STRUCTURE, CHEMISTRY AND CHARGE DENSITY IN MBE GROWN TMDS INVESTIGATED BY 4D-STEM

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Epitaxial growth is a route to achieve highly crystalline continuous 2D layers. Molecular beam epitaxy (MBE) could lead to the fabrication of large scale and well-oriented single crystals with a great flexibility in the choice of elements and a potential in building van der Waals stacks, alloys or 2D layers doped either electrically or with magnetic elements. In order to achieve the synthesis of the desired complex materials and to explore their functionalities, the ability to survey the structural configuration and their properties at the atomic scale is essential. High resolution scanning transmission electron microscopy (STEM) has become the most powerful technique for providing detailed atomic structure by Z-contrast direct imaging and the associated chemical information by electron energy loss spectroscopy (EELS). While charge density is among the most fundamental solid-state properties determining chemical and electrical characteristics, measuring local charge density together with detailed atomic structure poses a challenge in advanced microscopy for atomistic understanding of 2D materials. A new STEM acquisition technique so-called four-dimensional STEM (4D-STEM) consists in recording a diffraction pattern for each position that the electron probe scans on the sample in order to map multiple information found in reciprocal space [1]. Analyzing the deviation (Center of Mass: CoM) of the transmitted beam position gives access to the electric field with atomic resolution, and to the electrostatic potential and the charge density through Poisson's equation [2-3].

In this work, we first demonstrate a multi-scale analysis of MBE grown WSe_2 using the orientation and 1H-1T' phase maps reconstructed by the 4D-STEM diffraction datasets obtained over micron areas. A histogram of the orientation angle was generated for direct comparison with X-ray diffraction (XRD) data acquired at the mm scale. This step makes it possible to link large- and atomic-scale diagnostics and finally to explain the mis-orientation in layers investigated by the XRD relating to their atomic structures. Secondly, the electric field map around single dopant vanadium atoms incorporated in WSe_2 was reconstructed by the CoM measurements and then converted into local electrostatic potential and charge density maps. The quantitative interpretation of the experimentally obtained charge density was studied by comparison with the DFT calculation and the ability to identify a single impurity atom is discussed by comparing with z-contrast imaging and EELS spectrum imaging.

This 4D-STEM technique is still exploratory and far from being used in a standard way to study synthesized 2D materials. The methodology highlighted here will open the use of electron microscopy to provide atomic scale multiple information on structure, chemical and electric characteristics appearing in 2D layers and their heterostructures.

References

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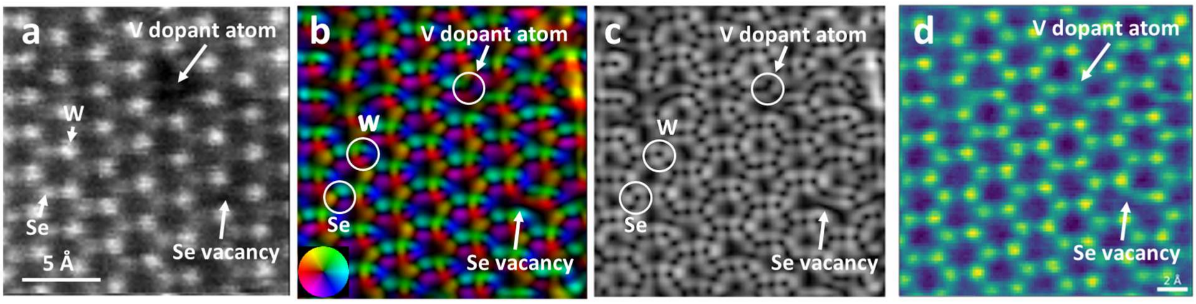

Figure: (a) Virtual HAADF image, (b) projected electric fields vector map, (c) projected electric field strength map (0-65V) and (d) projected electrostatic potential map; of vanadium doped WSe_2 monolayer grown by MBE. All maps were reconstructed from a 4D-STEM dataset.

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