Invited Lecture

Accelerator based 3D-structure determination with MeV electrons

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Electron diffraction (ED) is an extremely powerful tool for 3D structure determination of crystalline materials like 2D-layered transition metal dichalcogenides (TMDs), minerals, pharmaceutical compounds, or proteins. By providing 5 to 6 orders of magnitude stronger interaction with matter compared to X-rays, electrons are ideally suited for studying nano- and microcrystals. This strong interaction comes along with a limitation on the maximum sample thickness. For laboratory-based electron microscopes, which are typically operating in the 100 – 300 keV range, this limit is typically about 50 nm for high-Z samples and about 300 nm for low-Z materials. Thus, sample preparation may impose a serious challenge for ED experiments. Utilizing higher electron energies in the MeV range, the maximum sample thickness is increased by a factor of about 2 to 4, which allows sample preparation under an optical microscope.

Here we present first results from 3D structure determinations carried out at DESY's high-energy electron diffraction facility REGAE (RElativistic Gun for Atomic Exploration) in Hamburg, Germany [1]. The radio-frequency based linear accelerator REGAE generates ultrashort electron pulses with energies ranging from 2.5 to 6 MeV. For the investigation of solid samples, the experimental chamber of REGAE is equipped with an ultrahigh vacuum (UHV) compatible single axis goniometer allowing to collect high-quality diffraction data over a large rotation angle (Figure 1, left). At a distance of 5 m downstream from the sample, diffraction images are recorded on a Jungfrau 1M detector allowing the direct detection of electrons. With this setup, a continuous-rotation data set of the sheet silicate muscovite was recorded with 3 MeV electrons. After data reduction, structure solution was straightforward. Dynamical refinement with Jana2020 indicates a sample thickness of about 600 nm and the final R1-value is about 11% [2]. This result highlights the potential of high-energy ED at REGAE. Other samples recently investigated at REGAE include TaS_2, known for its super-lattice structure originating from a charge density wave of the sample (Figure 1, right).

A further advantage of REGAE is the much larger space available around the sample, allowing the installation of more sophisticated experimental setups for sample manipulation and excitation such as environmental cells for room temperature experiments with biological samples or cells for gas-loading experiments of catalysts.

In the near future we aim at extending the structure determination capabilities from the ground state of the samples to the time domain by utilizing the unique femtosecond time resolution available at REGAE. The presentation will summarize the present status and provide an outlook on the planned hardware extensions of REGAE.

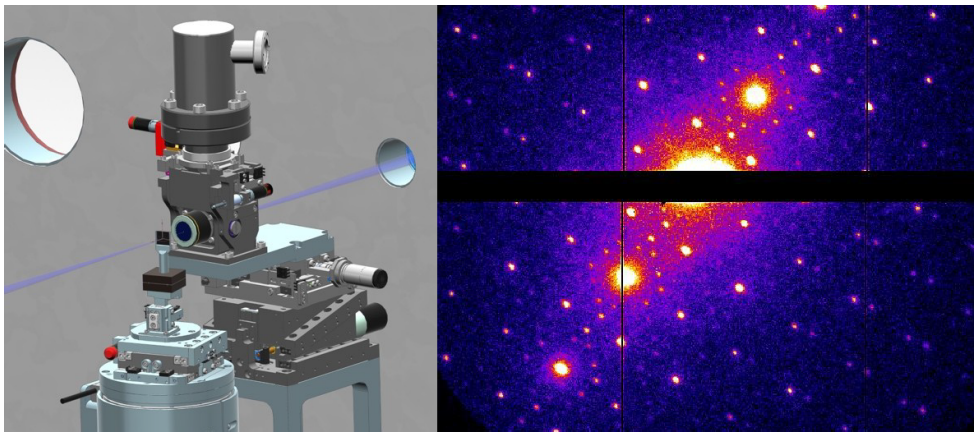

Figure 1. Left: Technical drawing of the UHV-compatible crystallography setup installed in the experiment chamber of REGAE. It consists of an inline sample viewing microscope and high-precision e-Roadrunner goniometer. Right: 3 MeV electron diffraction pattern of a TaS_2 single crystal with super lattice reflections recorded on a direct detection Jungfrau 1M detector.

[1] Manz, S., Casandruc A., Zhong, Y., et al. *(2015). Faraday Discuss.* **177**, 467−491.

[2] Klar, P.B., Krysiak, Y., Xu, H., Steciuk, G., Cho, J., Zou, X., Palatinus, L. (2023). *Nat. Chem.* **15**, 848−855.