

2D Photoluminescence Mapping of Transition Metal Dichalcogenides

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Abstract

Transition Metal Dichalcogenide (TMD) crystals are semiconductors with properties that make them promising candidates as electronics components. In particular, bulk TMD material has an indirect band gap while monolayer (1L) material transitions to a direct band gap [4]. To create atomically thin (including 1L) samples of TMD crystals on silicon substrates, the “scotch tape” method is employed. The resulting samples are then characterized using an imaging spectrometer to create a 2D map of the photoluminescence, which allows identification of the monolayer regions. In future work, multidimensional coherent spectroscopy will be done on the 1L regions to learn more about the linewidth of 1L TMD material.

1 Introduction

Transition Metal Dichalcogenide (TMD) crystals such as WSe_2 exhibit unusual band structure in that bulk material (that is, two or more layers) have an indirect band gap, while monolayer (1L) crystals have a direct band gap [4]. These and other properties, such as the accessibility of K and K' valleys by circularly polarized light [5], make WSe_2 and other TMDC materials appealing as possible components in electronics.

To better understand these materials and their homogenous linewidth in particular, 2D coherent spectroscopy provides a valuable tool. In 2D spectroscopy, four-wave mixing measurements are taken and the Fourier transform of the output delay is plotted as a function of the Fourier transform of the time delay between the first two input pulses [1]. In simpler terms, a plot of absorption frequency v. emission frequency is created and can be used to learn more about the homogeneous and inhomogeneous components of broadening in the crystal's linewidth.

However, before such measurements can be taken, we must create a sample to measure. Although both chemical vapor deposition and mechanical exfoliation are viable routes for creating monolayer TMD crystal, the properties of the monolayer have been found to depend on the method used [3]. Moreover, the homogenous linewidth has been measured for CVD-grown WSe_2 , but has not yet been measured for a mechanically exfoliated sample [2].

Optical microscopy allows us to examine WSe_2 material and identify probable monolayers. To confirm these regions really are 1L, we measure the emission spectrum of this area of the sample when illuminated by a 532 nm laser. Because of the direct band gap structure for 1L, only the 1L regions to emit photoluminescence, so the presence of an emission peak on the candidate area (and the lack of such a peak anywhere else) will confirm that the area is 1L and thus suitable for use in 2D spectroscopy to determine the homogeneous linewidth of monolayer WSe_2 .

2 Procedure

We used the “scotch tape” method of mechanical exfoliation to thin the bulk WSe₂ down to few- and mono-layer thickness. We placed a small amount of a WSe₂ crystal on one a piece of tape. We then repeatedly stuck that piece of tape on another piece of tape, taped sticky-side up to a glass slide, rubbing on top of the upper tape lightly with a finger or other rounded object each time to ensure good contact and transfer. Once the tape on the slide appeared well populated with WSe₂, we placed a small (5 mm by 5 mm) square of PDMS on a second glass slide (ensuring no air bubbles were underneath) and then placed that slide PDMS-side down on the WSe₂ tape. We held the two slides in place for approximately 10 seconds, applying slight pressure, then carefully separated the slides.

We now repeated the process to transfer the WSe₂ to the silicon substrates. We first cleaned the substrates by swiping the surface with lens paper wetted with acetone, then placed the substrate face-down on the PDMS. We heated the glass slide, PDMS, and silicon on a hot plate to around 100° C for 60 seconds, then removed the silicon for imaging with the optical microscope.

For the photoluminescence measurements, we used a Horiba iHR550 with a Synapse CCD. A 532-nm laser was sent in normal to the sample, and the reflected light was filtered with a 600-nm longpass filter before entering the spectrometer.

3 Results

Imaging with the optical microscope allowed identification of probable monolayer regions based on the Fabry Perot effect. Different WSe₂ thicknesses cause the microscope’s white reflected light to constructively interfere at different wavelengths so that the crystals are colored according to number of layers. Figure 1 shows one such image with a monolayer region of approximately 100 square microns.

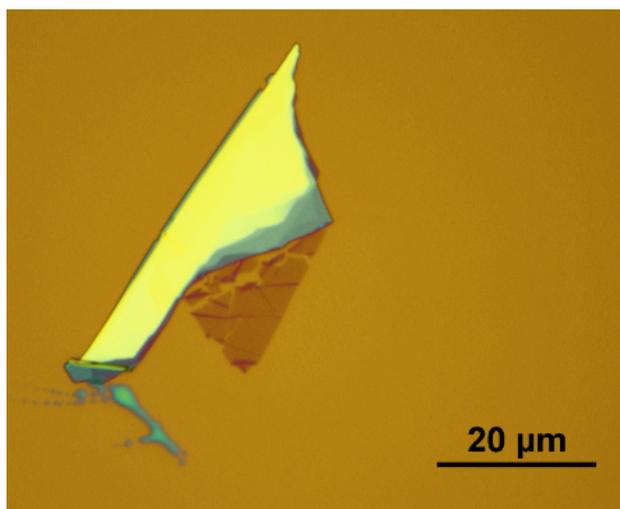


Figure 1: 100X image from an optical microscope showing a monolayer region as well as thicker WSe₂; the two are distinguishable by color thanks to the Fabry Perot effect.

We located the monolayer candidate on the substrate by imaging with the laser spot and a CCD and were able to detect a PL signal at room temperature. Figure 2 shows the peak we saw on the sample; when we moved the beam off the monolayer region, the peak disappeared.

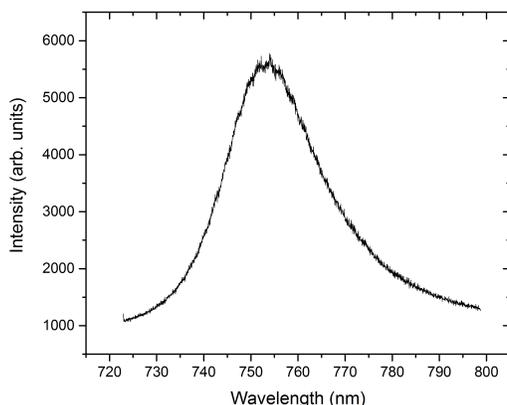


Figure 2: The emission spectrum of the candidate monolayer WSe_2 as measured by a Horiba iHR550 imaging spectrometer. Note that the peak, which centers around 750 nm, disappeared when the beam was moved off the sample.

4 Conclusions and Next Steps

Given that we were able to detect a signal at room temperature, and given that the literature reports a significant increase in signal when cooled, we expect that we could increase our signal by cooling with helium or even nitrogen. If need be, we could use this improved signal and already-written code to take data and create a 2D map of the sample (in LabVIEW and Matlab, respectively). However, this is not necessary to move on to the four-wave mixing measurements.

Now that we have confirmed the presence of a monolayer region on the order of several microns in diameter, we can begin probing the sample with four-wave mixing to create 2D spectra. From the 2D spectrum, we can extract the homogeneous and inhomogeneous linewidths and compare with the measured values for CVD-grown WSe_2 .

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