Atomically-resolved exciton emission from MoS_2 on ultrathin hBN decoupling layer

Lysander Huberich[1], Eve Ammerman[1], Jian Chen[2], Sotirios Papdopoulos[3], Chengye Dong[4], Joshua A. Robinson[5,4,6], Takashi Taniguchi[7], Kenji Watanabe[8], Oliver Gröning[1], Lukas Novotny[3], Tomás Neuman[2] and Bruno Schuler[1]

[1]nanotech@surfaces Laboratory, Empa – Swiss Federal Laboratories for Materials Science and Technology, Dübendorf 8600, Switzerland
[2]Institute of Physics, Czech Academy of Sciences, Prague 18200, Czech Republic
[3]Photonics Laboratory, ETH Zürich, Zürich 8093, Switzerland
[4]Two-Dimensional Crystal Consortium, Pennsylvania State University, University Park, PA 16802, USA
[5]Department of Materials Science and Engineering, The Pennsylvania State University, University Park, PA 16082, USA
[6]Department of Chemistry and Department of Physics, The Pennsylvania State University, University Park, PA, 16802, USA
[7]International Center for Materials Nanoarchitectonics, National Institute for Materials Science, 1-1 Namiki, Tsukuba 305-0044, Japan
[8]Research Center for Functional Materials, National Institute for Materials Science, 1-1 Namiki, Tsukuba 305-0044, Japan

lysander.huberich@empa.ch

Atomically thin 2D materials with deliberate single atomic defects are emerging as foundational components for next-generation optical and electronic devices. Using Scanning Tunnelling Microscopy (STM) and electron-induced STM-Luminescence (STML), we investigate the atomically resolved electronic and optical emission properties of MoS_2 and embedded single defects. To achieve excitonic emission, we decouple MoS_2 from the graphene (Gr) substrate by inserting a bilayer of hBN (Fig. a). This decoupling increases the electronic bandgap, extends defect charge state lifetimes and prevents quenching of the emission. Spatially resolved spectral mapping of the pristine MoS_2 heterostructure reveals sharp emission lines corresponding to excitons and trions, exhibiting nanoscale sensitivity to local potential fluctuations and variations in the dielectric environment. We further identify the optical fingerprints of single atomic defects including oxygen substitution defects (O_s), sulfur vacancies (Vac_s^-), and negatively charged defects (CD^-) assigned to CH_s^-. O_S and Vac_S^- suppress pristine emission, while CD^- give rise to defect-bound exciton complexes (A^-X) 250 meV below the MoS_2 exciton (Fig. b,c). Sub-nanometer-resolved STML measurements elucidate the charge-mediated excitation mechanism and the plasmonic nanocavity-induced spectral shifts of defect-bound excitons. These findings establish an atomically precise correlation between structural, electronic, and optical fingerprints of single atomic defects in MoS_2, enabling deterministic engineering of quantum emitters in 2D materials.

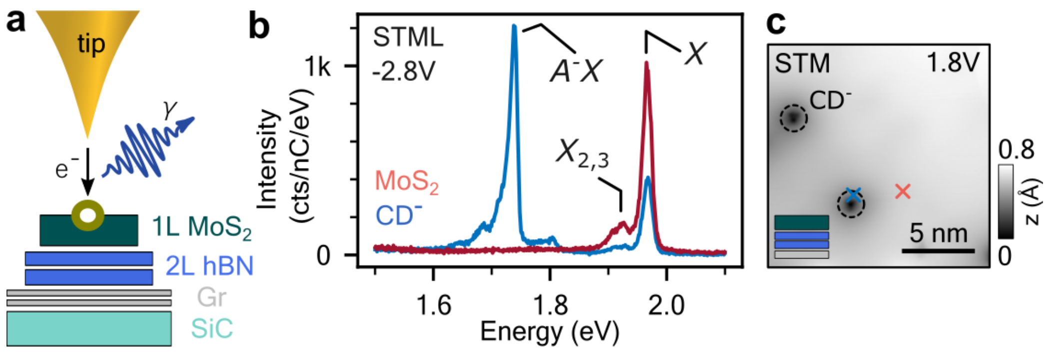

Figure 1: a) Sketch of MoS_2/hBN/Gr heterostructure in the STM. **b)** STML emission spectra of pristine MoS_2 (red) and negatively charged defect (CD^-) (blue). **c)** STM topography image of MoS_2 heterostructure with STML measurement locations indicated.