Two-Dimensional TMD as a Single Photon Source and a Quantum Sensor

CNF Project Number: 2126-12

Principal Investigator(s): Gregory Fuchs, David Muller

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, Oxford COBRA, 5X g-line Stepper, Wire Bonder, DISCO Dicing Saw, SC4500 Odd-Hour Evaporator, AFM – Veeco Icon, Zeiss Supra SEM, Zeiss Ultra SEM

Abstract:

Electronic and optical properties of two-dimensional transition metal dichalcogenides (2D TMDs) are readily tuned by strain and external fields due to their atomic thickness. Tensile strain is known to confine excitons and create single photon emitters in 2D TMDs. We are working toward using electron microscopy to directly measure the strain to better understand the strain confinement of excitons. Also, the exciton emission energy in TMDs can be tuned with an external field and we tried using an interlayer exciton in homobilayer tungsten diselenide (WSe_2) as a quantum sensor for the polarization change in ferroelectric materials in vicinity.

Summary of Research:

Single photon emitters with high brightness and purity are key components for quantum communication and information technologies. Monolayer transition metal dichalcogenides (TMDs) have been established as promising sources of single photon emitters, and a recent study showed that the hybridization of strain-localized excitonic state and localized defect state induces single photon emission in monolayer WSe_2 [1]. To better understand the interplay among strain, exciton, and defect, it is crucial to have a clear picture of strain localization of excitons.

Electron microscopy is promising for the direct measurement and quantification of strain responsible for single photon emission in monolayer WSe_2. We were able to create single photon emitters by stacking monolayer WSe_2 on top of a nanorod-gap array. Monolayer WSe_2 is folded into nanogaps (90 nm) between nanorods, and the high strain point at the wrinkle confines excitons to create single photon emitters with high purity (Fig.1).

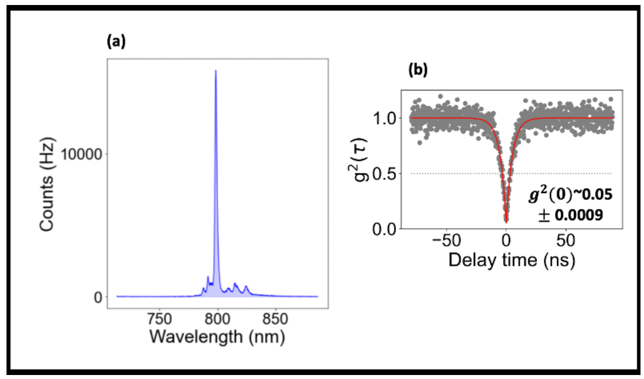


Figure 1: (a) Spectrum from the fold at the nanogap site. We observed an emission peak with a narrow linewidth, which implies a strong exciton confinement. (b) Time correlation measurement of the emitter created at the nanogap. We observed the antibunching with $g^2(0)$ value ~ 0.05, which unambiguously shows the high purity of the single photon emitter.

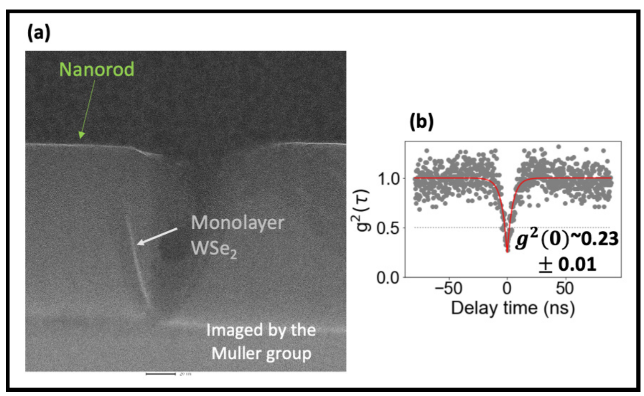
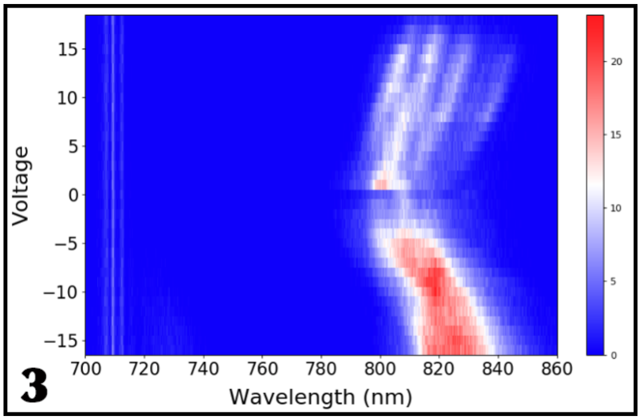


Figure 2: (a) TEM image of a nanogap site with the sharply folded monolayer WSe_2. (b) Time correlation measurement of a quantum emitter created at the sharp fold in (a). $g^2(0)$ is 0.23, which shows a single photon nature of the emitter.



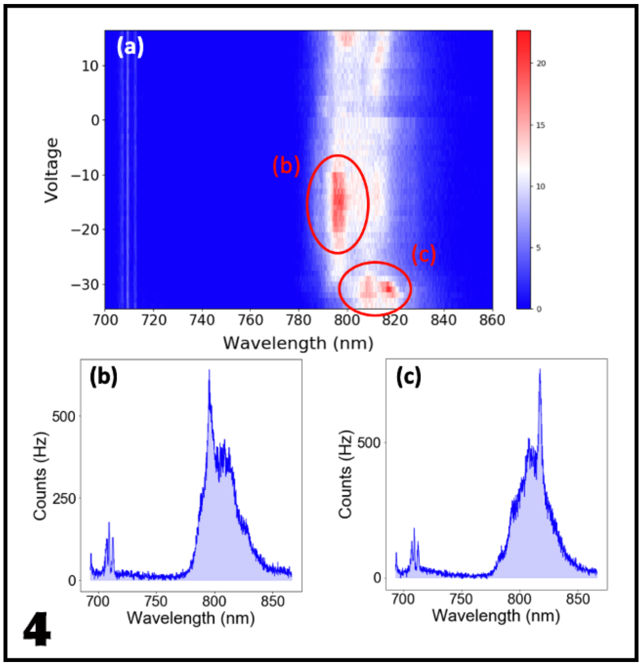


Figure 3, top: Photoluminescence modulated by V_{gb}. V_{tg} is adjacent to 2L-WSe$_2$, and it directly pumps charge carriers into 2L-WSe. We can see a clear doping effect. Figure 4, bottom: (a) Photoluminescence modulated by V_{tg}. V_{tg} is adjacent to BTO, and it switches the polarization in BTO. (b) and (c) show the change in population proportion of $\Lambda - K$ and $\Lambda - \Gamma$ indirect excitons under the polarization switching in BTO.

To better understand the strain confinement of excitons at the wrinkle apex, Transmission electron microscopy (TEM) imaging was attempted on the nanogap sites in collaboration with the Muller group at Cornell University. By TEM imaging, we confirmed that the high strain achieves a strong confinement of a single exciton. Anti-bunched quantum emitters were observed at sharp folds (Figure 2). At relatively flat and gradual folds, the strain was expected to be small. This resulted in creation of multiple emitters.

One of the pre-requisites for the strain analysis is that the atom columns in the material should be aligned with the beam axis. However, monolayer WSe$_2$ is thin and flimsy. We were unable to align the beam axis with the atom columns in WSe$_2$. To allow the strain analysis, it is important to have a support layer that gives a structural support so that the curvature of wrinkle is uniform throughout a monolayer WSe$_2$.

The next step is using a thick support layer underneath monolayer WSe$_2$ which will enable the strain analysis.

2D TMDs can be used as quantum sensors as well due to their sensitivity to their immediate environment. In homobilayer (2L) WSe$_2$, the Bloch states of valleys at each point in the momentum space have different orbital compositions, and this makes wavefunctions reside at different positions in real space and form interlayer exciton (2). Due to the spatial distance between an electron and a hole, interlayer excitons possess a dipole moment in an out-of-plane direction. This dipole moment can be modulated by an external field (2-4), so an interlayer exciton can be used to sense a polarization-induced field in ferroelectric materials in vicinity (4,5).

We studied the optical property change in 2L-WSe$_2$ in response to the polarization change in ferroelectric BTO in 2L-WSe$_2$/BTO hybrid heterostructure. A gate voltage was applied to modulate the polarization in BTO *in situ*. We observed that the spectral change in response to the polarization switching. When the bottom gate (V_{bg}), which is adjacent to 2L-WSe, was modulated, we observed a clear doping effect (Figure 3). When we modulated the top gate (V_{tg}), which is adjacent to BTO, the population proportion of indirect $\Lambda - K$ and $\Lambda - \Gamma$ excitons was changed at different voltage values (Figure 4). We wanted to see the hysteresis, but BTO started leaking at higher voltages, so the higher range voltage sweep was not possible. Next step is using a much thicker BTO membrane to minimize the leaking and study the photoluminescence change in WSe$_2$ when the polarization switches in BTO.

Conclusions and Future Steps:

Deterministic activation of single photon emitters is crucial for quantum technology application, and a better understanding of exciton confinement that leads to single photon emission is required. Electron microscopy on the strained monolayer WSe$_2$ will enable us to directly measure and quantify the strain responsible for single photon emission. Homobilayer WSe$_2$ was used to sense the polarization change in ferroelectric BTO. Interlayer exciton emission energy and spectral composition are expected to be modulated by the polarization-induced field in ferroelectric BTO. Thick BTO will be used to minimize the leaking and allow a wide range of voltage sweep.

References:

[1] Linhart, L., et al, Phy. Rev. Lett. 123, 146401 (2019).

[2] Huang, Z., et al, Phy. Rev. Lett. B 105, L041409 (2022).

[3] Altaiary, M., et al, Nano Lett. 22, 5, 1829-1835 (2022).

[4] Wang, Z., et al, Nano Lett. 18, 1, 137-143 (2018).

[5] Li, T., et al., Nano Lett. 17, 922.927 (2017).

[6] Li, P., et al., Adv. Funct. Mater. 2201359 (2022).