INVITED

Multimodal tip-enhanced spectroscopy

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Combining tip-enhanced Raman and photoluminescence with active atomic force tip interaction, I will discuss novel approaches for the study of photo-physical and photo-chemical processes in molecules and nano-solids. As example, we nano-image the exciton behavior and its correlation with defects, grain boundaries, and local strain in different transition metal di-chalcogenides (TMDs). Based on exciton-plamon coupling we achieve a 10^5-fold enhancement of the photo-luminescence yield [1].

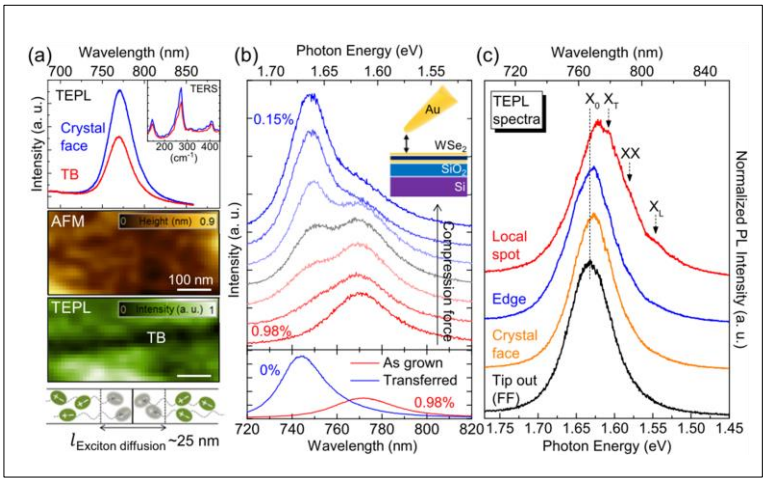

Fig. 1. (a) TEPL spectra and image of the as-grown ML WSe_2 at crystal face and twin boundary regions. (b) Evolution of TEPL spectra with increasing compressive force by the tip, giving rise to a release of the tensile strain of the crystal.

Further, we are able to achieve TEPL spectroscopy of the otherwise forbidden radiative emission from excitonic dark states with the optical antenna tip coupling to its out-of-plane transition dipole moment [2]. We achieve room temperature contrast, not possible in conventional approaches, due to the ultrafast radiative dark exciton to the tip-antenna mode with few-nm tip-sample gap localized mode volume induced Purcell factor of > $2x10^3$. With the atomic-force microscope controlled antenna tip we demonstrate correlative nano-opto-mechanical switching and programmable modulation of the dark exciton emission. This hybrid tip-enhanced nano-spectroscopy and –imaging method allows to probe and control neutral-, multi-, localized-, and dark-excitons and their correlation with lattice and electronic structural heterogeneities in 2D materials and molecular systems.

References:

1. Park, K.-D. et al., "Hybrid tip-enhanced nanospectroscopy and nanoimaging of monolayer WSe_2 with local strain control," *Nano Lett.*, Vol. 16, No. 4, 2621–2627, 2016.
2. Zhou, Y. et al., "Probing dark excitons in atomically thin semiconductors via near-field coupling to surface plasmon polaritons," *arXiv:1701.05938*, 2017.