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Nanoscale Heterogeneities in Monolayer MoSe₂ and WSe₂ Revealed by Correlated SPM and TERS

Two-dimensional semiconductors, specifically the broad class of transition metal dichalcogenides (TMDs) attract significant attention of research community in recent years due to the wealth of interesting and potentially applicable phenomena observed in these materials. In order to control the performance of devices based on TMDs, they must be characterized at the scale relevant to the corresponding application, which in most cases today corresponds to a few tens of nanometers [1].

Here we report on the direct application of scanning probe microscopy (SPM) cross-correlated with tip enhanced Raman scattering (TERS) imaging for characterization of rather unexpected locations of grain boundaries in single layer CVD grown MoSe₂, as well as the evolution of these grain boundaries in the course of sample aging and their transfer from the original substrate via wet etching transfer procedures. Detailed analysis of the TERS maps of MoSe₂ on Au revealed that there exist two types of nanoscale (few to several tens of nm across) domains, one featuring a resonant response (as should be expected for 638 nm laser), and the other featuring mostly a single peak near 240 cm⁻¹ (typical for non-resonant conditions).

Additionally, TERS maps on WSe₂ revealed the presence of small, 100 - 300 nm triangular areas with zero Raman response. Cross-correlation of the TERS data with the topography, surface potential and friction images revealed that large flakes had a significant number of perfectly triangular holes. Such perforations in TMD flakes can be extremely beneficial for hydrogen evolution reactions, as it has been demonstrated that the most efficient way of improving the performance of TMDs in fuel cells is to increase the ratio of edge length to surface area [2]. This ratio in the samples examined here was up to 3 - 4 times higher compared to homogenous continuous flakes. These perforated flakes can be transferred to any surface, including corrugated ones, which should inevitably cause some strain also beneficial for hydrogen catalytic activity.

References

- [1] English, C. D. *et al.*, *IEEE Int. Electron Devices Meet.* **5.6.**, 131–134 (2016).
- [2] Guoqing Li, *et.al.*, *JACS* **138**, 16632-16638, (2016)