Light and Matter Interactions in TMDC Systems

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I. Introduction

In recent years, research into the quasiparticle, polariton, has produced some promising results with regard to the creation of Bose-Einstein condensates (BEC). These polaritons offer some distinct experimental advantages which allow for the possibility of BECs at room temperature thus leading to future applications [4]. One area of materials which has grown in interest due to its help with the creation of polaritons is transition metal dichalcogenides. TMDCs have been found to have strong excitonic properties which are necessary for the creation of polaritons [2, 4]. These TMDCs offer a range of useful properties for a variety of light-matter interaction experiments and can be obtained through various fabrication techniques, each with their respective advantages and disadvantages.

A. Transition Metal Dichalcogenides

Transition metal dichalcogenides (i.e. TMDCs or TMDs for short) are a group of layered materials of the form MX_2 where the M represents a transition metal – such as Molybdenum or Tungsten – and the X represents a chalcogen from group sixteen of the periodic table like sulfur or selenium. The resulting compounds, like $MoSe_2$ or WSe_2, have a hexagonal structure and obtain some typical and some unique qualities of semiconductors when they make the transition from bulk material to monolayers (two-dimensions).

Having a monolayer thickness of about 0.6-07 nm, two-dimensional TMDCs transition to having a direct band gap in their energy band structure while their bulk material counterparts have indirect band gaps [2, 3]. On top of their direct band gaps, 2D TMDC semiconductors have fairly high charge carrier mobility, as well as optical sensitivity and other beneficial electronic properties [3]. These materials have now become a focus of increasing interest in the field of polariton physics where they are being used as quantum wells to form polaritons, due (at least partially) to the fact that they have an exciton binding energy of up to 500 meV [5].

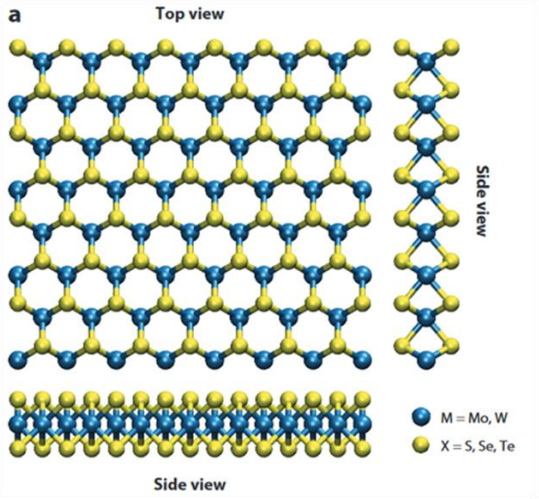

Figure 1. Top and side view of TMD structure courtesy of [2].

B. Light-Matter Interaction: Polariton Physics

A polariton is quasiparticle of light and matter formed by the strong coupling of a photon and an exciton (a bound electron-hole pair) [4]. To create a polariton in our lab, a TMDC sample in a microcavity is hit by a laser to excite electrons in a given material to the conduction band and create holes in the valence band. If an excited electron relaxes to the conduction band minimum and binds with a hole at the valence band maximum, it will create an exciton and release a photon into the microcavity. The photon will then bounce off of the distributed Bragg Reflectors (DBRs) parallel to the TMDC layer to keep it inside of the cavity, and if it binds with the exciton it will create a polariton.

These polaritons possess two main properties which give them an experimental advantage over regular excitons – at least for the formation of Bose-Einstein condensates (BEC). A polariton has an effective mass of roughly four orders of magnitude smaller than an exciton, and the phase coherent wave from the photon is easier to extend through the structure of the crystal despite any potential crystal defects [4]. Together, these two properties ultimately make it easier to produce BECs at room temperature. I did not participate much with the various exciton and polariton experiments directly; instead, my main contribution to the various experiments was from the fabrication of TMDC monolayers and the preparation of samples which would then be studied.

II. Sample Preparation and Characterization

There are two different main ways to create layers of TMDCs: the top-down method (exfoliation), or the bottom up method – also known as chemical vapor deposition (CVD). While the CVD method can generally create larger, and more uniform samples, the method of mechanical exfoliation – nicknamed the 'scotch tape' method (though that is a bit of a misnomer for our research group since we never used scotch tape specifically) – has proven to yield higher quality monolayers at relatively small sizes [3]. In the future, new methods for producing larger size samples, generally on the scale of >10 μm, will greatly aid this kind of research, but for now our lab used the mechanical exfoliation method.

Mechanical exfoliation is the process of using some kind of adhesive, in our case PDMS, to quickly remove flakes of a material and then further peel off thinner flakes until it is suspected that monolayers might be present (usually due to a slight change in color when light is reflected). We then placed our exfoliated samples onto a substrate such as silicon. In order to check for potential monolayers, an optical light microscope was used to scan substrates.

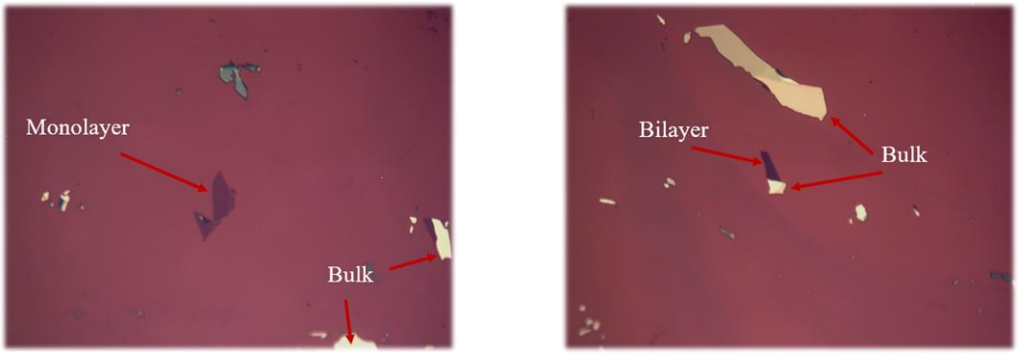

Figure 2. Monolayer and bilayer samples (respectively) of WSe_2 on a Si substrate with bulk samples for reference.

In terms of color, monolayers generally almost blend into the background of the substrate that they are exfoliated onto. Beyond using the optical microscope to check for monolayers, once images like those in Figure 2 are recorded there are a couple of ways to further check if they are actually monolayers. The first involves a computer program to compare the contrast between the layer and the background. If the contrast is roughly 2-4% it is a monolayer; if it is more than that, the sample is most likely a bilayer or thicker. The other dependable method is to use a photoluminescence test with a laser and spectrometry. Monolayer TMDCs have sharp and narrow peaks at known wavelengths (depending on each material) and if the measured peaks match the known values, a monolayer is confirmed.

III. Sample Transfer Procedure

In order to transfer the fabricated monolayers from one substrate to another or onto a DBR to create a quantum-well optical cavity microstructure, a specific microscope set up was employed. The setup, shown in Figure 3, consisted of an XY stage which allowed us to move a substrate to the desired position – such as to find the desired monolayer with the microscope – and then use the XYZ stage on the left-hand side of the figure to move a transparent PET adhesive stamp above the monolayer in question. When both the stamp and the desired sample are in view of the microscope, the temperature of the heat plate can be raised to around 40° C to weaken the Van der Waal bonds of the atoms of the monolayer. This allows the stamp to pick up the monolayer or other samples so that they may be placed later on the desired substrate or DBR.

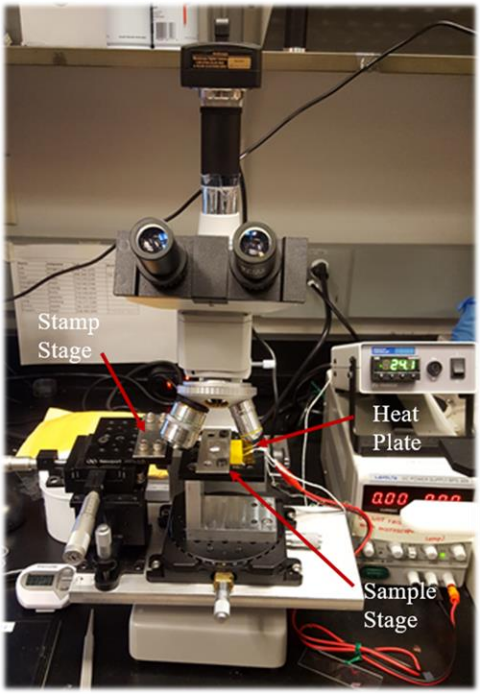

Figure 3. Microscope set-up for sample transfer.

IV. Light-Matter Interaction: Coherent Perfect Absorption and Looking Forward

About halfway through the summer, my mentors and I became interested in the topic of coherent perfect absorption and how our work with monolayer TMDCs might be used with it. Coherent perfect absorption (CPA) can occur when two counter propagating waves (lasers) are incident on some kind of thin film – in our case, a monolayer of $MoSe_2$. In order for CPA to be achieved, the two beams must be of equal intensities and must be interfering destructively with

the sample placed at an antinode [1, 6]. If these conditions are met, the sample can absorb up to 100% of the incoming light.

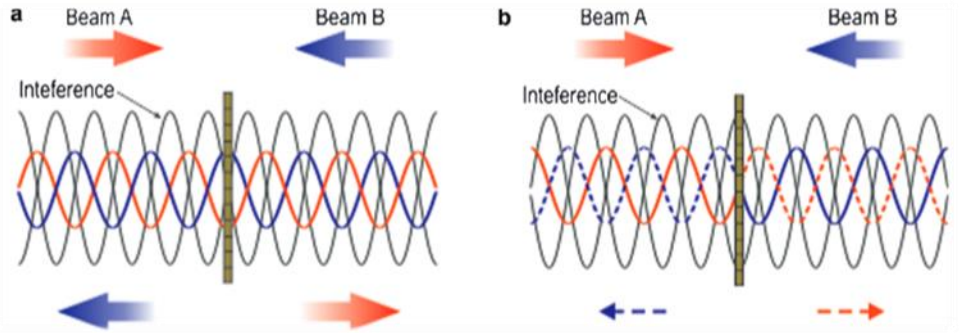

Figure 4. *a)* Destruction wave interference and *b)* constructive wave interference. Image courtesy of [6].

Figure 4, *a*), shows two counter-propagating waves interfering destructively, and Figure 5 shows our CPA setup. Our laser was a red 750 nm beam (approximately 100 μW) split into two beams at the first beam splitter (where the blue arrow begins) in order to pass through our $MoSe_2$ sample from opposite directions. I transferred the sample onto a double-polished sapphire substrate – a transparent substrate – and attached it using PDMS onto an aluminum sample holder which I had drilled a hole through in order to avoid as much unwanted interference as possible. Our beams were then aligned to strike the same spot on our samples, and therefore the same spot on our camera and photodetector. Though most research is still in the early stages, some potential applications of CPA that have been discussed so far are related to sensing and molecular detection, photovoltaics, and even radar cloaking [1]; however, experiments with CPA have not been performed with TMDCs before now.

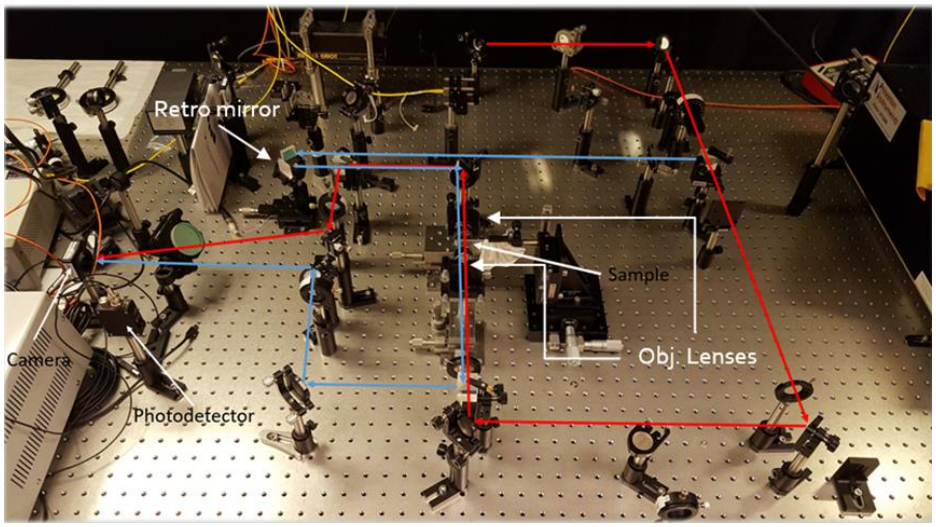

Figure 5. CPA experimental set-up, red and blue arrows indicating the path of the beams leading to a camera or photodetector.

Figure 6 demonstrates our preliminary results. In Figure 6 *a)*, once background light was subtracted, the intensity of the combined beams resulted in a sinusoidal pattern where the average maximum intensity divided by the average minimum gave a result of 3.43. This suggests that our lasers were properly aligned and were at roughly identical intensities. When the average

maximum intensity was divided by the average minimum for the monolayer (Figure 6 *b)*), a result of 1.5 was obtain – a similar value of 1.27 was obtained for the same procedure on a bulk layer. More work will need to be done in order to fully understand these results pertaining to CPA on two-dimensional TMDCs.

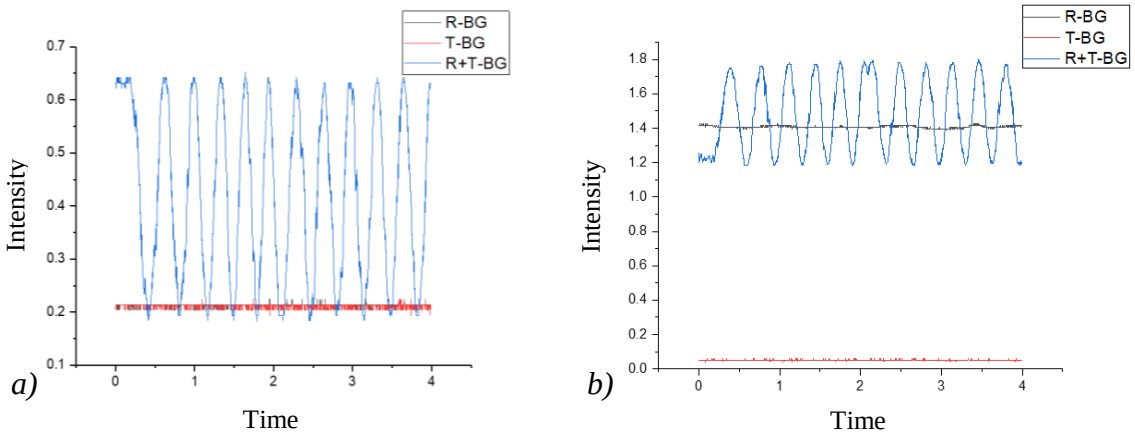

Figure 6. Preliminary intensity results of the two beams on the double polished sapphire substrate in *a)* and on the $MoSe_2$ sample in *b)*.

As work like this continues, there are two main areas to look at: one relating to the CPA experiment specifically and one relating to the experiments of the research group as a whole. Regarding the CPA experiment, we were unable to move beyond initial testing of our $MoSe_2$ sample due to time constraints. In the future, more work can be done with $MoSe_2$ as well as with samples of different materials to observe if they each exhibit similar phenomena. As for the research of TMDCs and all of their related experiments, there is the need to search for new methods of exfoliation. As previously mentioned, mechanical exfoliation tends to yield higher quality samples; however, mechanical exfoliation – at this point – does not have a high reliability for creating uniform samples of larger sizes. Therefore, there is a need to experiment with different exfoliation techniques – perhaps as different temperatures, or speeds of exfoliation, or using different adhesives.

V. Acknowledgements

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