# Rapid and large FOV mapping of 60° grains in epitaxial MX2 with a segmented detector

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Owing to the rapid development in growth of synthetic $MX_2$ (where M is the metal and X is the chalcogenide) materials in recent years, their average grain sizes are fast approaching several tens to hundreds of micrometers [1]. Epitaxial growth on a single crystalline template is the favored approach to achieve wafer-scale and industrially compatible growth of high-quality single crystalline $MX_2$ monolayers. Despite the control over orientation provided by templated approaches, selective growth of domains with a single orientation while avoiding the anti-parallel orientation (or 60° rotated domains) has proven to be challenging [2]. Inclusion of such anti-parallel domains inevitably leads to the formation of 60° grain boundaries which are detrimental to electron mobility due to the presence of mid-gap states [3]. It is therefore necessary to develop methods to enable rapid and large field of view (FOV) mapping of the anti-parallel domains in epitaxial $MX_2$. We present a novel approach using segmented detector geometry to rapidly map the 60° grain boundaries over a large FOV in epitaxially grown $MX_2$. This relies on the asymmetry of diffraction peak intensities which split into two families Ka and Kb due to reduced symmetry from 6-fold to 3-fold in a monolayer of $MX_2$ [4].

The specimens are prepared by transferring the $MoS_2$ from sapphire to a carbon coated TEM grid using a tape assisted method described elsewhere in detail [3]. The conventional dark-field TEM (DF-TEM) is carried out on a Thermofisher Metrios 80-200 TEM operating at 200 kV by placing the smallest objective aperture on any first order diffraction spot. A large number of images with a certain overlap are automatically acquired in a grid pattern, preprocessed, and stitched together to form a montage with significantly larger FOV. This process is repeated for the opposite diffraction spot to obtain the complementary image. The novel segmented detector approach is carried out in a Helios5 FX dual beam operated in a “STEM in SEM” setup at 2kV and equipped with a segmented annular detector. The six segments of the detector are operated such that only 3 of the 6 segments are active for the acquisition and transmitted beam is centered as shown in schematic Fig.1 a. In this mode, as the three segments are separated from each other by 120°, the detector segments selectively collect the intensities from either the Ka or the Kb spots depending on the orientation of the grains resulting in a contrast which helps differentiate between 60° grains in monolayer $MX_2$.

The segmented detector approach is first tested on a small grain sample with high density of 60° twins. Fig.1(b) shows a color overlay of the two complementary images wherein the contrast in a monolayer is reversed between the two images making the layer appear either more green (0°) or more red (60°). The boundary separating these two regions can be either a β or γ grain boundary [3]. The images are segmented using a gaussian mixture model and assigned classes to obtain statistics on 60° twin inclusion in monolayer as well as various stacking orders in bilayer $MoS_2$ (Fig.1(c)). The method is then applied on large grain samples and compared with the more conventional montage DF-TEM approach used so far. Fig.1(d) shows a similar color overlay obtained by montaging several DF-TEM images acquired at 200kV. The contrast between the main orientation and 60° rotated domains is rather weak and images are noisier compared to those obtained with a segmented detector at 2kV shown in Fig. 1(e). Moreover, the stacking in bilayer cannot be uniquely identified with the conventional DF-TEM due to influence of unintended specimen tilt [4] which in case of the segmented detectors is overcome by design through radial integration. A drawback, however, is that 2kV electrons don’t have enough energy to see through the carbon support.

The segmented detector in SEM provides superior results from a large FOV in a fraction of the time it takes for montage acquisition and processing, i.e., few minutes compared to several hours or days. In addition. in bilayer, the stacking order can be easily assigned and not influenced by sample mistilt.

**Graphic:**

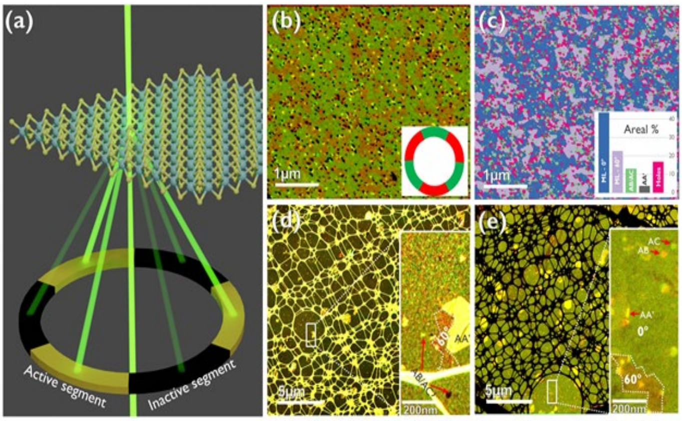

**Figure 1**: *(a) Schematic of six segmented annular detector on monolayer* $MoS_2$ *(b) Color overlay of the two complementary images obtained from the two modes of operation on a small grain sample. The inset shows the detector segments color coded with the same color as used in the color overlay. (c) Segmented image after clustering where the inset shows the areal percentage of various classes. Comparison between montage DF-TEM acquired on Metrios @ 200kV (d) and segmented detector STEM on Helios5@ 2kV (e) on different parts of the same large grain sample. Insets are magnified regions.*

**Keywords:**

Segmented, Grains, Epitaxial, $MX_2$, Stacking.

**Reference:**

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