Four-dimensional STEM: A new approach to probe structure, chemistry and electronic properties in 2D materials

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Structural defects are known as a parameter-tuning knob for material properties. In particular, two-dimensional (2D) materials, with their unique infinitely large basal planes and atomic thickness, exhibit a vast range of atomic defects that present unparalleled degrees of freedom for tailoring physical properties. The ability to engineer atomic defects at the nanometer scale opens up unique opportunities to modify and enhance electrical, optical, mechanical, and magnetic properties. On the contrary, inevitable intrinsic growth-related defects appearing in synthesized materials such as grain boundaries (GBs) modify local electronic properties in a complex way, causing a discrepancy between the properties measured in synthesized materials and those theoretically predicted from a perfect model system. Identifying atomic defects and correlating their structural and chemical configurations with local electronic properties are therefore essential for synthesizing complex materials with desired characteristics and exploring their functionalities. Aberration Corrected Transmission Electron Microscopy (AC-TEM) has become the most powerful technique for providing detailed local atomic structure of 2D layers such as graphene and transition metal dichalcogenides (TMDs). Recently a new STEM acquisition technique so-called four-dimensional STEM (4D-STEM) has demonstrated its great potential to fulfil information either on up-scaled structures or on local electric states [1]. The technique consists in recording a diffraction pattern for each scanning beam position on the sample, enabling to visualize multiple structural information in micrometer scale. Analyzing the deviation of the transmitted beam position (Center of Mass: CoM) in 4D-datasets also gives access to the local electric field at atomic scale, and to the electrostatic potential and the charge density through Poisson's equation [2].

In this talk, the application of 4D-STEM for studying various TMDs synthesized for 2D-based device materials will be presented. Mapping domain junction in WS_2 monolayer and $PtSe_2$ multilayer film will illustrate the multi-scale analysis providing insight into the density and distribution of key atomic defects and the correlation between structure and property [3-4]. Atomic scale CoM imaging highlights the capability of 4D-STEM to probe negative charge accumulation around single dopant atoms in MBE grown WSe_2 monolayer. The electrostatic potential landscape in realistic structural configurations reveals the charge state of dopant atoms strongly influenced by other defects in their environments [5].

References

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