

Fabrication and Characterization of Transition Metal Dichalcogenides for the Production of Single Photon Emitters

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Abstract. *This work aimed to fabricate and characterize a single-photon emitter based on WSe₂ monolayers conformed onto tungsten nanopillars via strain engineering. Optical measurements showed enhanced emission intensity and redshift in strained regions, features typical of single-photon emitters in nanopillar structures. However, photon anticorrelation yielded $g^2(0) = 0.67 \pm 0.022$, indicating classical behavior. The low purity may result from lack of encapsulation, multiple defects, and limited spectral filtering. Still, the results show potential for advancing these emitters in quantum technologies.*

1. Introduction

The fabrication of deterministic single-photon emitters (SPEs) is essential for the efficiency of quantum communication, computing, and metrology systems. Spatial and spectral control of photon emission ensures that only one photon is emitted per excitation, enabling the determinism required for integration into scalable photonic circuits and emerging quantum technologies [AHARONOVICH, ENGLUND and TOTH 2016].

SPEs based on quantum dots (QDs) and color centers are widely studied but require cryogenic operation and face challenges regarding precise emitter localization and production scalability [TOTH and AHARONOVICH 2019]. In contrast, defect engineering in transition metal dichalcogenides (TMDs), such as tungsten diselenide (WSe₂), has emerged as a promising alternative.

The intentional creation of defects in TMDs enables exciton confinement in specific regions, facilitating single-photon emission [AZZAM, PARTO and MOODY 2021]. This technique, known as strain engineering, uses localized mechanical deformations to induce strain and consequently excitonic confinement states that promote quantized light emission [LINHART et al. 2019].

Despite their potential, these emitters still face challenges related to emission efficiency and reproducibility, due to the complexity of emission mechanisms and the

precise control of structural defects [TOTH and AHARONOVICH 2019]. Recent research aims to overcome these limitations by employing substrates with nanopillars to induce controlled local strain, improving SPE emission in TMDs [PARTO et al. 2021].

This work aims to fabricate and characterize a single-photon emitter based on a WSe₂ monolayer (ML) conformed over tungsten nanopillars, employing strain engineering to induce controlled defects and excitonic confinement.

2. Theoretical review

WSe₂ is a TMD with a hexagonal crystal structure that, in its ML form, presents a direct bandgap due to the band structure reorganization at high-symmetry points in reciprocal space (Figure 1a) [YUANZHENG et al., 2018]. This ML consists of a tungsten layer between two selenium layers, bonded covalently in-plane and by van der Waals forces out-of-plane [LIU et al., 2013]. The lack of spatial inversion symmetry in WSe₂ ML makes it non-invariant under inversion operations [XU et al., 2014]. Combined with the strong spin-orbit coupling from tungsten orbitals, this leads to energy level splitting at the K⁺ and K⁻ points of the Brillouin zone [XIAO et al., 2012]. Consequently, each valley exhibits distinct spin states, allowing selective excitation with circularly polarized light — σ^+ for K⁺ and σ^- for K⁻ [XIAO et al., 2012].

Photoexcited electron-hole pairs form excitons with high binding energy, enabling their stability at elevated temperatures in 2D TMDs [MUELLER and MALIC, 2018]. Their optical recombination can be bright or dark (Figure 1b), depending on spin alignment and selection rules. Neutral excitons (X₀) consist of opposite-spin pairs; dark excitons involve misaligned spins or momentum-forbidden transitions [MAK et al., 2010]. Structural defects may also trap excitons, forming defect-bound states within the bandgap [AZZAM, PARTO and MOODY, 2021].

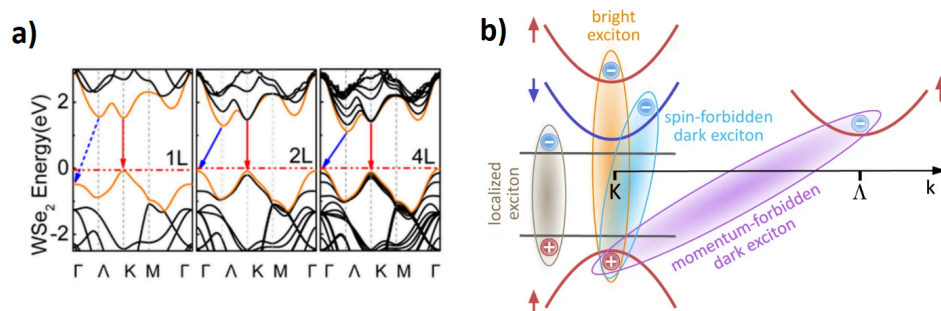


Figure 1. (a) Band structure of WSe₂ for different thicknesses; arrows indicate indirect (blue) and direct (red) transitions [YUANZHENG et al., 2018]. (b) Exciton types in spin-orbit-coupled TMDs [MUELLER and MALIC, 2018].

SPEs in nanopillars use mechanical strain engineering to locally deform TMD MLs, inducing exciton confinement and enabling single-photon emission. The ML conformation funnels excitons into low-energy regions, where defect states and localized excitons generate intense, narrow, redshifted emissions [LINHART et al., 2019]. These effects depend on pillar geometry, ML quality, and defect density. Despite

advances, no unified theoretical model yet fully describes strain- and defect-induced single-photon emission in 2D TMDs [PARTO et al., 2021].

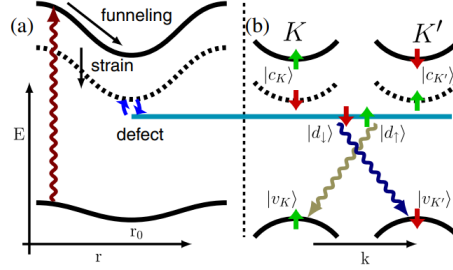


Figure 2. (a) Band variations from ML conformation and confinement; (b) Degeneracy breaking in reciprocal space due to defect and dark band changes [LINHART et al., 2019].

The quantum nature of the emission was verified via photon antibunching using the Hanbury Brown and Twiss (1956) setup, where the signal is split into two paths and detected independently to compute $g^2(t = 0)$; values below 0.5 confirm non-classical emission [TOTH and AHARONOVICH, 2019]. Purity, defined as $1 - g^2(0)$, reflects the likelihood of sequential single-photon emission. In applications like quantum key distribution (QKD), high purity and extraction efficiency ($g^2(0) < 0.1$) are required to outperform classical sources and reduce statistical uncertainty [AZZAM, PARTO and MOODY, 2021]. Although $g^2(0) < 0.5$ confirms non-classical emission, it does not ensure the deterministic emission of a single photon. Instead, it quantifies the statistical suppression of multiphoton events. Therefore, values of $g^2(0)$ significantly below 0.5 are desired to approach true single-photon operation, but additional techniques such as pulsed excitation and lifetime control are required to guarantee one-photon-per-pulse emission [SENELART, SOLOMON and WHITE, 2017].

3. Methodology

The sample was prepared by mechanically exfoliating WSe₂ crystals onto a polydimethylsiloxane (PDMS) membrane. MLs were identified via optical microscopy using apparent color patterns, as described by Puebla et al. (2022), and then transferred onto a silicon substrate with a 285 nm SiO₂ layer containing pre-patterned tungsten nanopillars. These nanopillars were fabricated using Focused Ion Beam (FIB) with 300 nm height, 200 nm diameter, arranged in a 9 × 9 matrix with 4 μm spacing. The ML was transferred and conformed using a PDMS-based system heated to 60 °C. Raman and photoluminescence (PL) spectroscopies were used to confirm the ML thickness, while Atomic Force Microscopy (AFM) assessed topography and structural integrity over the nanopillars.

Optical analysis employed aligned mirrors to direct a right-circularly polarized laser beam into the cryostat, targeting the K⁺ valley. The sample was held at 3.4 K and irradiated with 660 nm and 730 nm lasers. Emissions were recorded using a spectrometer coupled to a Charged Coupled Device (CCD) detector. Photon antibunching was measured using two avalanche photodiodes in a Hanbury Brown and Twiss (HBT) setup to obtain the $g^2(t)$ correlation function.

4. Results and Discussion

Raman and photoluminescence (PL) spectroscopies confirmed that the transferred material corresponds to a WSe₂ ML, as evidenced by the characteristic positions of the optical peaks, in agreement with the literature [YUANZHENG et al., 2018]. AFM revealed wrinkles, perforations, and well-conformed regions over the nanopillars. Figure 3 shows the ML profile with a measured height of ~260 nm, close to the designed 300 nm. A stretched-sheet-like conformation is ideal, as it enhances exciton confinement and supports efficient single-photon emission [LINHART et al., 2019].

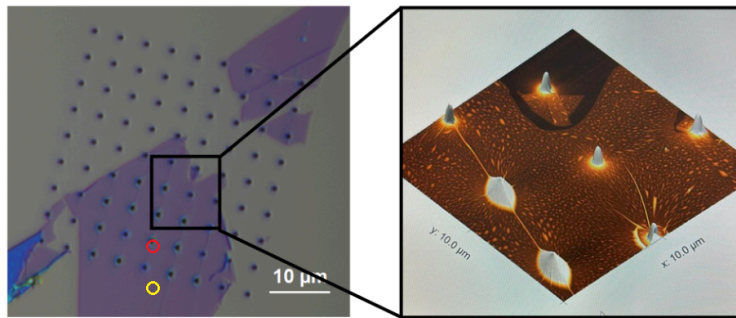


Figure 3. WSe₂ (purple) conformed over tungsten nanopillars. Yellow and red circles mark measurement points on the ML and nanopillar. Right: AFM image of the sample.

Spectroscopic measurements of excitonic emission were conducted. Figure 4.a presents the free-space spectrum on the flat ML; Figure 4.b shows emission from the nanopillar. The latter exhibited peak narrowing, higher intensity, and redshift, attributed to strain-induced confinement [PARTO et al., 2021; LINHART et al., 2019]. The nanopillar emission was over 25× stronger, indicating high extraction efficiency, likely enhanced by strain–defect interaction [PARALIKIS et al., 2024]. Based on the emission observed in Figure 4.b and a total acquisition time of 5×1 seconds, the photon emission rate was estimated to be approximately 2 kHz.

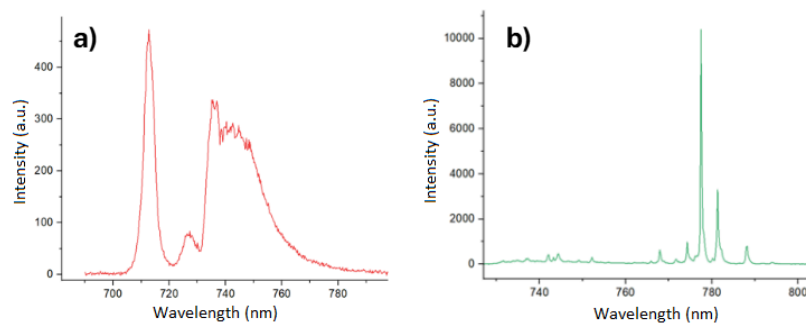


Figure 4. (a) PL measurement on the ML; (b) PL measurement on the nanopillar.

To suppress unwanted light states, the emission was filtered through a single-mode fiber and bandpass filters (Figure 5.a), reducing intensity by over 70% and allowing antibunching measurements. Under these conditions and with an acquisition time of 3×1 seconds, a photon detection rate of approximately 100 Hz was obtained.

Final characterization involved photon antibunching using two avalanche photodiodes in a Hanbury Brown and Twiss setup. The $g^2(t)$ function after 12 hours (Figure 5.b) yielded $g^2(0) = 0.67 \pm 0.022$, indicating $\sim 33\%$ purity—above the $g^2(0) \leq 0.5$ quantum threshold, suggesting classical behavior [TOTH and AHARONOVICH, 2019]. Coexisting emitters or overlapping defect states may have degraded the purity, despite the high emission intensity.

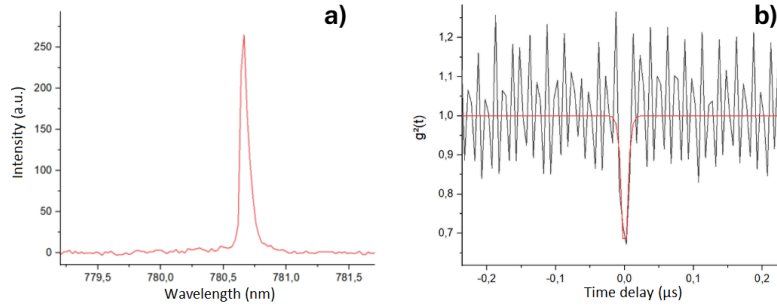


Figure 5. (a) PL measurement on the ML coupled to single-mode fiber; (b) PL measurement on the nanopillar.

5. Conclusion

The sample fabricated via strain engineering, with a WSe₂ ML conformed over tungsten nanopillars, showed optical behavior consistent with the literature, including enhanced emission intensity, peak narrowing, redshift, and spatial localization — effects linked to strain- and defect-induced exciton confinement [PARTO et al., 2021; LINHART et al., 2019].

Photon antibunching measurements yielded $g^2(0) = 0.67 \pm 0.022$, indicating classical emission rather than true single-photon generation [TOTH; AHARONOVICH, 2019]. Low purity may result from multiple emitters and spectral filtering limitations, highlighting the need for better defect control and optical isolation. Encapsulation and advanced defect engineering are recommended, as shown by Parto et al. (2021), who achieved $g^2(0) \approx 0.28$ at 150 K and $g^2(0) \approx 0.05$ at 5 K.

These findings highlight that 2D-material-based SPEs, such as those in TMDs, offer promising features compared to attenuated classical sources and well-established solid-state emitters, combining potential operation at elevated temperatures with scalability — key aspects for the next generation of quantum technologies.

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