EMIL: THE ENERGY MATERIALS IN-SITU LABORATORY BERLIN – A NOVEL CHARACTERIZATION FACILITY FOR PHOTOVOLTAIC AND ENERGY MATERIALS

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ABSTRACT: A knowledge-based approach towards developing a new generation of solar energy conversion devices requires a fast and direct feedback between sophisticated analytics and state-of-the-art processing/test facilities for all relevant material classes. A promising approach is the coupling of synchrotron-based X-ray characterization techniques, providing the unique possibility to map the electronic and chemical structure of thin layers and interface regions – with relevant in-system/in-situ sample preparation or in-operando analysis capabilities in one dedicated laboratory. EMIL, the Energy Materials In-situ Laboratory Berlin, is a unique facility at the BESSY II synchrotron light source. EMIL will be dedicated to the in-system, in-situ, and in-operando X-ray analysis of materials and devices for energy conversion and energy storage technologies including photovoltaic applications and photo-electrochemical processes. EMIL comprises up to five experimental end-stations, three of them can access X-rays in an energy range of 80 eV – 10 keV. For example, one key setup of EMIL combines a suite of advanced spectroscopic characterization tools with industry-relevant deposition facilities in one integrated ultra-high vacuum system. These deposition tools allow the growth of PV devices based on silicon, compound semiconductors, hybrid heterojunctions, and organo-metal halide perovskites on up to 6'' sized substrates.
EMIL will serve as a research platform for national and international collaboration in the field of photovoltaic/photocatalytic energy conversion and beyond. In this paper, we will provide an overview of the analytic and material capabilities at EMIL.
Keywords: Buried contacts, Interfaces, Characterization, Deposition, PV Materials, Spectroscopy, Thin Film

1 INTRODUCTION

One of the main challenges for today's global society is a reliable, cost-effective and environmentally-friendly supply of energy. Renewable energies will likely carry the major load within a future sustainable energy system and photovoltaics (PV) as well as photo(electro)chemical fuel production will play important roles. Technology development and mass production have pulled down the costs of PV during the past decades. However, in order to accommodate the necessary economic constraints to implement PV on a global scale, substantial cost reductions are further needed and the integration of PV in a supply system tackling the fluctuating availability of solar radiation is a must. This additionally calls for economically suitable solutions for solar fuel production and energy storage (batteries, hydrogen storage etc.).

To achieve these ambitious goals on the required time horizon, a knowledge-based approach to materials research will become necessary with a fast and direct feedback between sophisticated analytics and state-of-the-art deposition systems which are capable to process complete devices. In particular, it requires the deliberate design of materials and interfaces. Thus, the understanding of the electronic, optical, chemical, and structural properties of materials involved becomes very important. The relevant active interfaces that determine device operation are often not directly accessible for spectroscopy since they are buried under a contact material (e.g. in a solar cell heterojunction) or under a thin liquid film (in a photoelectrochemical device). Such buried interfaces usually become accessible by removing the respective layers. However, through this procedure, changes to the interface properties will be induced,

Figure 1: The EMIL infrastructure (left) at the BESSY II storage ring (left, courtesy of hammeskrause bda). For more details see http://www.helmholtz-berlin.de/projects/emil/.

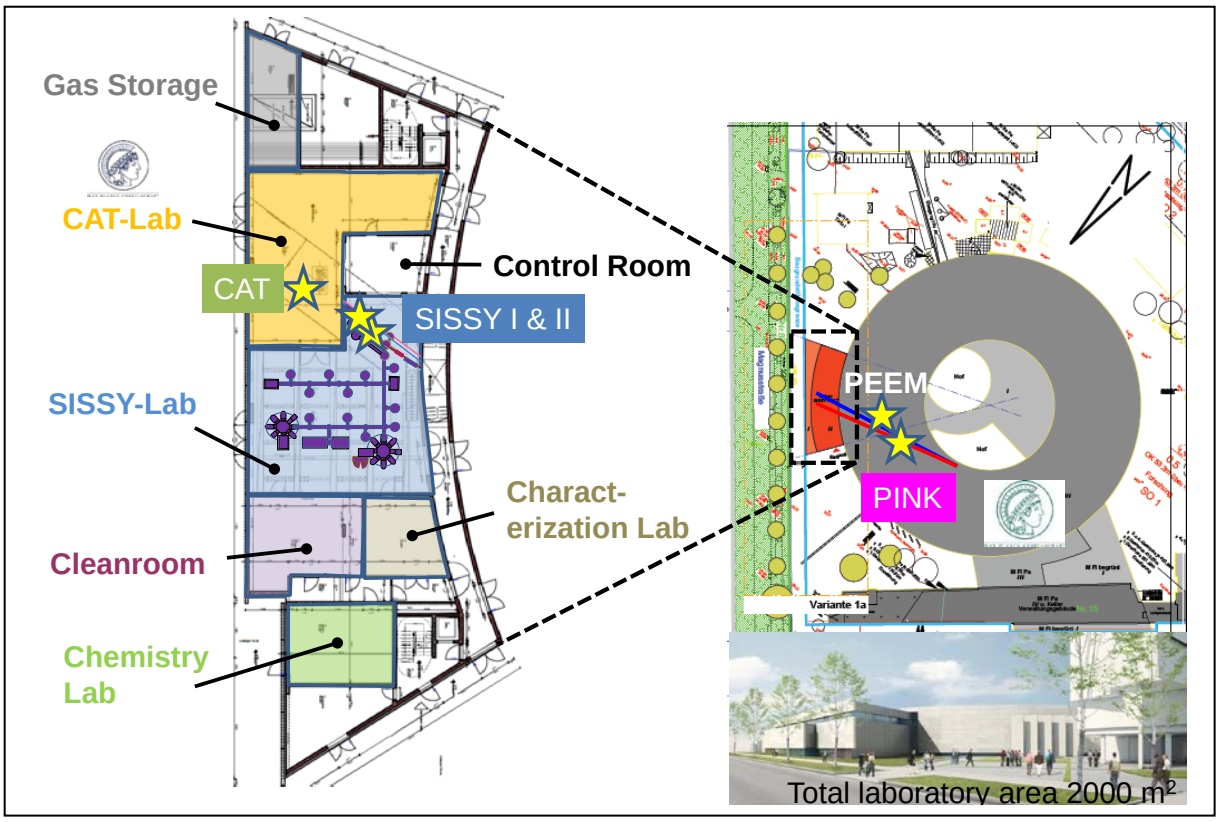

Figure 2: Sketch of the laboratory layout of EMIL and its location at the BESSY II synchrotron light source.

leading to strong deviations with respect to the relevant interface properties (e.g. induced by the missing/artificially altered junction partner or due to contamination).

An approach to tackle this problem is the coupling of synchrotron-based X-ray characterization techniques with a variety of information depths – such as soft and hard X-ray photoelectron spectroscopy and -microscopy (XPS/XPEEM), as well as X-ray absorption (XAS) and emission/fluorescence (XES/XRF) spectroscopy, with relevant deposition, post-deposition treatment, and test capabilities in one dedicated laboratory infrastructure

Despite the detailed information that can be obtained through synchrotron-based spectroscopy, two main limitations are currently restricting the broad application of synchrotron light sources in energy materials research worldwide: (i) The feedback times between spectroscopy at the synchrotron and performance results for materials and device development are too long to allow for a truly knowledge-based design due to limited access to synchrotron sources. (ii) Surface contamination as often found on *real-world* samples sometimes hinder or even prevent an unambiguous data interpretation. (iii) The limited X-ray energy range at most beamlines does not allow a continuous use of soft and tender/hard X-rays as would be mandatory to study surfaces and buried interfaces on the same sample without moving to another experimental setup. (iv) Conventionally, the study of the chemical and electronic structure of surfaces and interfaces of materials and layer stacks by X-ray spectroscopies is performed in an ultra-high vacuum environment. For some energy conversion devices, in particular in the field of (photo)electrochemistry, this represents a significant deviation from the environment they are usually operated in, recently raising skepticism about the relevance of the derived results for the device.

2 EMIL'S APPROACH

The aforementioned scientific challenges require an in-system/in-situ/in-operando X-ray spectroscopy approach in a dedicated laboratory infrastructure ensuring continues access to synchrotron radiation to follow the growth of materials and the formation of functional interfaces as well as the evolution of their properties during successive device processing and operation. EMIL will employ multi-chamber small (11 × 11 mm^2) and large-area (6" wafers) thin-film deposition tools across all relevant material classes, which are connected through ultra-high vacuum (UHV) transfer systems to advanced characterization tools complemented by spectroscopic analysis capabilities at ambient and atmospheric pressure to aid the development of real-world materials for PV, (photo)electrochemical, and (photo)catalytic devices. The analytical facilities feature a combination of complementary state-of-the-art X-ray analytical techniques in three end stations at a dedicated beamline delivering high-brilliance photons in the energy range of 80 – 10,000 eV. The availability of such a wide X-ray energy range from soft to hard X-rays will allow studying the physical and chemical properties of buried interfaces due to the easily tunable information depth accessible at the EMIL beamlines. Hard X-ray energies further facilitate the study of liquid/solid interfaces in electrochemical processes such as water splitting.

EMIL's technical specifications are pushing the limits of synchrotron-based X-ray analysis. The EMIL beamlines, to be commissioned in 2016, will provide a photon flux of up to 10^{13} $s^{-1}0.1A^{-1}$ at a resolution $E/\Delta E$ > 10,000. Spot sizes of 45 × 10 μm^2 and 120 x 10 μm^2 for soft and hard X-ray energies, respectively, will be

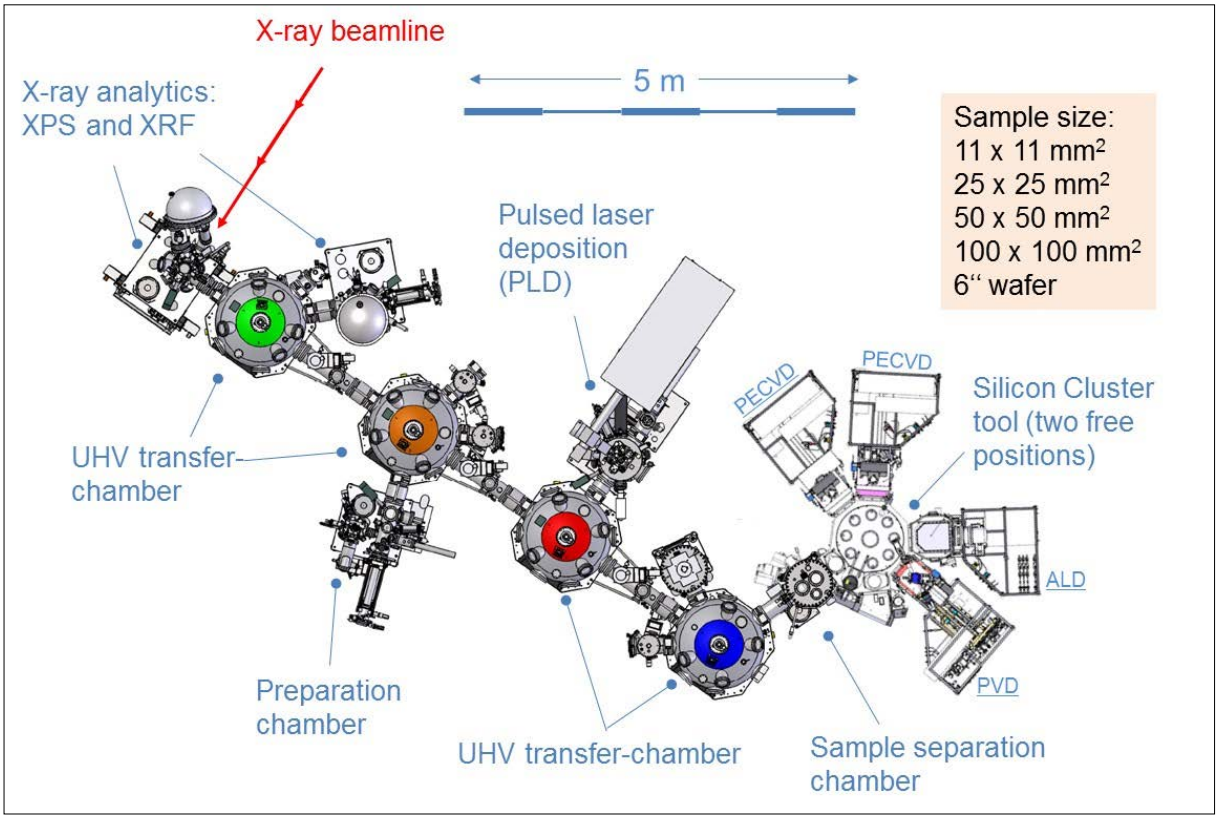

Figure 3: Current layout of the SISSY@EMIL deposition and analyis tool.

realized (see [1]).

2.1 Infrastructure and X-ray characterization tools

The EMIL building is a separate infrastructure with a total floor area of 2000 m^2 that is attached to the BESSY II synchrotron (Fig. 1) with direct access to the dedicated X-ray beamlines. EMIL will consist of two large laboratory infrastructures: SISSY (Solar Energy In-Situ Laboratory at the Synchrotron operated by HZB) and CAT (Catalysis Research for Sustainable Energy Supply operated by Max Planck Society, MPG) (Fig. 2). In particular, SISSY will feature a combination of the afore-mentioned state-of-the-art X-ray analytical techniques, and CAT will focus on fundamental research on catalytic processes under ambient conditions employing near-ambient-pressure hard X-ray photoelectron spectroscopy (HAXPES). Additionally, in the CAT@EMIL laboratory near-ambient pressure HAXPES will be combined with in situ energy-dispersive XRD (ED-XRD). The simultaneous observation of surface and structural bulk properties during compound semiconductor film formation will thus be possible.

In addition, EMIL will have a chemistry lab and a clean room as well as a gas handling system that can supply the SISSY lab with up to 30 different gases (Fig. 2). EMIL will be complemented by two additional end stations located on the experimental floor of BESSY II: a photoelectron emission microscope (PEEM) and a X-ray emission spectroscopy (XES) end-station PINK, capable of operation in a tender x-ray energy range (2-10 KeV) – see Fig. 2.

2.2 Deposition tools

In more detail, SISSY@EMIL has dedicated deposition and preparation facilities across all materials classes. It allows for the preparation of hybrids and novel material combinations as well as combinatorial synthesis approaches. Most of the aforementioned functionality will be hosted in a UHV system for processing and analysis of up to 6” sized substrates which are handled using automated vacuum transfer systems. This minimizes the time needed for sample transfer and thus reducing the probability of contamination, while enabling step-by-step deposition/analysis sequences. In its current planning phase EMIL hosts a silicon deposition cluster tool with up to six deposition chambers that are directly connected to the automated UHV transfer line. The initial layout features two plasma-enhanced chemical vapor deposition (PECVD) chambers, one atomic layer deposition (ALD) chamber and one sputter deposition chamber (see Fig. 3), enables deposition on up to 6” size substrates, i.e., spanning the range from “academic” samples up to fully industry-compatible wafers.

By a pulsed laser deposition tool (PLD, see Fig. 3), EMIL will also enable composition-dependent libraries of the chemical and electronic structure of oxides with complex stoichiometries to be built for stable photoelectrodes for water splitting devices. By combining gradient composition/stoichiometry samples made with PLD with spatially resolved X-ray analysis a much higher sample characterization throughput will be possible.

At CAT@EMIL, promising photoelectrodes will be characterized in-operando by near-ambient pressure HAXPES (in collaboration with MPG). The stability and reactivity of such novel photoactive electrodes will be studied under real conditions, i.e., near-ambient pressure and under potential control. The aim is to in-situ probe the surface/near-surface chemical composition (and how it changes) during operation in the dark and under illumination.

These preparation/test facilities will be extended in

the near future to also include compound semiconductor processing (e.g. chalcopyrites, kesterites). The cluster design is such that is allows for full device preparation without vacuum breaks, by combining a flexible MBE absorber preparation chamber, as well as a sputtering chamber for buffer and window preparation. This tool will be especially geared towards research on compound semiconductor based multijunction solar cells as well as solar fuel generation devices by photoelectrochemistry (PEC). Furthermore, it is envisioned to add preparation capabilities for III-V materials, a glove-box and vacuum-based organic semiconductor processing capabilities for organic PV (OPV), and hybrid concepts (e.g. perovskites) as well as for emerging materials such as graphene, nanocomposites, and hybrid materials for solar spectrum shaping (photonic up- and down-conversion) or materials for (photo)catalysis. Also a characterization cluster with STM, AFM and micro-Raman setups will complement the analytic toolbox of EMIL.

The unique capabilities of EMIL's characterization/ preparation platform for in-system/in-situ and in-operando analysis will enable both synergetic use of the unique synchrotron-based analysis environment and a cross-system approach in device design, explicitly envisioning hybrid organic/inorganic or combined PV/PEC devices.

2.3 Beamtime application and cooperation

EMIL is part of the large-scale user facility BESSY II and therefore will be accessible after commissioning to external users from academia and industry through the BESSY II access platform GATE. There will be several variants of beamtime available to external users depending on the form of collaboration (short term, long term), and the kind of required experiments. In particular, a fast beamtime access for industrial users and for fast test experiments is envisioned.

3 SUMMARY

Up to now, a knowledge-based approach for optimizing solar energy devices is extremely time-consuming as the parameter space for material and device design is very large, and access to synchrotron-based analytics is limited. Due to the high reactivity of the involved materials and interfaces (e.g., for photovoltaic materials) and the very specific environment they are working in (e.g., in aqueous solutions for catalysts), the mandatory *in-system*, *in-situ*, or *in-operando* analysis of film growth or of devices under working conditions is in the core focus of the EMIL infrastructure. EMIL will therefore be a unique facility, enabling entirely new routes of energy materials development. EMIL will serve as a dedicated research platform and will be available for national and international collaboration.

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