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 photoluminescence yield [1]. 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 $> 2 \times 10^3$. 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 nanospectroscopy 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.

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.

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