Four-dimensional STEM: A new approach to probe defects in synthesized 2D materials

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Two-dimensional (2D) materials provide a versatile platform for structural engineering, where defects, stacking order, and elemental combinations can be tailored to tune physical properties. However, achieving controlled synthesis remains a key challenge, as intrinsic atomic defects become both powerful tuning parameters and unavoidable sources of discrepancies between experimental measurements and ideal theoretical predictions. Probing these defects at the atomic scale is thus essential to understand their formation and properties, but must be complemented by large-scale analysis of their distribution to build realistic structural models, where materials are described as patchworks of atomic defects that determine their general properties. Aberration-corrected scanning transmission electron microscopy (AC-STEM) has been known as a powerful and reliable technique for imaging atomic defects in atomically thin 2D layers, with a possibility to associate chemical information using spectroscopies. Recently, four-dimensional STEM (4D-STEM) has emerged as a technique that can capture local information by recording a diffraction pattern at every scanning beam position, providing quantitative data that complements atomic-scale structural and chemical imaging and also enables large-scale structural analysis. For instance, analyzing the deviation of the transmitted beam position (Center of Mass: CoM) in 4D-datasets acquired at atomic scale gives access to the local electric field around individual defects, and to the electrostatic potential and the charge density through Poisson's equation [1,2]. On a larger scale, extracting specific diffraction signals collected over extended areas allows the localization of diverse types of defects. Advances in microscopy and computational capabilities now enable the collection and processing of large datasets, but this also makes their interpretation increasingly complex and requires careful analysis combined with simulated datasets and DFT calculations to extract reliable structural and physical insights.In this talk, the application of 4D-STEM to the study of synthesized 2D materials will be demonstrated, illustrated with several examples realized in our group. Atomic-scale CoM imaging highlights the capability of 4D-STEM for quantitatively detecting negative charge accumulation around single dopant atoms in MBE-grown WSe_2 monolayers. The charge state of dopant atoms is determined by comparison with simulated datasets including DFT calculations and residual aberrations measured by the ptychographic reconstruction of the probe [3]. Mapping domain junctions in transition metal dichalcogenide (TMD) monolayers and multilayer films demonstrates the utility of large-scale defect analysis, providing an overview of the distribution of key atomic defects identified through atomic-resolution imaging and DFT calculations. This allows the statistical correlation of structural defects with the experimentally measured electrical properties of the films [4,5].

[1] C. Ophis , *Microscopy and Microanalysis* **25**, 563 (2019).
[2] K. Muller et al., *Nature Communications* **5**, 5653 (2014).
[3] D. Dosenovic et al., *ACS Nano* **18**, 23354 (2024).
[4] D. Dosenovic et al., *2D Materials* **10**, 054024 (2023).
[5] E. Desgué et al., *arXiv* **2503**, 20659 (2025).