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Parallel Acquisition of Real and Reciprocal Space Data in Transmission SEM

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Recent developments of transmission diffraction setups for scanning electron microscopy (SEM) showcase the power of diffraction analysis combined with low energetic electrons [1], [2, 3]. There are several reasons for pushing these developments further. First, SEM instruments are much more widespread and show a simpler set than transmission electron microscopy (TEM). Therefore, the accessibility is significantly higher and also the barrier for usage is reduced. Secondly, the large sample chamber and extended field of view make SEM instruments highly attractive for fast screening of materials and implementation of dedicated *in situ* setups. Moreover, several TEM techniques can be straightforwardly transferred to SEM. Even 4D-STEM, a popular technique in TEM, managed to enter SEM, where the lower acceleration voltage provides much increased contrast and reduced knock-on damage [2]. These advantages are particularly relevant for 2D materials [5].

In 4D-STEM, the electron beam scans over the sample in a 2D pixel array and for each scanning point, the corresponding diffraction pattern is acquired, resulting in a 4D data stack. Each 4D data stack includes an enormous amount of information, which has to be processed by dedicated algorithm and analysis software. Virtual dark-field images are one prominent example of using those data to reconstruct countless images by arbitrary apertures shapes with no physical limitations. More advanced analysis enable strain and orientation mappings and revealing electromagnetic properties [4]. 4D-STEM has increased significantly in popularity over the last few years by recent developments in camera technology offering frame rates up to 80.000 frames per second (fps), which reduces drastically the needed acquisition time. However, even the fastest cameras cannot support important *in situ* experiments, where a continuous monitoring of the ongoing reaction is necessary to elucidate the occurring phenomena. A simple image/video recording with a 1k x 1k pixel size and an acquisition time of 1 fps requires a total frame rate of 1.000.000 fps. No available camera even comes close to such acquisition times.

The lack of such a combined transmission and diffraction imaging setup for *in situ* experiments together with the auspicious perspectives was the driving force for our current development. In this work, we introduce a unique transmission setup in SEM, which enables fast STEM imaging with simultaneous spatially averaged diffraction analysis. Figure 1a-b) illustrates the concept of our setup. It builds on our developed LEND setup [5], which uses a combination of fluorescent screen and in-chamber mounted CMOS camera for acquisition of diffraction patterns. In addition, a secondary STEM detector is installed below the fluorescent screen, which contains a centric hole (0.4 mm / 0.8 mm) through which transmitted (or diffracted) electrons can pass for STEM imaging.

Our concept offers regular STEM imaging with sufficiently high frame rate for dedicated *in situ* experiments combined with acquisition of averaged diffraction patterns to receive the reciprocal space data simultaneously. The parallel acquisition of real and reciprocal space information opens up new possibilities for receiving key information about complex reactions and microscopic mechanism. The utilized STEM detector, as an integrating detector, is faster than any direct detection camera regarding the achievable frame rate, thus facilitating *in situ* experiments. In addition, this setup allows not only STEM imaging and diffraction analysis but also simultaneous detection of other common signals, such as secondary electrons (SE) and back-scattered electrons (BSE). In total, up to four different signals can be simultaneously recorded for detailed analysis. Furthermore, apart from bright-field (BF) also dark-field (DF) imaging can be achieved by moving the fluorescent screen in a position so that a diffracted beam passed through its hole. In Figure 1c-d) the 3D model and the installed setup in the SEM are shown.

The first conducted experiment was the analysis of a thin MoS_2 flake covered with gold (Au) particles/islands formed by solid state dewetting. Figure 2a depicts all four acquired signals (SE, BSE, STEM (BF), diffraction). SE (yellow) offers topographic information; all tiny structures are visible on the surface together with a detailed view on the Au nanoparticles. Elemental contrast is provided by BSE (blue) imaging. Here, it is easy to distinguish between both phases, where MoS_2 reveals a slightly darker contrast compared to the Au particles because of the lower atomic number. The fluorescent screen is positioned with the hole centered around the incident/transmitted electron beam. Therefore, the undiffracted electrons reach the STEM detector and generate a typical BF signal (black) offering information about the inner structure of the sample (dominated by mass-thickness and Bragg contrast). Pronounced bend contours are visible due to the bending of the thin MoS_2 flake. The fourth recorded signal is the diffraction pattern (green), which is an averaged pattern over the whole scanned sample area showing the typical hexagonal pattern of MoS_2. No reflections from the Au particles are visible since the particles/islands are too thick to be transmitted by the low-energetic electrons.

In this work, we want to demonstrate the successful implementation of our presented setup by means of several examples. Furthermore, it is intended to combine the described setup with our *in situ* heating stage [6] to perform unique *in situ* heating experiments together with acquiring all available signals simultaneously (see Figure 2b). The heating stage utilizes DENSsolutions Wildfire Nano-Chips, which enables precise and fast heating and cooling of the specimen. The parallel access to real and reciprocal space during annealing experiments can help to unravel the complex interplay of phenomena during thin film processes, like, e.g., grain coarsening, dewetting and texture evolution during solid-state dewetting of metal films [6, 7].

In the future, an independent and motorized movement of the fluorescent screen (with attached STEM detector) is planned to enable fast and easy switching between bright-field and dark-field imaging modes. Moreover, we want to develop and include intelligent scan strategies to amplify the current capabilities. Besides simple line-by-line scanning, defined scan areas of interesting sample regions can be flexibly chosen and the corresponding recorded diffraction patterns can be assigned to those regions. For instance, if one STEM image (taken with 1 fps) is subdivided into 100 patches which are scanned subsequently the corresponding diffraction patterns of these regions could be collected with a frame rate (of the LEND camera) of 100 fps, which is feasible. Furthermore, methods of artificial intelligence might be useful for recognizing autonomously significant sample areas during STEM imaging, track those areas and adjust the scan strategy in a way that diffraction patterns from these areas are collected parallel to STEM imaging [8].

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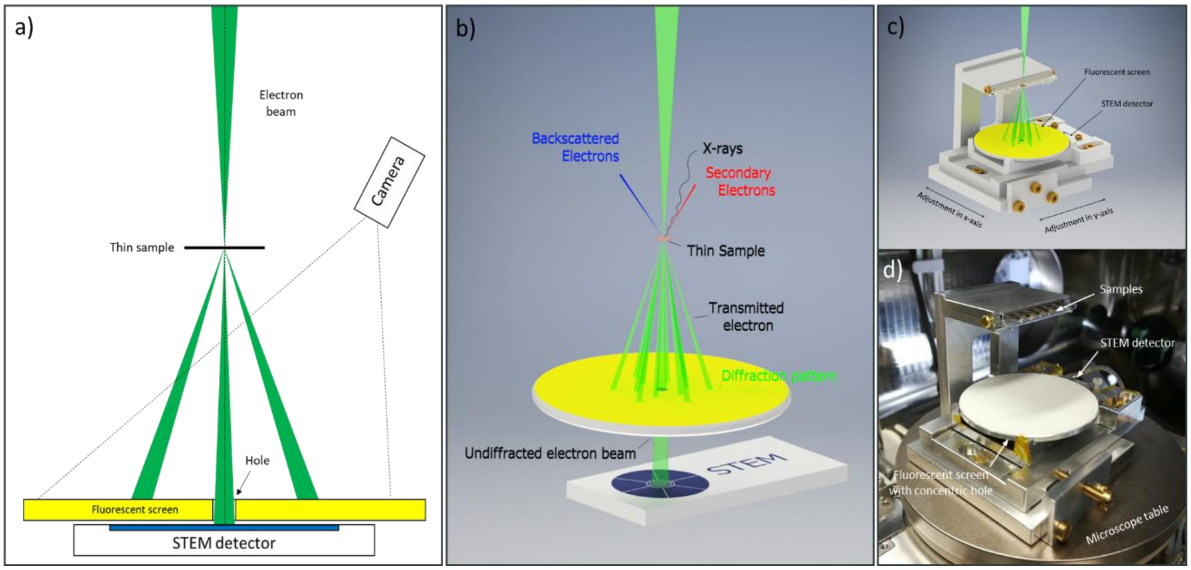

Figure 1. Developed setup for combining parallel STEM imaging with diffraction analysis. a) 2D sketch illustrates the proposed setup, which consists of an independent STEM detector placed below a fluorescent screen. The occurring (averaged) diffraction patterns are recorded by an in-chamber mounted CMOS camera. b) Illustration of all available signals in SEM (SE, BSE, STEM & diffraction), which can be collected independently for image formation without suppressing any of them. c) CAD model of the whole setup. d) Installed setup in the SEM.

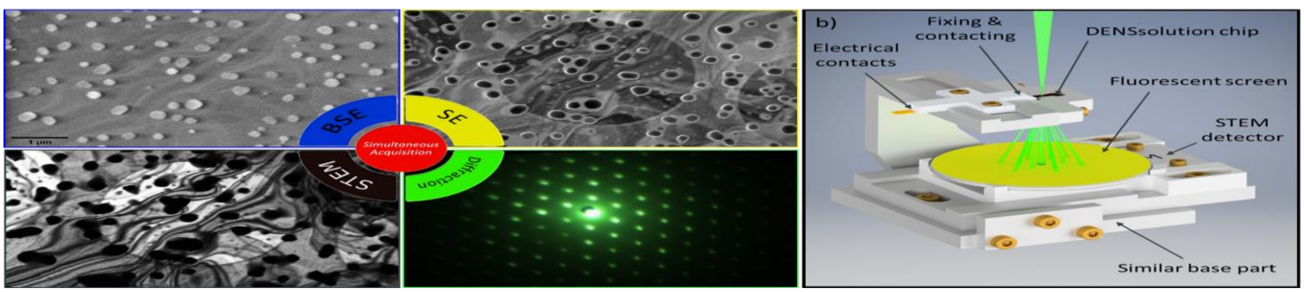

Figure 2. a) Investigation of MoS2+Au nanoparticles sample using all available signals (SE, BSE, STEM (BF) & averaged diffraction pattern). SE (yellow) offers topographic information, BSE (blue) provides elemental contrast, STEM (BF) reveals the inner structure dominated by mass-thickness and Bragg contrast and the averaged diffraction pattern (green) exhibits the crystallographic structure. b) Illustration of the next development. Upgrading the current setup to an *in situ* heating stage to perform unique heating experiments.

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