An Investigation into the Properties of Magic Angle Twisted Bilayer MoS2

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Abstract
Molybdenum disulfide (MoS2) is an inorganic van der Waals crystal composed of molybdenum and sulfur that can be exfoliated to one atom thick levels. MoS2 is a semiconductor and provides strong-light matter interactions. If two separate monolayer MoS2 are placed vertically with small angle twist between the crystals (~5º, also known as magic angle), one can obtain strongly correlated states. In this research project, we have fabricated magic-angle twisted bilayer MoS2 (MATB) by using a deterministic transfer process. We are studying MATB samples by using nondestructive micro-Raman spectroscopy to determine how strong correlation between charge carries modify the crystal vibrations.

Introduction
It has been known for some time that MoS2 exhibits semiconductive properties when exfoliated to a single layer, but recent models have shown that bilayer MoS2, with the layers having a slight 4º to 5º (magic angle) offset may have properties arising from the altered periodicity of the material. In particular, the magic angle is thought to change the spectrum of MoS2 by either shifting the existing peaks in amplitude or wavelength or by adding a third peak. Raman spectroscopy is used to analyze this spectrum and search for any added peaks or effects arising from this offset. The most immediate application of this research would be device fabrication and photodetection, with other possible applications in nano and opto electronics.

Method
The samples of MATB are prepared through a meticulous multistep process starting from bulk MoS2, and ending with MATB. First, the bulk MoS2 is exfoliated down to a monolayer via mechanical exfoliation onto SiO2. Then once two monolayers are identified the transfer process can begin. This process starts by cutting a small piece of Polyethylene terephthalate (PET) and using it, with the help of the transfer stage, to pick up one of the monolayers from its piece of SiO2. Then the monolayer on the PET is lowered onto the second monolayer but with a 4 to 5 degree twist angle (the so called ‘magic angle’). This stack is then transferred onto a new piece of SiO2. The sample is then washed with Dichloromethane (DCM) to remove the PET. The new sample is then analyzed via Raman spectroscopy to search for the unique signature of the magic angle in the sample’s spectrum. This process starts by first calibrating the Raman spectrometer by taking the peak position of pure SiO2, and from this determining an offset. Once calibration is done the sample’s spectrum can then be taken. This Raman data is then analyzed in python to search for the magic angle signature.

Results
1) Exfoliation of MoS2

The peak position of MoS2 at ~250 cm-1 is shown in blue and the Sulfur atoms shown in yellow. Figure 1: Atomic structure of monolayer MoS2, pictured above with the Molybdenum atoms shown in blue and the Sulfur atoms shown in yellow.

2) Exfoliation of MoS2

The peak position of pure SiO2 at ~450 cm-1 is shown in blue and the Sulfur atoms shown in yellow. Figure 2: Side view of MATB, with the substrate (SiO2) pictured in deep blue, the first layer of MoS2 pictured in light blue, and the twisted layer pictured on top in medium blue.

3) Monolayer MoS2 and MATB

Figure 3: Pictured left: Overview of dry transfer stage, with microscope in the center of the frame, and the heated stage with turntable directly beneath. Pictured above: Schematic diagram of dry transfer stage with microcontroller in yellow, microscope in purple, and stage in grey.

4) Scanning Raman data and Contour plot

Figure 4: Pictured left: Scanning Raman setup with sample stage center frame, objective lenses directly above, and fiber optic cable in blue in the background. Pictured below to the left: Schematic diagram of scanning Raman setup. Note that it has been simplified for clarity. The scanning Raman setup used for data acquisition uses alternatively 300 nm and 1200 nm gratings; 1200 nm for higher resolution, and 300 nm for higher range.

Figure 5: Pictured left: Monolayer of MoS2 at 50x magnification. Pictured right: Resulting MATB sample at 20x magnification. Notice the change in the MATB’s coloration in the twisted bilayer region.

Figure 6: Pictured left: Scanning Raman data of twisted bilayer MoS2 at Magic angle. Pictured right: Contour plot of sample in Fig. 5 with twisted region in green/yellow. The color bar denotes the number of counts.

Summary and Conclusions
Throughout the course of this research, it has been shown we are able to exfoliate MoS2 down to one layer through mechanical exfoliation, then using dry transfer techniques and our own self-made transfer stage, construct twisted bilayer MoS2, with the required magic angle. Then with our scanning Raman setup we have been able to attain and analyze the unique Raman signatures of not only bilayer MoS2 but twisted bilayer MoS2 as well, and have made headway into identifying the features associated with the 4º-5º degree magic angle.

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References