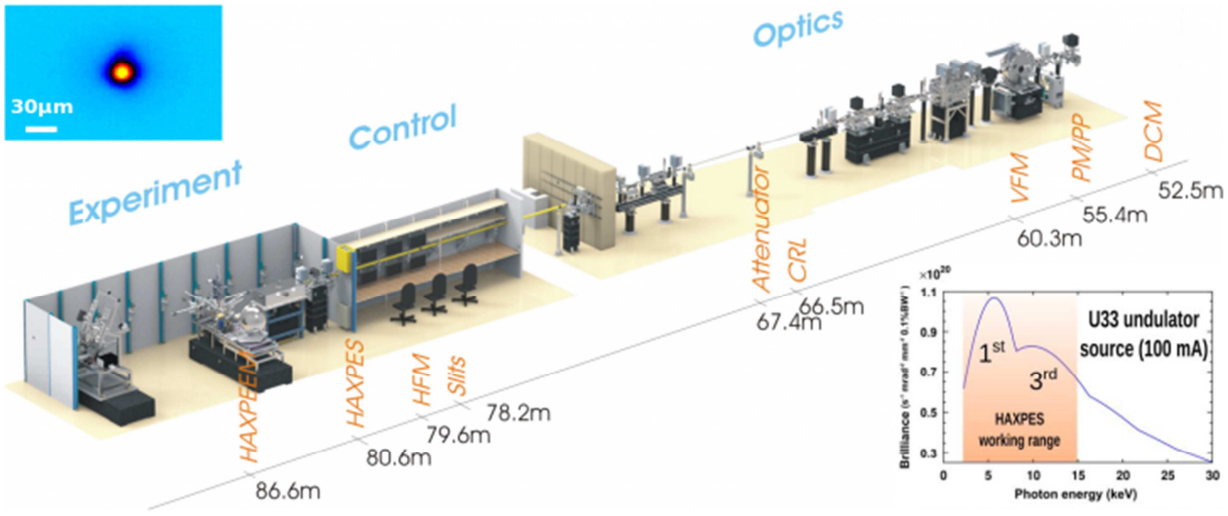

Figure 1 Rendered image of the P22 beamline. The beam enters the optics hutch from the right. The key optical components and their distances from the undulator are indicated. The experimental hutch to the left offers space for 2 in-line experimental stations; shown here are the HAXPES and the HAXPEEM stations. The inset on the top left shows the bean focused in the experimental hutch to a size of 20·16 µm^2. The inset on the bottom right shows the U33 undulator brilliance as a function of photon energy. The HAXPES working range is highlighted for the 1st and 3rd harmonic.

In addition, a stage with diamond phase plates (PP) is implemented downstream inside the PM vacuum vessel. In the hard X-ray regime, the in-plane polarization of a linear undulator beam can be conveniently varied utilizing the phase shift produced by diamond PP in the vicinity of a lattice reflection. The plate thickness typically amounts to 1-1.5 times the X-ray attenuation length, i.e. the beam transmission in Laue geometry is about 20-30%, however yielding a highly polarized photon flux. The device provides variable beam polarization [12], i.e. circular left/right and linear vertical, which can be used to study magnetic properties of buried layers [13] or to separate orbital contributions to the density of states in the valence band [14].

For optimum beam stability at the sample position, focusing is realized by horizontally deflecting mirrors. Each mirror has 2 stripes of reflective coating. A coating with B_4C, was chosen for highest reflectivity up to an energy of 10.5 keV (at 2.9 mrad glancing angle), and Pd for occasional work at higher energies.

A combination of a 1 m long cylindrical and a plane Si mirror placed in the optics hutch is used for vertical focusing (VFM). The mirrors are mounted in individual vacuum vessels based on an established DESY mirror chamber design and share a common granite base. The sagittal cylinder radius of 89.6 mm is expected to yield a ~10 µm vertical focus at 2.9 mrad glancing angle as shown by ray tracing calculations, only limited finally by residual slope errors. Deliberate defocusing can be easily facilitated by small offsets in the glancing angle. First tests at the beam line during commissioning confirmed a vertical focus size of ~16µm (FWHM) at 3.0m from the HFM without any limiting upstream aperture.

The bendable plane-elliptical mirror (HFM) [10] can be adjusted for optimal horizontal focusing at all experimental positions. The mirror is located 2 m before the first measurement position in the experimental hutch at 80.6m from the source. The insulating zerodur substrate of the mirror allows measuring the photoelectric drain current from the reflective stripes which is used for incident beam intensity monitoring and mirror alignment. First tests of the horizontal focusing demonstrate a very stable horizontal focus of ~20µm (FWHM) 3m from the HFM.

Alternatively, for intermediate focal sizes an in-house designed transfocator with 1D Be compound refractive lenses (CRLs) is available. The compact UHV compatible device is placed downstream in the optics hutch and offers moderate horizontal focusing down to about 80 µm (FWHM), depending on photon energy [15].

The experimental hutch provides ample space for two large end stations installed in-line. The experimental positions are connected to two gas cabinets which meet the individual experimental needs. Several instruments share this space in order to take advantage of the favorable conditions P22 offers for HAXPES. All instruments are designed such that they may be mutually exchanged with moderate effort and in short turnover times. Additional floor space including media supplies is provided outside, next to the hutch, for off-line instrument operation, maintenance and testing. In the following, we will briefly introduce the different instruments operated and/or being implemented at the beamline.

HAXPES spectroscopy instrument

The well-established HAXPES setup for solid state spectroscopy which has been in operation at PETRA III beamline P09 for several years [4,5] has been relocated to P22 (Fig. 2, left). A key feature of this instrument are HAXPES studies of device relevant multi-layer structures under working conditions by *in-situ* biasing and accurate electric characterization of electronic devices [16, 17].

A Phoibos 225HV analyzer [18] with its optical axis oriented along the horizontal X-ray polarization direction is used for electron detection up to 10.5 keV kinetic energy. The analyzer uses a delay-line electron detector with a high dynamic range of 10^6 [19]. The standard analyzer lens may be exchanged with a $\pm 30^o$ wide-angle acceptance lens for increased transmission and/or angle-dependent XPS studies.

Analyzer and analysis chamber are mounted on a motorized platform, which can be aligned vertically and horizontally normal to the incoming X-ray beam with very high accuracy (reproducibility <2 µm in the vertical direction). Samples are introduced to the analysis chamber via a quick access load lock which is attached to a sample storage carousel. From there, transfer is facilitated by a long transfer arm to the liquid Helium cooled 5-axis manipulator [20]. The manipulator head offers 4 spring-loaded electrical contacts which can be used for *in-situ* sample biasing, electrical characterization or to contact small mobile sample environments, e.g. sample heaters. Electrical characterization of functional multilayer stacks is facilitated by a precision source measure unit connected to the contacts of the manipulator via a switching matrix. This allows for versatile and fully automated measurement routines.

A simple stage for *in-situ* re-magnetization of magnetic samples as well as local gas dosing nozzles e.g. for oxidation are incorporated into the experimental chamber. An attached preparation chamber is set up for sample treatment (heating, cleaving, ion sputtering) and/or surface characterization. X-rays scattered from the sample surface can be recorded by 2D X-ray detectors place downstream of the analysis chamber. This configuration has gained particular interest for multilayer and total reflection X-ray standing wave measurements. The

analysis chamber is additionally equipped with a (Cr, Al) twin-anode X-ray source [21] and a high-energy (1-20 keV) electron gun [22].

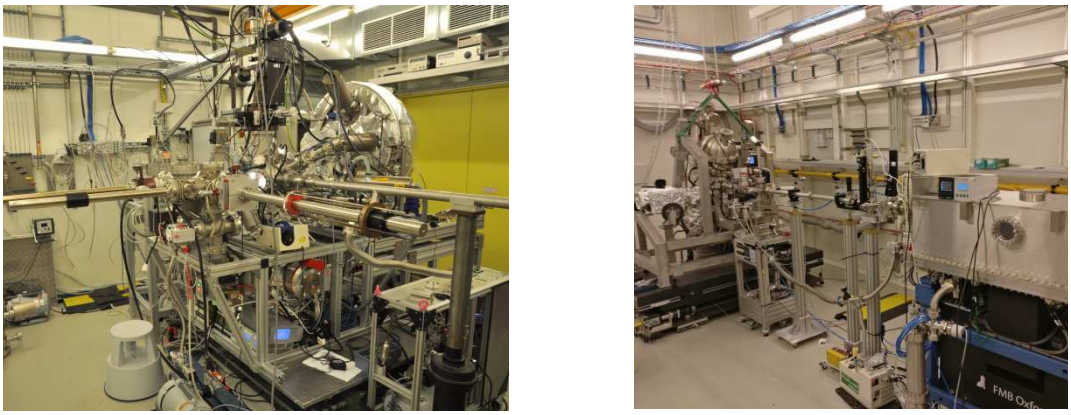

Figure 2: Left: HAXPES setup used for solid state spectroscopy. Right: HAXPEEM instrument in the new experimental hutch of P22.

An essential component for an efficient operation of both the beamline and the HAXPES instrument is a coherent software control of the experiment. Strong efforts have been made to develop an experiment control system which integrates the beamline optics and the HAXPES chamber including the electron spectrometer with emphasis on ease of use as well as flexible automated operation, sample handling and beam alignment. It is completely written in Python making use of the lab-wide TANGO device abstraction layer. The user interface comprises several widgets for standard tasks such as motor movements, beamline component control and data acquisition of the analyzer. The latter is remotely controlled by a custom designed command interface via TCP/IP and data visualization and storage is done on the beamline control computer. More complex control tasks can be realized by an integrated, Python based scripting layer which offers virtually unlimited flexibility.

The two in-line positions in the experimental hutch are shared between the HAXPES instrument described above and other special instruments which are temporarily installed and operated in collaboration with external research groups. In the following we give a short overview.

HAXPEEM: Full field imaged spectroscopy of lateral structures

Full-field photoemission microscopy combined with energy filtering in the hard X-ray regime can be used to resolve lateral structures on the sub-µm scale exploiting the advantages of HAXPES for depth sensitivity. A hard X-ray photoelectron emission microscope (HAXPEEM) has been developed and commissioned at beamline P09 a few years ago [23] and is now operated at beamline P22 (Fig. 2, right). It is a modified version of a commercial instrument [24, 25] where an immersion lens objective collects photoelectrons from the sample and forms a first real image. An electrostatic transfer optic retards the electrons and feeds the image into the entrance of a 125 mm hemispherical analyzer operating at constant pass energy. A variable aperture in the analyzer exit plane defines the transmitted energy bandwidth. After passing a second, symmetric hemisphere to cancel image aberrations, the image is recovered and a two-stage projector optics delivers a final total magnification of up to ~1500x. The photoelectron image is intensified by a double MCP stack to produce a visible image on a fluorescence screen. All potentials have been scaled up to allow for kinetic energies up to 10 keV. The instrument accepts flat, conductive samples of size 10x10 mm^2 and 1 mm thickness and has a storage stage for up to 4 samples for quick exchange. Following proof-of-principle experiments, the initial performance yielded a spatial resolution of 670 nm for imaging buried structures down to a depth of more than 10 nm below the surface, with an energy resolution of 1eV [23].

Photoelectron *k*-microscope

Photoelectron *k*-microscopy is a new approach to the mapping of the valence electronic structure. Full-field k-imaging with time-of-flight (ToF) energy recording is the maximum parallelization possible for data acquisition. In the soft X-ray range, a benchmark in recording efficiency was recently reached by taking 4D (E_B,k) data arrays in the complete 3D Brillouin zone (BZ) and several eV energy width (beamline P04 at

PETRA III) [26]. In the HAXPES energy range *k*-microscopy can be expected to provide valuable insights to the valence electronic structure of *buried* layers and interfaces. A high-energy prototype of such an instrument combining a PEEM immersion lens (up to 20 $Å^{-1}$ field of view) with a ToF drift tube for energy filtering will be available at P22. This instrument will take advantage of the unique time structure of the reduced bunch filling modes of PETRA III offering rather large bunch separation of up to 192 ns (with 40 bunches) owing to the large circumference of the storage ring (2.3 km). First test experiments are scheduled in July 2018. In general, cathode-lens type microscopes are very sensitive to space-charge induced energy shifts and therefore suitable strategies of correction and suppressions are currently being developed [27].

POLARIS: HAXPES at high-pressures

The study of *operando* catalytic transformations at interfaces between solids and liquids or gases play a crucial role in answering the most pressing questions in energy storage and conversion. The POLARIS setup at P22 (Fig. 3) is dedicated to investigate catalytic processes on surfaces and interfaces under realistic conditions, i.e. pressures beyond 1 bar and temperatures up to 450°C during the measurement [28].

The system is based on a commercial ambient pressure instrument [29] which combines a high-resolution electron analyzer with a differentially pumped pre-lens. This has been substantially modified by a novel pressure-cell design that integrates the front cone of the electron analyzer which allows windowless operation at pressures beyond 1 bar. The distance between sample and analyzer cone is only a few µm requiring well-focused X-rays and highly accurate instrument and sample alignment. Two high-precision manipulators position the sample with sub-µm resolution with respect to the front cone and the incoming X-ray beam. Reaction products can be followed with a residual gas analyzer in the differential pumping section of the electron analyzer. Recently, HAXPES measurements at 3.7 keV could be achieved at pressures up to 2 bars using He (Fig. 3, right) and temperatures up to 450°C allowing for the study of catalytic reactions under real conditions while the catalytic surface is turning over the reaction. The results push the pressure limits of ambient pressure photoelectron spectroscopy beyond the previous record of 1 bar. [30]

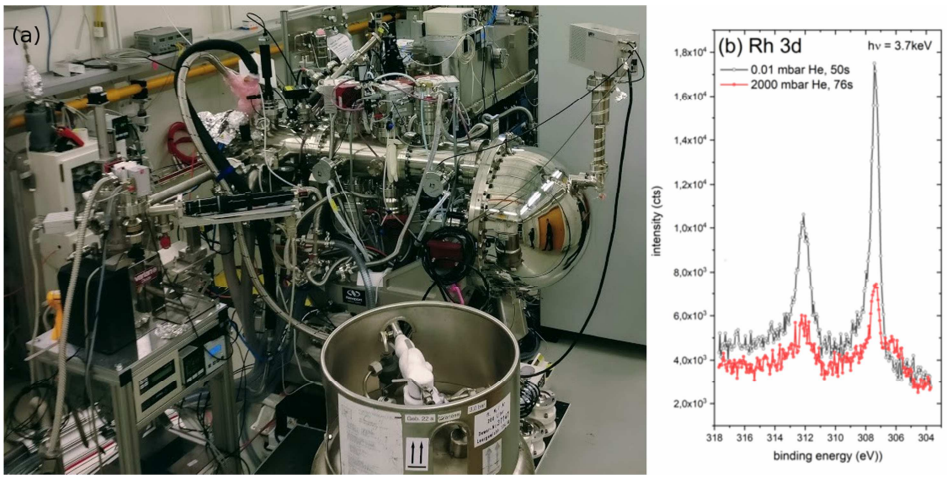

Figure 3 Left: POLARIS ambient-pressure instrument in operation at P22. Right: Core level HAXPES spectra of Rh 3d recorded at 0.01 and 2000 mbar of He. The spectra were taken during the initial commissioning with short accumulation times only.

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