Optical Properties of Locally Strained WSe_2 Monolayers

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Primary CNF Tools Used: JEOL 6300 electron-beam writer, GCA 5x stepper, Oxford 81 etcher

Abstract:

We investigate spectral properties of locally strained tungsten diselenide (WSe_2) monolayers. The local strain is created by layering the WSe_2 monolayer over a silicon dioxide (SiO_2) substrate pre-patterned with 150 nm-wide nanopillars. From spectral measurements, we show that strain shrinks the band gap resulting in local wells that concentrate carriers and redshifts the exciton recombination energy. Low temperature measurements show that the strain gradient is so far insufficient to create quantum confinement of carrier to generate single-photon emission.

Summary of Research:

Monolayer transition metal dichalcogenides (TMD) are direct bandgap semiconductors with a very strong light-matter interaction, large excitonic effects and possessing a new electronic degree of freedom, known as valley, that locks excitons to a given photon helicity [1]. Tungsten diselenide (WSe_2) is one such TMD, which has been shown to host quantum emitters [2]. Applications of quantum emitters inheriting valley properties are numerous, including dynamic generation of flying qubits with orthogonal polarization. While most reported emitters in WSe_2 monolayers are located at random, few reports show deterministic activation of quantum emitters via local strain by placing the WSe_2 monolayer over a patterned substrate [3]. The strain locally bends the band gap and creates quantum confinement of charge carriers, eventually leading to the emission of single photons upon recombination. Here, we report on the fabrication of nanopillar-patterned substrates, transfer and successful local straining of WSe_2 monolayers. We quantify the effect of strain on the band gap of WSe_2 at room temperature, while measurements at cryogenic temperatures remain elusive to resolve quantum emitters.

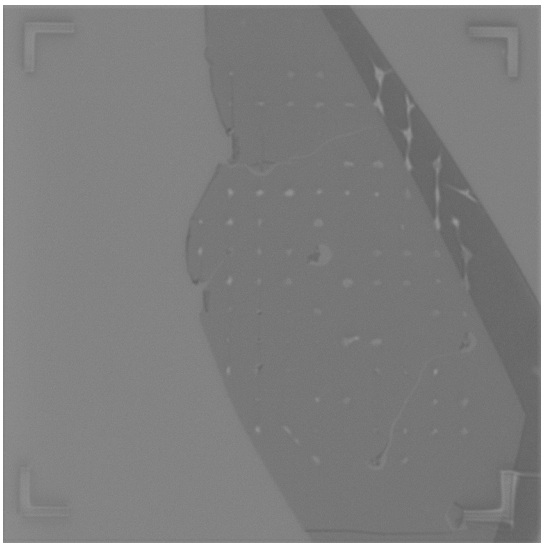

Figure 1: Optical microscopy image of a WSe_2 monolayer transferred over an array of nanopillars. Bright spots indicate tenting sites resulting in local strain in the WSe_2 monolayer.

The nanopillars are fabricated on a SiO_2 substrate via electron-beam lithography (JEOL 6300) using hydrogen silsesquioxane (HSQ) negative resist. The nanopillars are cylindrical with equal height and diameter of 150 nm. The spacing between pillars is 3 μm, which gives enough room for the WSe_2 monolayer to both conform to the pillars while layering on the substrate. To place an exfoliated WSe_2 monolayer deterministically over the fabricated nanopillars, we employ an all-dry transfer technique using polydimethylsiloxane (PDMS) stamps. Figure 1 shows an optical microscopy image of a mechanically exfoliated WSe_2 monolayer transferred on top of a nanopillar array. The tenting of the monolayer over the pillars is visible as bright spots due to increased contrast. Atomic force microscopy imaging confirms the conforming of the monolayer to the substrate and nanopillars.

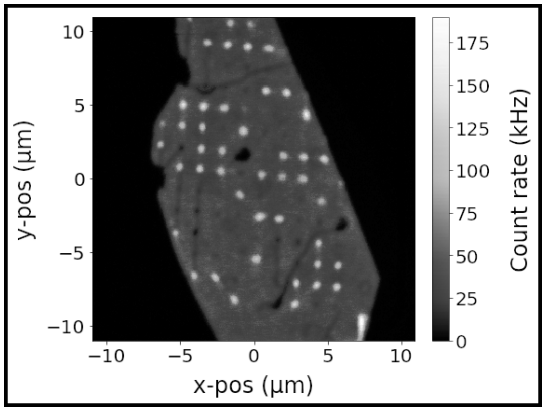

Figure 2: Room temperature photoluminescence scan of the WSe_2 monolayer displaying brighter spots at the nanopillar sites.

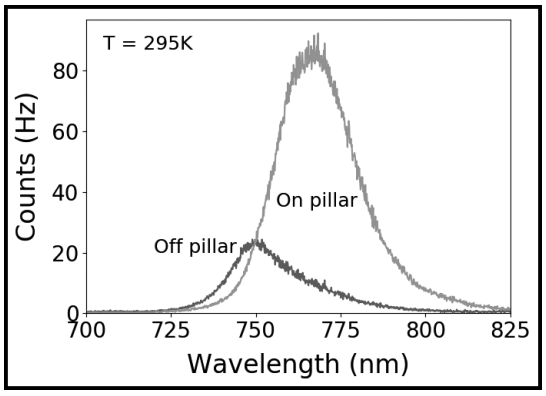

Figure 3: Spectrum of strained and unstrained WSe_2 monolayer. Besides brightness increase, a clear redshift of the emission at the pillar indicates strain that shrinks the band gap.

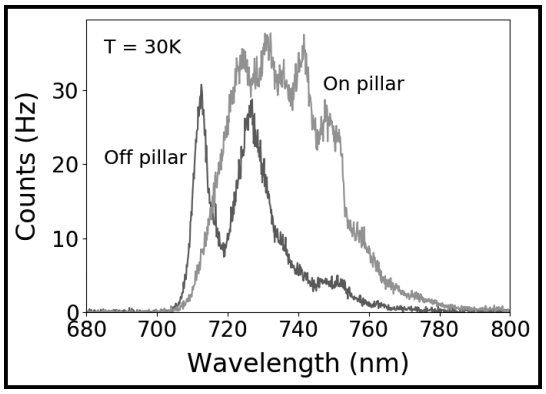

Figure 4: Spectrum of WSe_2 monolayer at T = 30 K. Besides the global redshift, no sharp features characteristic of quantum emitters are observed.

As-transferred WSe_2 monolayers necessitate no further treatment to be investigated optically. We perform spectral and micro-photoluminescence measurements in a confocal setup. Charge carriers are excited with a continuous wave laser at 532 nm with a power density of about 100 nW/µm^2. In the photoluminescence scan of Figure 2, we see a clear brightness increase at the pillar sites, indicating a larger carrier concentration. The strain shrinks the bandgap at the nanopillar sites, thus creating a local well where carriers get concentrated. This is consistent with the results of Figure 3, which shows a redshift of the exciton recombination energy by 35 meV (16 nm) at strained sites compared to unstrained WSe_2. For comparison, this is twice the value reported by Palacios-Berraquero, et al., in [3], which is promising for obtaining quantum emitters.

Quantum emitters in WSe_2 monolayers can only be resolved at cryogenic temperatures. We place the sample in a helium-flow cryostat and cool the sample to 30 K.

As shown in Figure 4, the emission of the unstrained WSe_2 is composed of the neutral exciton at 715 nm, negatively charged excitons at 735 nm and an additional peak at longer wavelengths originating from impurity-bound excitons [3]. At the pillar sites where the WSe_2 is strained, the spectrum is globally redshifted and is broadened towards longer wavelengths due to various local exciton traps naturally more populated at low temperatures. This data does not show correlation between strained sites and sharp emission lines characteristic of single quantum emitters. The temperature of the experiment is likely too high and resulting in a thermal depopulation of narrow exciton traps.

We conclude that the strain gradient created by the pillars is either too weak and/or too broad to confine a single exciton at 30 K but should be sufficient at temperatures below 10 K according to previous reports. Additionally, residual charging of the nanopillars from the electron beam during fabrication might quench the quantum emitters. Future generation of substrates will include electrical gating of the WSe_2 monolayer to control the charge environment of the emitters.

References:

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[2] A. Srivastava, M. Sidler, A. V. Allain, D. S. Lembke, A. Kis and A. Imamo.lu, Nat. Nanotechnology 10, 1038 (2015).

[3] C. Palacios-Berraquero, D. M. Kara, A. R.-P. Montblanch, M. Barbone, P. Latawiec, D. Yoon, A. K. Ott, M. Loncar, A. C. Ferrari and M. Atature, Nat. Comm. 8, 15093 (2017).