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Exciton Diffusion in a Monolayer MoS₂-WS₂ Lateral Heterostructure

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Abstract: In this paper, we observed preliminary exciton diffusion signature in a WS₂-MoS₂ lateral heterostructure at room temperature. Within center MoS₂ the diffusion appears to be 300 nm, along the WS₂-MoS₂ edge the diffusion appears to be more than 2 μm.

OCIS codes: (250.5230) Photoluminescence; (160.6000) Semiconductor materials; (300.6250) Spectroscopy, condensed matter

1. Introduction

Monolayer transition metal dichalcogenides (ML-TMDC) is a class of 2-D semiconducting quantum materials with a direct optical band gap, optical-robust, and with the potential of substantially altering optoelectronic devices, and define new classes of devices based on defect induced quantum phenomena such as charge density waves or single photon emission. Unique features such as large exciton binding energy (~ 500 - 800 meV), high exciton oscillator strength (~20% per monolayer) and short radiative lifetime (~ 100 fs to several ps) lead to effective light-matter interaction at room temperature [1]. In addition, multiple active layers can be embedded in Van der Waals heterostructures to provide a new platform to discover and eventually control the vast physics of exciton dynamics [1]. The dynamics of exciton diffusion in individual ML-TMDC were studied by various papers using pump-probe microscopy [2-4]. The carrier lifetime of typical ML-TMDC (e.g. MoS₂ and WSe₂) is in the ps timescale and the diffusion coefficient is about tens of cm²/s [2-4]. However, the transport across the boundary in the ML-TMDC lateral heterostructures and the diffusion confined along the thin edge of the outer layer is neither studied nor understood.

In this paper, we present some preliminary exciton diffusion measurements in a MoS₂-WS₂ lateral heterostructure. Across-the-boundary diffusion and long exciton travelling length (~2 μm) along the edge is observed.

2. Sample characterization and experiment procedure

The basic optical setup to study the exciton diffusion in this paper is a “custom-built” optical microscope with the additional capability of spatially, spectrally and temporally resolving the collected photoluminescence (PL). 2D WS₂-WSe₂ and MoS₂-WS₂ lateral heterostructure samples are prepared by chemical vapor deposition (CVD) method on Si substrate. The “hexagon” sample shape is captured by optical microscopy (fig. 1a-b). Raman spectroscopy using a 532 nm laser is used to distinguish the individual components (fig. 1c-d). PL spectrum of WS₂-WSe₂ collected at the boundary of two components clearly shows emission from both WS₂ at 640 nm and WSe₂ at 760 nm (fig. 1e). For a

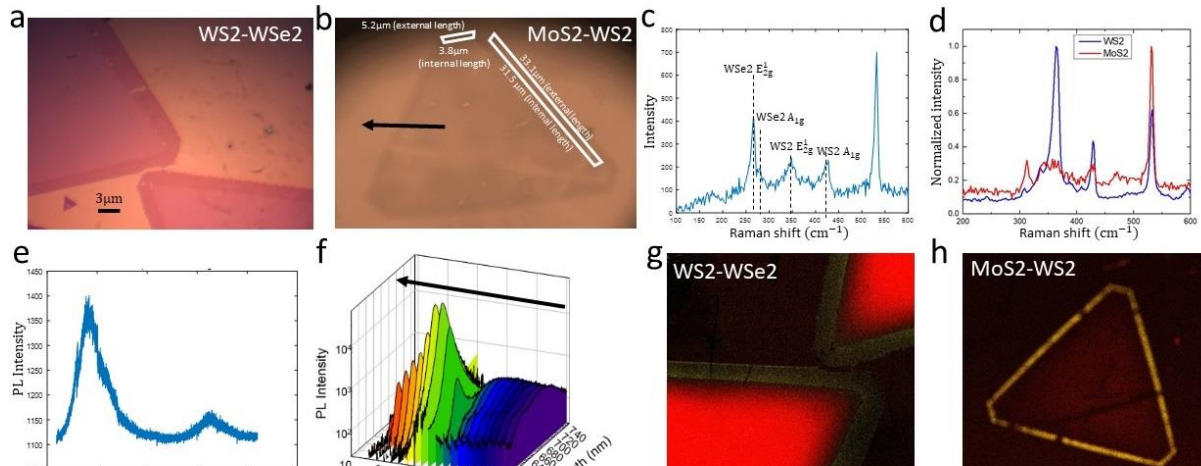


Fig. 1 Optical characterization of 2D lateral heterostructure samples. (a, b) optical image of WS₂(center)-WSe₂(edge) and MoS₂(center)-WS₂(edge) lateral heterostructure flakes. The black arrow is the laser scanning direction for (f). (c, d) Raman spectrum of heterostructures show individual spectra of each components. (e). PL spectrum of WS₂-WSe₂ heterostructure flake at the boundary show emission from both WS₂ at 640 nm and WSe₂ at 760 nm. (f). Series of PL spectrum collected from MoS₂-WS₂ heterostructure flake along the black arrow direction in (f). (g, h). PL mapping of WS₂-WSe₂ and MoS₂-WS₂ flakes. Red/yellow are the center/edge regions. A 532 nm laser is used as the excitation source for Raman and PL measurement. A 594 nm laser is used for PL mapping. All data are collected at room temperature.

MoS₂-WS₂ flake, a 532 nm laser is scanned along the black arrow in fig. 1b starting over MoS₂ across the heterostructure into the WS₂. The resulting series of PL spectrum show over the MoS₂ a low-intense and broad band around 690 nm at the center and a pronounced sharp band at 638 nm at the edge which is about 30 times stronger than the 690 nm emission (fig. 1f). The center MoS₂ region showing the low-intense broad emission band (low quantum yield) is mostly likely associated with defect emission. The confocal PL mapping near the diffraction limit by using oil-immersed objective (NA=1.4) are performed for both 2D lateral heterostructure samples to clearly resolve the cracks and defects on the flakes (fig. 1g-h). The emission from center and edge region are collected by two different detectors with different pre-set detection range corresponding to the center and edge emission wavelength. The exciton diffusion measurement is performed by exciting at center (MoS₂), as well as near the long/short edge (WS₂), and at the long edge (WS₂) on a flake. The spatial extend of the PL emission away from the excitation spot is collected. The laser and PL emission line profile are compared to study and characterize the extended PL emission profile.

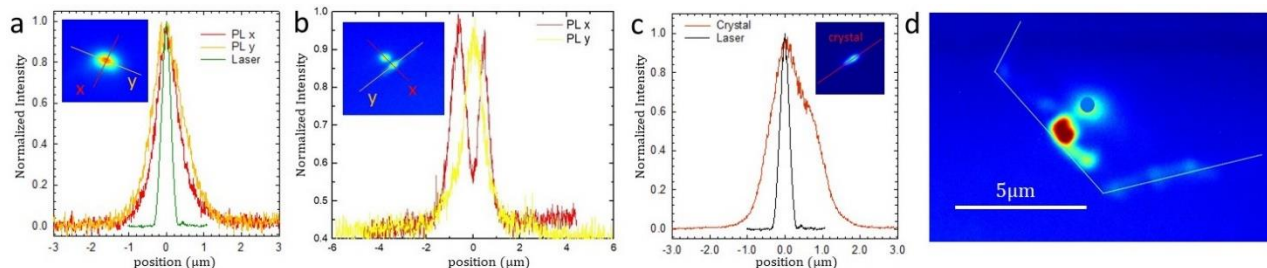


Fig. 2 Exciton diffusion measurement of a MoS₂-WS₂ lateral heterostructure sample. (a-c). CCD image of PL emission (inset) and their corresponding line profiles along specified directions collected by excitation only at the center (a), near the long edge (b) and at the long edge (c). (d). CCD image of PL emission collected by excite only near the short edge. The excitation spot is highlighted with a blue dot. A 532 nm laser is used as the excitation source. A550 nm long pass filter is used to filter out the laser before the CCD camera. All data are collected at room temperature.

3. Results and discussion

The confocal PL mapping of WS₂-WSe₂ and MoS₂-WS₂ lateral heterostructure flaks clearly resolve the PL emission from center and edge (fig. 1g-h). Cracks and defects on the flake of the PL mapping agree with the optical image in fig. 1a-b. The exciton diffusion measurement on a MoS₂-WS₂ flake is captured by a CCD camera while a circular laser spot of FWHM ~267 nm is shining on the center (fig. 2a), near the long edge (fig. 2b), at the long edge (fig. 2c) and near the short edge (fig. 2d) of a MoS₂-WS₂ flake. Fig. 2a shows a significant and asymmetric extent of the PL emission profile from circular laser profile to an elliptical PL profile in the x and y direction of 654 nm and 940 nm. Deconvolving the Gaussian FWHM ~ 940 nm in y direction with the excitation spot translates into a diffusion length of over 350 nm. When moving the excitation spot closer to the long edge, the PL emission map is further centered towards the WS₂ edge region and once close to the heterojunction the PL profile propagates along the edge as shown in fig 2. b. In the latter case (-excitation spot at the long edge), the PL emission map propagates along the edge up to 2μm away from the excitation spot while staying confined in the edge region (fig. 2c). Another interesting phenomenon is observed when pointing the excitation close to the short edge: several spots on the edge light up in a fragmented way that reflects the irregularity of the edge and propagate along the long edge again (fig. 2d). The possibility that the extended PL is caused by laser Airy ring in all measurement is ruled out by directly comparing the laser and PL profile. Regarding to the observed long diffusion length, we could propose two hypotheses. Firstly, charge transfer excitons with typically lower effective mass and binding energy are converted from excitons when they diffuse along the band structure at heterojunction so that the hole is in one material and the electron remains in the other. The larger diffusion length observed could be conceivable because of their longer lifetime. Secondly, the excitons could get separated in the presence of defects levels and specifically, at the heterojunction. The diffusion and luminescence observed could due to the charge diffusion within the material and the charge recombination. This work provides some insights of pronounced exciton diffusion with directional preferences in a defect-populated 2D lateral heterostructures that is not been reported so far. We will work systematically to further study the exciton physics in depth. A brand-new clean (TSFI-treated) sample will be prepared and compared with current results. To study how the impurity scattering can influence the diffusion processes, low temperature experiment will be performed to suppress the phonon contribution. Time resolved pump-probe spectroscopy will also be performed for study of the diffusion dynamics in the future.

4. References

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