Strain Tuning of Quantum Emitters in Monolayer Transition Metal Dichalcogenides

CNF Project Number: 2126-12

Principal Investigator(s): Gregory Fuchs

User(s): Jaehong Choi

Affiliation(s): Applied and Engineering Physics, Cornell University

Primary Source(s) of Research Funding: Air Force Office of Scientific Research Multidisciplinary Research Program of the University Research Initiative (AFOSR MURI)

Contact: gdf9@cornell.edu, jc3452@cornell.edu

Primary CNF Tools Used: JEOL 6300, 5X g-line stepper, Oxford COBRA, Oxford 81, DISCO dicing saw, CVC SC4500 odd hour evaporator, Veeco Icon AFM, wire bonder, Heidelberg mask writer - DWL2000

Abstract:

Tensile strain has been known to modulate the band gap of two-dimensional (2D) transition metal dichalcogenides (TMDs), which effectively funnels excitons and activates quantum emitters in a deterministic way. We used an array of cylindrical nanopillars to create quantum emitters and discovered that single photon emitters with high purity are often formed on wrinkles rather than on the pillar apex. We also studied strain-tuned interlayer excitons in heterobilayers of 2D TMDs. We discovered that the strain exerted by nanopillars activates interlayer excitons in WSe_2/WS_2 heterobilayers irrespective of twist angles. We confirmed that the strain profile of the pillar apex is complex and that spectra associated with the pillar apex have multiple emission peaks with higher background. Wrinkles formed around the pillars, on the other hand, often generate spectra with lower background.

Summary of Research:

Monolayer transition metal dichalcogenides (TMDs) have been actively studied for quantum technology and optoelectronics due to their unique properties such as strong excitonic binding, a direct band gap, and spin-valley locking [1]. It has been shown that tensile strain modulates the band structure of 2D TMDs [2-4]. Site-localized tensile strain reduces the local band gap enabling excitons to be funneled into the strain potential before recombination, leading to single photon emission. Nanostructures such as nanopillars [2-3], nanorods [5], and nanobubbles [6] have been used to deterministically activate single photon emitters through this strain confinement. However, the strain induced by these nanostructures often have complex profiles and this typically results in multiple confining sites or large confining potential that hampers the single photon emission.

Our group has strain-tuned monolayer WSe_2 with cylindrical nanopillars and discovered that the wrinkle formed nearby the nanopillar gives rise to single photon emitters with higher purity as with compared to the emitters from the pillar apex (Figure 1) [7].

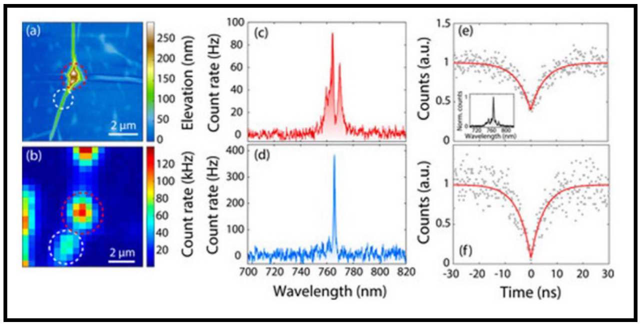

Figure 1: Comparison between pillar emission and wrinkle emission on WSe_2 stacked on nanopillars. (a) Atomic force microscopy image showing a pillar and wrinkles, (b) Photoluminescence map of the pillar (red) and wrinkle (white), (c),(e) spectrum and g2 measurement of emitters from the pillar apex. (d),(f) spectrm and g2 measurement of emitters from the wrinkle [7].

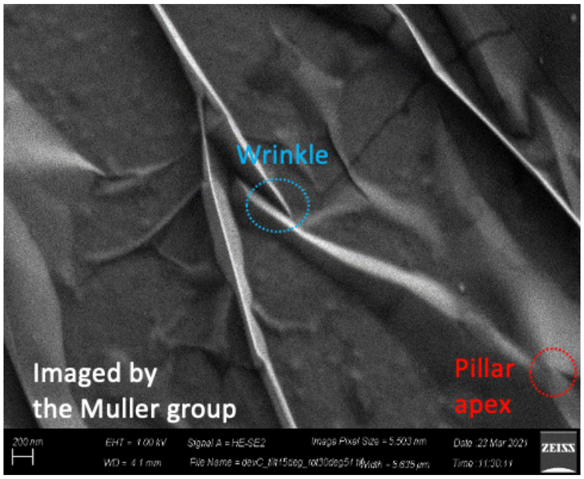

Figure 2: Scanning electron microscopy image of wrinkles formed on h-BN/WSe$_2$. The sharp folds on the wrinkle (blue) often exhibit sharp emission peaks.

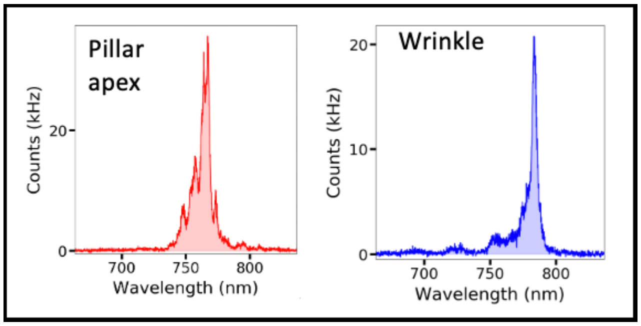

Figure 3: Spectra of interlayer excitons associated with the pillar apex (red) and wrinkle (blue).

The physics behind this finding can be better understood by quantifying the strain profile on the pillar apex and the wrinkle. With high resolution electron microscopy in collaboration with Professor David Muller's group, we found out that wrinkles with a sharp fold gives rise to single photon emitters with high purity (Figure 2). The characteristics of the strain exerted by wrinkles can be further explored by quantifying the strain gradient via electron microscopy, and this is in progress.

We also used nanopillars to activate interlayer excitons in a WSe$_2$/WS$_2$ heterobilayer. When two different TMD monolayers are stacked together, the electrons and holes find their energy minima in the composite material. When a bilayer is engineered such that the composite layer forms a type II band alignment, the coulomb-bound electron and hole can be separated into different layers. It has been found that the photoluminescence intensity of interlayer excitons is enhanced at 0° and 60° alignment angles, but significantly suppressed at intermediate angles due to the momentum mismatch [8]. We stacked WSe$_2$/WS$_2$ heterobilayer onto nanopillars and studied the strain effect.

We found that strain-tuned interlayer excitons exhibit fairly high photoluminescence intensity at any twist angle, which implies that momentum matching is not necessary when interlayer excitons are confined by the strain potential. We also observed that the spectra associated with wrinkles often exhibit sharper peaks with lower background compared to those associated with pillar apex (Figure 3), which is consistent with our work on monolayer WSe$_2$ [7].

Conclusions and Future Steps:

Nanopillars create a strain potential that confines excitons. We discovered that the emitters formed on wrinkles produce spectra with low background and sharp emission peaks. Also, strain-confined interlayer excitons exhibit high photoluminescence intensity irrespective of twist angle. Our next step is to quantify the strain profile on the wrinkle via electron microscopy, which will allow us to better understand the physics of wrinkles and design high-quality emitters more deterministically.

References:

[1] Mak, K., Shan, J., Nat. Photon 10, 216-226 (2016).

[2] Palacios-Berraquero, C., et al. Nat Commun 8, 15093 (2017).

[3] Branny, A., et al. Nat Commun 8, 15053 (2017).

[4] Chaves, A., et al. npj 2D Mater Appl 4, 29 (2020).

[5] So, Jae-Pil, et al. Nano Lett 21, 1546-1554 (2021).

[6] Darlington, T.P., et al. Nat Nanotechnol. 15, 854-860 (2020).

[7] Rapahel S. Daveau, et al. APL Photonics 5, 09615 (2020).

[8] Nayak K. Pramoda, et al. ACS Nano 11, 4, 4041-4050 (2017).