Quantum Emitters Activation in WSe_2 Monolayers via Hexagonal Boron Nitride Wrinkle Engineering

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Abstract:

Strain has been shown to be a key parameter to create quantum emitters in tungsten diselenide (WSe_2) monolayers. We report experimental observations of spatially and spectrally isolated single quantum emitters (QEs) in WSe_2 monolayers. We use hexagonal boron nitride (hBN) wrinkles nucleated from a nanostructured substrate to create strain at a size scale suitable to host single QEs, while avoiding other residual emission. In comparison, QEs stemming from direct strain points from the substrate yield multiple QEs per site and increased unwanted background emission.

Summary of Research:

Monolayer transition metal dichalcogenides are direct bandgap semiconductors with a strong light-matter interaction and possessing a new electronic degree of freedom, known as valley, that locks excitons to a given photon helicity [1]. Tungsten diselenide (WSe_2) has been shown to host bright and stable QEs [2]. While most reported emitters in WSe_2 monolayers are located at random, few reports show deterministic activation of QEs via local strain by placing the WSe_2 monolayer over a patterned substrate, in the form of nanopillars [3]. Strain engineering of WSe_2 monolayers is key to create QEs on demand and with position control [4].

Our approach uses a nano-patterned substrate as an indirect means to communicate strain to WSe_2 via a thin hBN layer, which appears to be suitable to create single QEs. In this work, we use a substrate patterned with nanopillars [3,4] and make a comparative study of emitters forming directly on the pillars versus along hBN wrinkles that propagate between pillars.

The nanopillars are fabricated from a SiO_2 substrate via electron-beam lithography (JEOL 6300) using M-aN 2403 negative resist. The nanopillars are etched into SiO_2 and are cylindrical with a diameter of 200 nm and height of 300 nm. Exfoliated WSe_2 monolayers and hBN are transferred using a polycarbonate (PC) on polydimethylsiloxane (PDMS) stamp technique. We study the samples using a homebuilt confocal microscopy setup with a 637 nm continuous wave laser at 10 K.

Figure 1 shows an atomic force micrograph (AFM) of a sample consisting of a hBN/WSe_2 heterostructure on a SiO_2 nanopillar substrate. The photoluminescence map from this sample shows that WSe_2 on the nanopillar (indicated with a large circle in Figure 1) emits most intensity because of the high strain while a less intense spot is visible along a hBN wrinkle (small circle in Fig.1).

The spectrum collected from the center of the nanopillar is shown in Figure 2 and shows two sharp features along with a broad emission from 755 nm to 775 nm.

Figure 3 on the other hand shows a spectrum taken on the wrinkle (see small circle in Fig.1). This spectrum features a single sharp peak at 765.8 nm with minimal background emission surrounding the peak. The AFM image reveals a kink in the wrinkle, i.e., increased strain, which can explain the creation of a quantum emitter at this specific site. At this location, the wrinkle is 90 nm high and has a full-width at half-maximum (FWHM) of 160 nm. The collected light from this spot is filtered through a 3 nm bandpass filter and sent to a Hanbury-Brown-Twiss interferometer for auto-correlation measurement. The time correlation between the two APD signals for the wrinkle QD is plotted as $g^{(2)}(t)$ in the

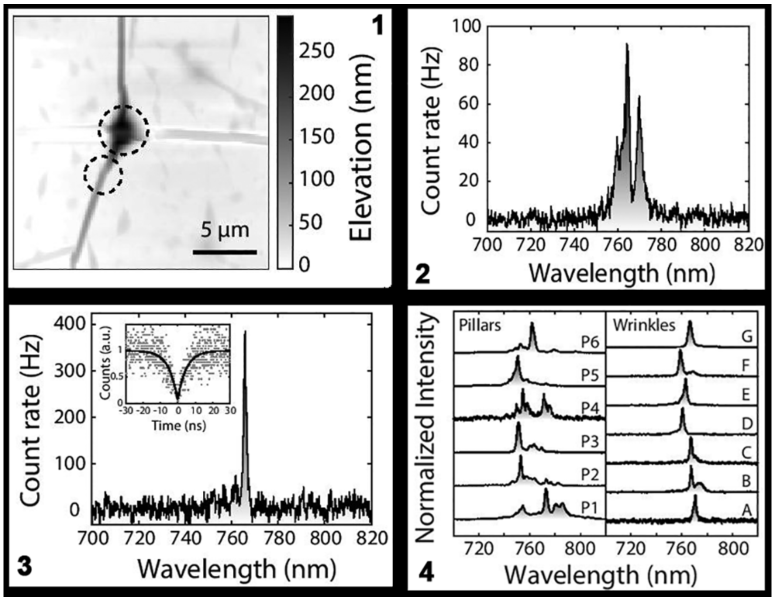

Figure 1: *Atomic force micrograph of the sample consisting of a WSe_2/hBN heterostructure on top of SiO_2 nanopillar substrate. Pillars bring most strain to WSe_2, however, hBN wrinkles bring more subtle strain suitable for creating cleaner single quantum emitters.* ***Figure 2:*** *Spectrum of light collected from the top of the nanopillar. Several peaks on top of a broad background are visible.* ***Figure 3:*** *Spectrum of light collected from the wrinkle, as circled in Fig.1. One clean peak is visible, meaning a single quantum emitter is created at this spot. Inset: second-order correlation function demonstrating the single-photon nature of the emission.* ***Figure 4:*** *(left) Spectra collected from the top of 300-nm high nanopillars. (right) Spectra collected from emission centers located along hBN wrinkles. The spectra systematically display a single peak and lower background level.*

inset of Figure 3. From the fit we extract $g^{(2)}(0)$ = 0.087, demonstrating the single-photon nature of the emission.

The spectrum of Figure 2 does not allow us to quantify $g^{(2)}(0)$ for the signal coming from the top of the nanopillar.

In another sample, we have studied six nanopillar-based QEs and seven wrinkle-based QEs. The spectra collected from the top of the pillars are shown in Figure 4 (left panel) and display one or several lines attributed to QEs and a broad emission background. In contrast, spectra that are collected along hBN wrinkles away from the pillars, shown in Figure 4 (right panel), consistently display a single peak, i.e., a single quantum emitter probed at a time, while the background level is significantly reduced.

We study the amount of background light emitted at the same frequency of the emitters on a 3~nm spectral window to quantify the effect on single-photon purity. We find that 25% of the signal is composed of unwanted background light for nanopillar-based emitters whereas this number is 15% for wrinkle-based emitters in comparison. This latter result intrinsically increases single-photon purity of wrinkle-based QEs.

Conclusions and Future Steps:

The use of a nano-patterned substrate to indirectly induce hBN wrinkles, may be a good choice to create single WSe_2 QEs with low background. Emitters created with hBN wrinkles are spectrally isolated from other emitters and have an increased single-photon purity, which is an advantage for application purposes. Future work should address the deterministic positioning of QEs along hBN wrinkles.

References:

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