Single-atom microscopy and spectroscopy on 2D materials

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The possibility of performing real-space imaging and spectroscopy analysis at the atomic scale via low-voltage aberration-corrected scanning transmission electron microscopy (STEM) has provided exciting new opportunities for the study of two-dimensional materials, especially for defects and interfaces. Pushing the sensitivity of STEM spectroscopy techniques down to the single atom level is expected to open a new avenue for probing the local functionalities of materials, but remains challenging. In this presentation, I will discuss our recent results on pushing the sensitivity of single-atom spectroscopy techniques using dopants in monolayer graphene as a model system. With this relatively stable single-atom model system, we can explore the ultimate sensitivity of EELS at 60 kV where the dose level is no longer the limiting factor. We show that the sensitivity of single-atom vibrational spectroscopy analysis can be pushed to the chemical-bonding level and this technique could be applied to explore local vibrational signatures at defects and interfaces in 2D materials [1]. I should also discuss briefly our recent progress of atom-by-atom isotope mapping using vibrational spectroscopy technique. As for core-loss excitations, we show that electronic states contributed by specific unoccupied pz orbital around a four-fold coordinated Si point defect in graphene can be mapped out using atomic-resolution energy-loss near-edge fine structure (ELNES) spectroscopy [2]. In addition, local coordination measurement can be achieved with single-atom sensitivity via extended energy loss fine structure (EXELFS) analysis.

[1] M. Xu et al., *Nature Materials* **22**, 612-618 (2023).
[2] M. Xu et al., *Physical Review Letters* **131**, 186202 (2023).