Irradiation of Nanostrained Monolayer WSe_2 for Site-Controlled Single-Photon Emission up to 150K

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Abstract: Utilizing strain and defect engineering techniques, a novel method of designing site-specific single-photon emitters in 2D-WSe_2 is developed that achieves emitters with high-yield, purities above 95%, and extended working temperatures up to 150 K.
OCIS codes: OCIS codes: (270.0270) Quantum optics; (350.4238) Nanophotonics and photonic crystals, (160.2220) Defect-center materials.

1. Introduction: In recent years, quantum-dot-like emitters in Two-Dimensional (2D) WSe_2 have been identified as a novel source of quantum light [1–3]. 2D WSe_2, in contrast to conventional III-V semiconducting quantum dots, offer significant advantages, including a high photon extraction efficiency due to its atomic thickness, the potential for scalable, site-controlled and deterministic manufacturability [4], and ease of integration with mature photonic technologies via simple transfer methods [5]. However, a longstanding challenge facing 2D WSe_2 Single-Photon Emitters (SPEs) lies in their low working temperature and thermal instability. The ultra-sharp emission lines associated with SPEs that appear ~50-200 meV below the free exciton of WSe_2 are observable only at cryogenic temperatures and quench above 30K due to low confinement potential and quantum yield [2,6]. Recent studies have attributed the origins of these SPEs to localized intervalley defect excitons that form when the excitonic energies are lowered and overlap with a valley symmetry-breaking defect state at highly strained regions [7]. This reveals the pivotal role of both strain and defects in the creation of SPEs. Therefore, engineering high-quality SPEs ideally requires placement of only a single valley symmetry-breaking defect (to minimize the non-radiative recombination and achieve high quantum yield) at the center of a strain field (to achieve high confinement potential). In this work, by using electron beam (e-beam) irradiation as a controllable method to induce defects in WSe_2, along with engineering strain fields using dielectric nano-pillar structures, we are able to create site-specific SPEs in WSe_2 with high yield, high single-photon purity, and possible working temperatures up to 215 K.

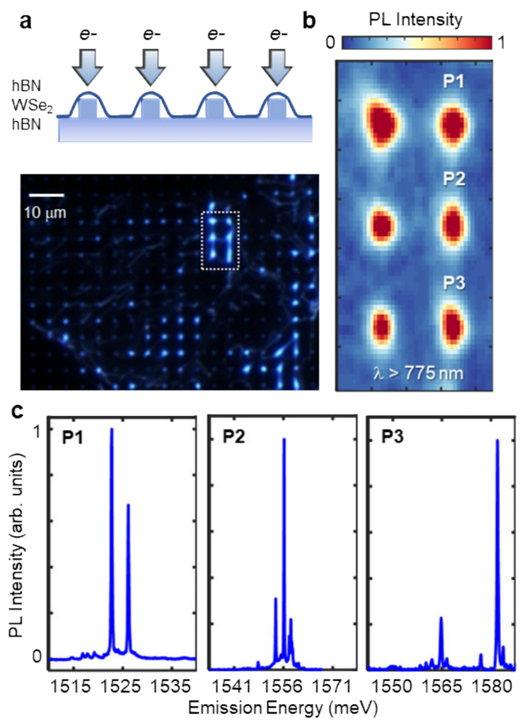

Fig. 1: Site-controlled quantum emitter array via strain and defect engineering. (a) Schematic of the fabricated structure. The bottom figure shows the dark-field image of the sample. Bright emission is observed from the top of the pillars due to enhanced exciton funneling. (b) Integrated Photoluminescence map for wavelengths above 775 nm, showing the bright emissions associated with single-photon emitters from the top of the nano-pillar. (c) Photoluminescence spectrum captured from different pillar sites showing the sharp emission lines associated with SPEs.

2. Results: A monolayer WSe_2 sample encapsulated by h-BN was prepared and transferred onto a Si/SiO_2 substrate with a predefined nano-pillar dielectric array with height and diameter of 200 nm and 150 nm, respectively. Subsequently, the top of the nano-pillar sites was irradiated with e-beam at a highly focused spot size (<10 nm) and an accelerating voltage of 100 keV to introduce crystalline defects. Fig. 1a (bottom panel) presents the dark-field optical image of the final assembled structure where the brightly illuminated regions correspond to the nano-pillar sites. Integrated Photoluminescence (PL) maps (Fig. 1b) of six irradiated nano-pillars (white dashed box region in Fig. 1a) were taken at 5 K for energies below 1.6 eV and provides evidence for the formation of bright emission lines below the WSe_2 free exciton. Fig. 1c shows the PL spectrum at the location of three different irradiated nano-pillars where the emergence of ultra-sharp emission lines associated with SPEs in these materials is evident.

With a closer look at Fig. 1c, it is readily observable that sharp emission lines appear to form in pairs with an energy spacing of about 3-5 meV. The features of these pairs resemble those of an exciton-biexciton where the inherent fine-structure splitting results in two doublets, but the sign of the splitting is reversed in each pair as a consequence of the emission cascade (Fig. 2a). The polarization-resolved PL spectrum of the pairs further corroborates the radiative cascade (Fig. 2b). The integrated intensities of the pairs show distinct sub-linear and super-linear characteristics as previously observed for exciton-biexciton emissions in WSe_2 [8] (data not shown). Note that the physical origin of these exciton-biexciton-like features is still ambiguous; previous studies have shown that such features can also be attributed to the hybridization of defect states with localized excitons [7]. Finally, second-order correlation measurements were performed using a Hanbury Brown and Twiss setup that confirms both emission lines act as single-photon emission sources with purities as high as 90%. (Fig. 2c). Our fabrication method was able to achieve a success rate of over 85% in engineering SPEs per site where the spectral purity of many of the emitters was measured to be above 95% (with an average purity of 92%).

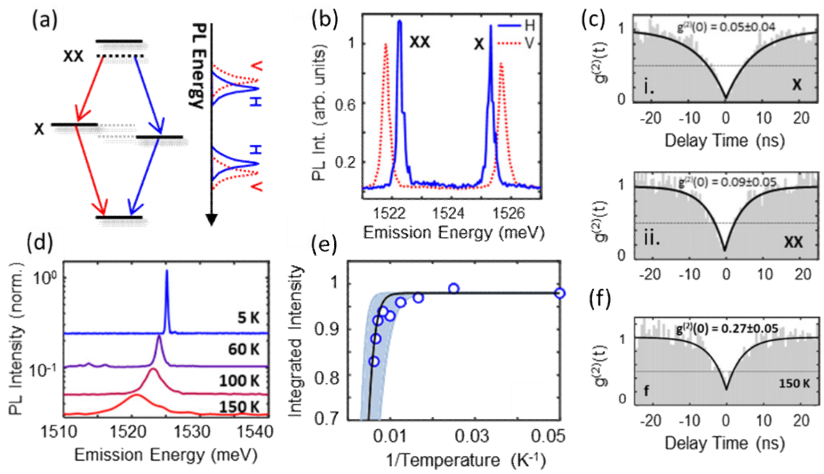

Fig. 2: (a) Cascaded emission of biexciton. (b) PL Spectrum of single-emitter. Two pairs of coupled doublets are observed. (c) Second-order correlation measurement. Both X and XX exhibit $g^{(2)}(0) < 0.1$, showing high purity single-photon emission. (d) Evolution of the PL spectrum as a function of the temperature. (e) Arrhenius plot of the integrated intensity. The PL peak starts to quench around 150 K (f) Second-order correlation measurement at T=150 K exhibiting $g^2(0) < 0.3$, evidencing that the single-photon nature persists up to our maximum measurement temperature of 150 K and likely persists beyond 200 K.

Next, we look at the evolution of the single-photon PL emission from 5 K to 150 K (Fig. 2c). It is readily apparent that the emission line is distinguishable up to 150 K. The relative redshift of the emission line with an increase of temperature is in line with the reduction in the bandgap following Varshni's empirical relationship. The Arrhenius plot of the integrated intensity of the SPE versus temperature shows that the integrated intensity quenches to e^{-1} at approximately 215 K (Fig. 2e). Fig. 2f shows the second-order correlation measurement of the SPE and demonstrates that the emitters exhibit anti-bunching up to 150 K with $g^{(2)}(0) = 0.27$. Given the high average purity (~0.25) of the array of emitters at 150 K in combination with the Arrhenius data, it can be expected that the single-photon nature of the SPEs can last up to 215 K. To our knowledge, no transition metal dichalcogenide SPEs have been able to operate to 150 K without Purcell enhancements.

3. Conclusion: By decoupling the strain and defect engineering processes in the design of WSe_2 single-photon emitters, we were able to achieve high yield in the deterministic positioning of quantum emitters with high purity (95% at 5 K) that can preserve their single-photon nature to above 150 K. This method was also successful at deterministic engineering of exciton-biexciton-like features with a radiative cascade that can pave the way for the realization of entangled photon-pair sources. It can be expected that by utilizing the methodology described in this work, and integration with plasmonic cavities, our work sets the stage for future scalable elevated-temperature high-quality 2D WSe_2 quantum light sources.

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