東京大学大学院工学系研究科 総合研究機構
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Single-atom microscopy and spectroscopy on 2D materials

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Aberration-corrected scanning transmission electron microscopy (STEM) operated at low accelerating voltage can now provide real-space imaging and spectroscopy analysis at the atomic scale and even with single atom sensitivity. Reducing the energy of the incident electron has been proven crucial for the study of two-dimensional materials and carbon-based nanomaterials. While imaging at the single atom level is becoming routine operation in many state-of-the-art STEMs, pushing the sensitivity of spectroscopy techniques down to the single atom level would provide new opportunities to probe the material functionalities, but remains relatively challenging. In this regard, 2D materials, especially doped graphene samples, could provide an ideal platform for exploring the new limit of single-atom spectroscopy techniques.

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)