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Acquisition of Closed-Cycle Optical Cryostat to Investigate 1D and 2D Material Plasmonic Structures

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14. ABSTRACT
This DURIP project funded the acquisition of a closed-cycle optical cryostat that was installed in winter of 2018. Specifically, we purchased a Montana Instruments Cryostation that is cable of sample temperatures down to 3.5 K, with electrical feedthroughs and high NA optical access. The cryostat has been in constant use since its installation and continue to be used in the study novel 1D and 2D material (WSe2, graphene, boron nitride, carbon nanotube) structures for applications ranging from nonlinear plasmonic to single photon emitters for quantum information applications. The PI (Schaibley) is currently leading the project 'Plasmonic Amplification through On-Chip, Four-Wave Mixing in Hybrid 2D Material Plasmonic Structures,' funded by AFOSR-YIP 2017 (FA9550-17-1-0215). The PIs (Schaibley and LeRoy) also have a current AFOSR pilot project entitled: 'Towards Single Photon Transistors with 1D and 2D Materials' (FA9550-18-1-0049). Our research has applications to the development of novel optoelectronic and computing device technologies which is crucial to maintaining the technological, scientific, and cybersecurity superiority of the United States. The PI (Schaibley) is also engaged in a collaboration with Dr. Josh Hendrickson (AFRL) to investigate both 2D material plasmonic structures and single quantum emitters, and is writing a proposal to send a student intern to work at AFRL in 2020 through the NSF INTERN program. This DURIP has allowed for the construction of a dedicated optical setup that has greatly enhanced the productivity of our AFOSR research projects. Specific accomplishments include demonstration of a 2D semiconductor plasmonic modulator, and evidence of charging of moire excitons in bilayer heterostructures for deterministic integration with carbon nanotube plasmons.

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1. Project background:

This DURIP project funded the acquisition of a closed-cycle optical cryostat that was installed in winter of 2018. Specifically, we purchased a Montana Instruments Cryostation that is cable of sample temperatures down to 3.5 K, with electrical feedthroughs and high NA optical access. The cryostat has been in constant use since its installation and continue to be used in the study novel 1D and 2D material (WSe_2, graphene, boron nitride, carbon nanotube) structures for applications ranging from nonlinear plasmonic to single photon emitters for quantum information applications. The PI (Schaibley) is currently leading the project "Plasmonic Amplification through On-Chip, Four-Wave Mixing in Hybrid 2D Material Plasmonic Structures," funded by AFOSR-YIP 2017 (FA9550-17-1-0215). The PIs (Schaibley and LeRoy) also have a current AFOSR pilot project entitled: "Towards Single Photon Transistors with 1D and 2D Materials" (FA9550-18-1-0049). Our research has applications to the development of novel optoelectronic and computing device technologies which is crucial to maintaining the technological, scientific, and cybersecurity superiority of the United States. The PI (Schaibley) is also engaged in a collaboration with Dr. Josh Hendrickson (AFRL) to investigate both 2D material plasmonic structures and single quantum emitters, and is writing a proposal to send a student intern to work at AFRL in 2020 through the NSF INTERN program. **As described below, this DURIP has allowed for the construction of a dedicated optical setup that has greatly enhanced the productivity of our AFOSR research projects.**

2. Description of system and integration with existing equipment:

We purchased a Montana Instruments Cryostation closed-cycle optical cryostat that can achieve sample temperatures of 3.5 K to 300 K, can be integrated with a high NA objective (at least 0.6) and has sub-micron sample positioning capabilities. The system is cryogen free with zero operating costs, has best-in-the-class vibrations of <5 nm, and best-in-the-class temperature stability of <10 mK peak to peak. It also includes 12 electrical feedthroughs allowing for gate voltages to be applied to the sample structures. It is a fully automated system which allows for students to be quickly trained and will facilitate high productivity.

A photograph of the cryostat system is shown in Figure 1. It consists of the closed-cycle optical cryostat system integrated with a dedicated setup for optical and plasmonic measurements. The system was installed in the PI's (Schaibley) state-of-the-art optics lab is equipped with **two** continuous wave high resolution (50 kHz linewidth) Ti:sapphire lasers, (M^2 Solstis), a Coherent 900D (120 fs pulse width) Ti:sapphire laser system, and two Andor CCDs and spectrometers. The lab now has three optical tables with three separate cryostat systems allowing for independent experiments to proceed in parallel. All three lasers are tunable from 700 nm to 1000 nm. The lab has a time correlated single photon counting system for time resolved photoluminescence and g^2 measurements. The lab is also equipped with numerous diode laser sources, modulators, optical and electrical components.

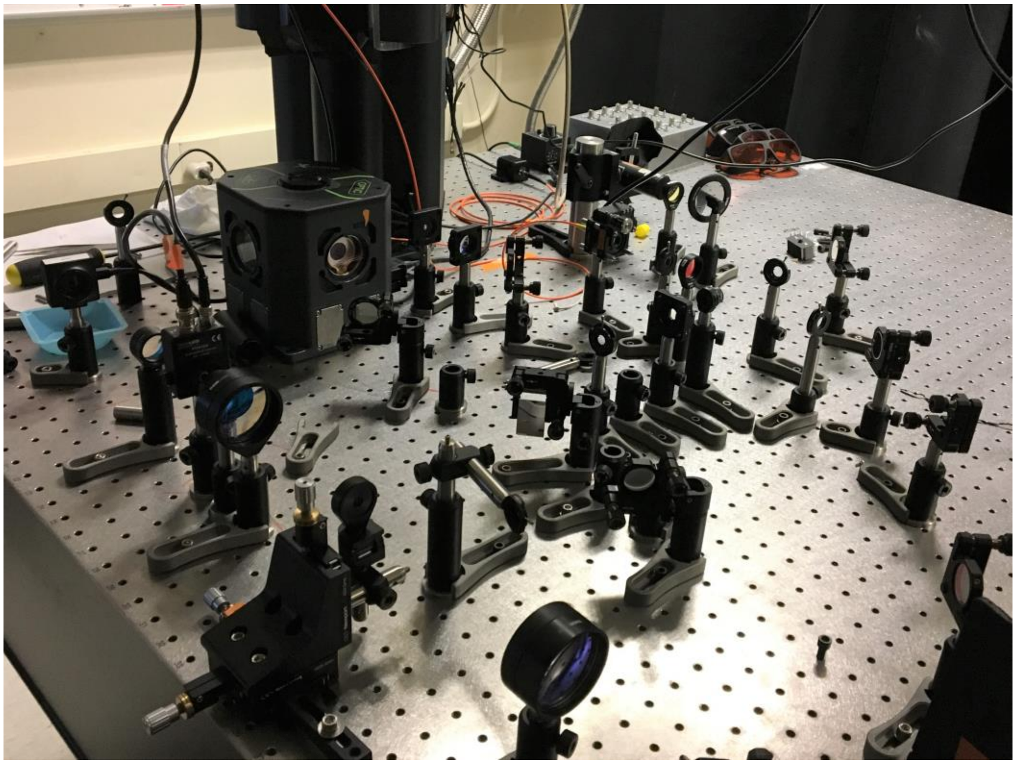

Figure 1| Photograph of the integrated cryostat setup.

3. Description of AFOSR projects that have been enhanced from the DURIP:

3.1 Schaibley- "Plasmonic Amplification through On-Chip, Four-Wave Mixing in Hybrid 2D Material Plasmonic Structures"

The objective of this project is to demonstrate plasmonic amplification using 2D materials as nonlinear plasmonic elements. Since the DURIP award, we have been able to construct a dedicated optical setup (Figure 1), that is used to study 2D material plasmonic structures. This greatly enhanced the productivity of the project and **aided in the publication of the paper "2D Semiconductor Nonlinear Plasmonic Modulators," published in Nature Communications[1].** Our approach is based on integrating monolayer semiconductor transition metal dichalcogenides on top of metallic plasmonic structures, as depicted in Figure 2a. Monolayer WSe_2 was obtained through mechanical exfoliation from bulk crystals. In order to electrically isolate the WSe_2 from the metallic waveguide (to avoid quenching of excitons), it was encapsulated with 5-10 nm thick 2D material insulator (hexagonal boron nitride, hBN). The hBN-WSe_2-hBN heterostructure was transferred onto the prefabricated metallic waveguide structure using a polymer based dry transfer technique (polycarbonate film on PDMS stamp). The alignment was carried out under a microscope allowing for sub-micron alignment of the 2D heterostructure to the center of the waveguide.

The hybrid hBN-WSe$_2$-hBN/plasmonic structure was investigated optically at 4 K in the Montana cryostat. A narrow bandwidth tunable laser was focused to a diffraction limited beam stop on the input grating. The light scattered out of the output coupler was isolated using spatial filtering and detected with an amplified silicon photodiode. The transmission spectrum is shown in Figure 2b. The black data show the transmission spectrum the hybrid hBN-WSe$_2$-hBN/plasmonic structure, and the red show a reference bare waveguide. At the exciton resonance (1.74 eV), the transmission is reduced by approximately 73% due to the presence of the WSe$_2$ layer, indicating very strong coupling between SPPs and WSe$_2$ excitons. By comparing these data to the photoluminescence spectrum, we can identify the dip in the SPP transmission as originating from the WSe$_2$ neutral exciton (X^0).

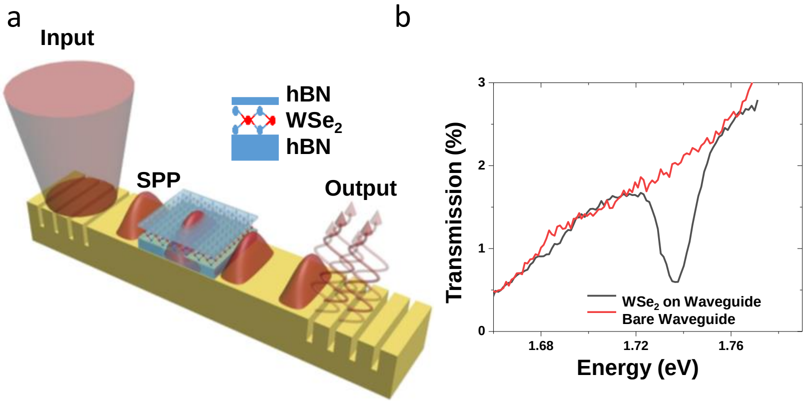

Figure 2| Plasmonic modulator device and linear response. **a** Depiction of the 2D material plasmonic device. SPPs are launched at the input of the device by focusing a free space laser onto an input coupler. The SPPs propagate through the waveguide where they can interact with excitons in the active WSe$_2$ layer, encapsulated in hBN. The SPPs are coupled back to free space photons by an output grating coupler. **b** Transmission data of the hBN-WSe$_2$-hBN plasmonic device (black). The normalized transmission of the bare waveguide is shown (red).

In order to demonstrate SPP controlled SPP propagation, we coupled pump and probe lasers into the waveguide structure (depicted in Figure 3a) and perform frequency dependent pump probe measurements (differential transmission- DT). The DT/T spectra are shown in Figure 3b for three different SPP pump energies. Again, we obtain the strongest DT response when both pump and probe are resonant with the neutral exciton near 1.74 eV. Remarkably the magnitude of the DT/T response is approximately 4%, enhanced by an order of magnitude over the optical pumping case. Figure 3c shows the a linear power dependence, consistent with a third order nonlinear response. In collaboration with Rolf Binder, we were able to determine that $\mathrm{Im}\,\chi^{(3)}$ to be $-10^{-20}\,\frac{\mathrm{m}^3}{\mathrm{V}^2}$, which

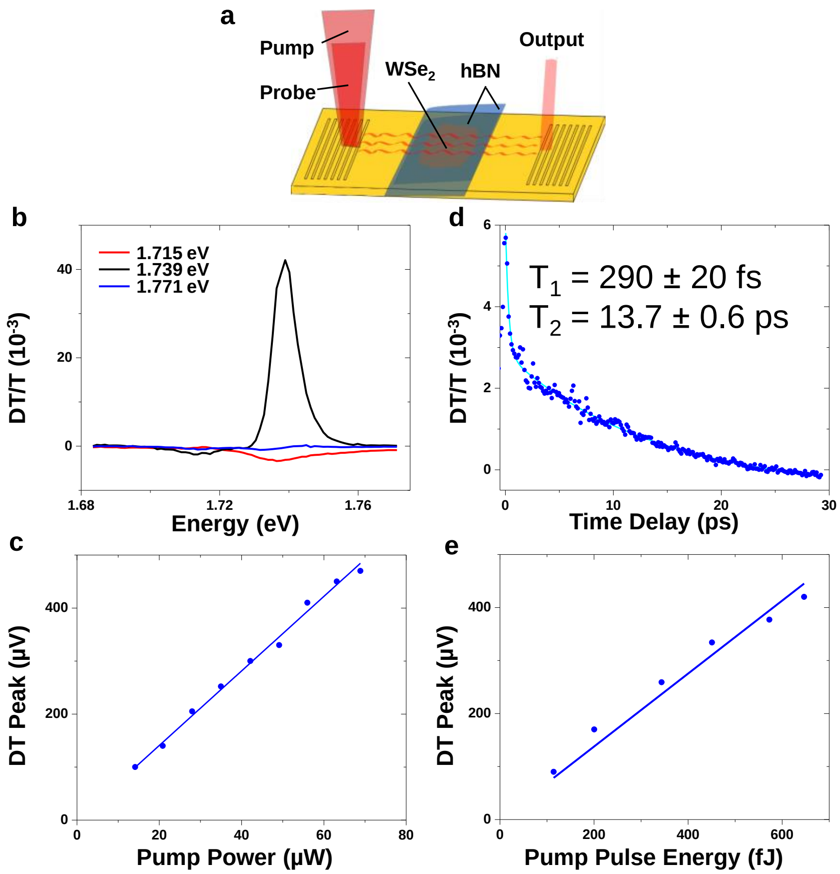

Figure 3| SPP control of SPP propagation**.** **a** Depiction of the SPP-SPP pump-probe measurements. Pump and probe SPPs are launched at the input grating coupler. SPPs propagating through the device interact with excitons in the WSe$_2$ layer. In this plasmonic modulator configuration, the pump SPP saturates the exciton absorption, resulting in an increase in probe SPP transmission. **b** SPP pump energy dependence of the DT/T spectrum. **c** SPP pump power dependence of the DT signal near the peak of the exciton response (pump 1.739 eV, probe 1.741 eV). **d** Time resolved DT/T response for a similar device measured at 11 K with a photon energy of 1.736 eV. The blue points are the data and the cyan curve is a biexponential fit whose time constants are shown. **e** SPP pump pulse energy dependence of the DT response measured near zero time delay.

is consistent with previously reported values of the third-order nonlinear susceptibility for WSe_2. Figure 3d shows time resolved pump probe response. A biexponential fit to the data show a fast ~290 fs and a slower 13.7 ps component to the decay. The fast component of the decay is on par with the fastest reported plasmonic modulators. Figure 3e shows the pump pulse energy dependence of the DT response, with relatively low switching energies of ~ 500 fJ per pulse.

We are currently using the system to investigate plasmonic amplification via "cross" waveguide structures that allow for us to achieve the phase conjugate four-wave-mixing geometry. Our preliminary data show that we are indeed able to observe a cross propagating differential transmission signal, but we are currently working on improving our structure to achieve parametric amplification.

3.2 Towards Single Photon Transistors with 1D and 2D Materials

The objective of this pilot project is to understand and demonstrate plasmon propagation in metallic single walled carbon nanotubes (SWCNTs) and couple them to localized excitons in 2D materials. In order to achieve this goal, we have developed the capability to grown SWCNTs on hBN substrates (a requirement for Luttinger liquid plasmons) and have begun developing novel quantum emitters whose locations can be deterministically placed. The long term goal of this project is to realize a single photon transistor architecture where nanoscale plasmons hosted to SWCNTs are strongly coupled to quantum emitters in 2D materials. Below we explain the results and current challenges.

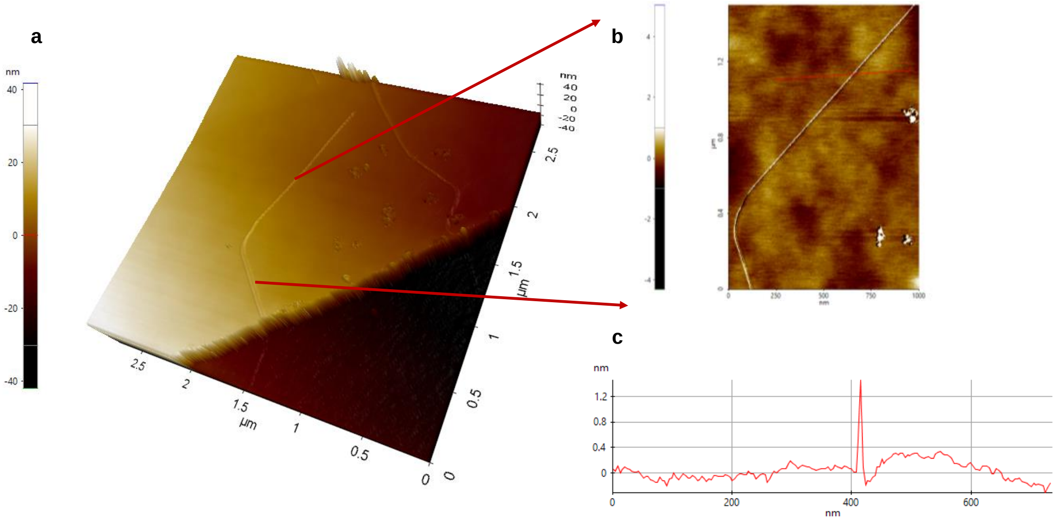

Figure 4| SWCNT on hBN| **a** AFM height image of a SWCNT grown on hBN via CVD. **b** Higher resolution AFM of **a** The red line shows the location of the line cut which is plotted in **c**.

3.2.1 CVD SWCNTs on hBN and propagating surface plasmons

We have developed a reliable fabrication process to grow SWCNTs on hBN. hBN was exfoliated on pre-cleaned SiO_2(90nm)/Si substrates. Once hBN flakes with thickness 10-50 nm were identified with an optical microscope, the substrates were spin-coated with two layers of poly (methyl methacrylate) PMMA and baked at 160 ^{0}C. Using e-beam lithography, small rectangular patterns were written on the edges of the flakes to allow targeted dispersion of carbon nanotube precursors. The catalyst was prepared from a solution of 10mg of MoO_2, 30mg of Al_2O_3 and 40 mg $Fe(NO_3)_3$ in 30 ml of methanol. Precursor was dispersed from methanol solution on to the patterned area of hBN/SiO_2. After dispersion of the precursors, the substrates were baked at 170 ^{0}C for 8 minutes to remove remaining solvent.

The PMMA was washed off by soaking it in acetone for 40 minutes and 5-10 min in methanol respectively. Consequently, the substrates were transferred to a CVD growth furnace. The CNT growth was started after the samples were heated to 900 ^{0}C while Ar gas was flowing at a rate of 990 ml/m. When the temperature reaches 900 ^{0}C, H_2 at a flow rate of (710 ml/m) and CH_4(900 ml/m) were introduced. The total growth time for the CNT was 10 minutes. After growth was complete the samples were cooled at a rate of 5 degrees/min while Ar gas is flowing.

AFM of SWCNTs grown on hBN substrates in shown in Figure 4. Since only 1/3 of SWCNTs are metallic, we then identify the metallic tubes using electric force microscopy (EFM). In 2018, LeRoy and Schaibley were awarded a ~$1.1M NSF MRI to purchase a cryogenic scanning near field optical microscope (cryo-SNOM) which will be installed in spring 2020. In a collaboration with the company (Neaspec), we have performed room temperature SNOM measurements on our CVD grown SWCNTs and observe evidence of propagating Luttinger liquid plasmons. We have found that some of the SWCNTs that appear metallic under EFM, do not show plasmon propagation in the SNOM measurements. Therefore, in future SWCNT devices, we plan to first characterize the plasmon propagation using SNOM before we fabricate the proposed far-field plasmonic couplers. In addition, we have identified several fundamental questions about plasmon propagation in SWCNTs that have not previously been explored including: what is the wavelength and temperature dependence propagation length of Luttinger liquid[2] plasmons? We plan to further investigate these propagating plasmons as function of temperature and wavelength in our cryogenic SNOM, then fabricate plasmonic couplers around allowing for far field measurements in the Montana cryostat.

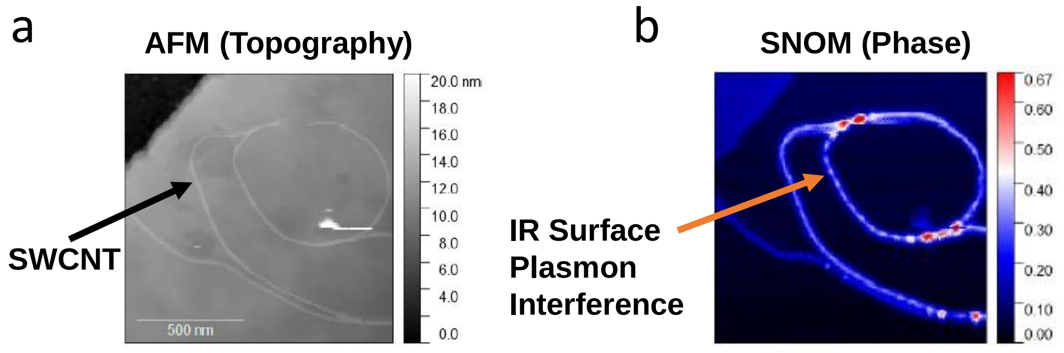

Figure 5| a AFM topography of metallic SWCNT grown on CVD. b SNOM measurement of IR plasmons propagating in SWCNT.

3.2.2 Integrating with single quantum emitters in 2D materials

Our long term goal is to interface SWCNT plasmons with single quantum emitters in 2D materials to realize a single photon transistor architecture[3]. In order to realize this goal, we have been investigating novel single emitters in 2D materials that offer the potential to realize deterministic placement, which will enable efficient coupling to SWCNT and other plasmonic waveguides. One of the most promising 2D material quantum emitters is hosted in WSe_2-$MoSe_2$ heterostructures as depicted in Figure 6a. Here, the quantum emitter is an interlayer exciton, which is shared between the layers. The moiré lattice between the two lattices has been reported to give rise to a confinement potential that traps single interlayer excitons, resulting in narrow lines that are reminiscent of other single quantum emitters[4]. We have also observed evidence of these spectrally narrow emission lines (Figure 6b), and are actively working on interfacing these emitters with lithographically defined structures to realize deterministically placement. One of the key requirements of our lithographic approach is the ability to control the emitter's energy with applied gate voltage. To study this, we fabricated the WSe_2-$MoSe_2$ heterostructures (Figure 6a) and performed gate dependent photoluminescence (Figure 6c). We see that at gate voltages near ~1.25 V some of the narrow emission lines turn off, and others turn on, which is consistent with previous reports of charge tuning InAs quantum dots. We are currently fabricating more structures that will allow for the systematic investigation of this effect. If these emitters can be charged reliably, they will potentially allow for novel spin-valley qubits that have been predicted to host a variety of novel properties[5] that are advantageous for quantum technologies.

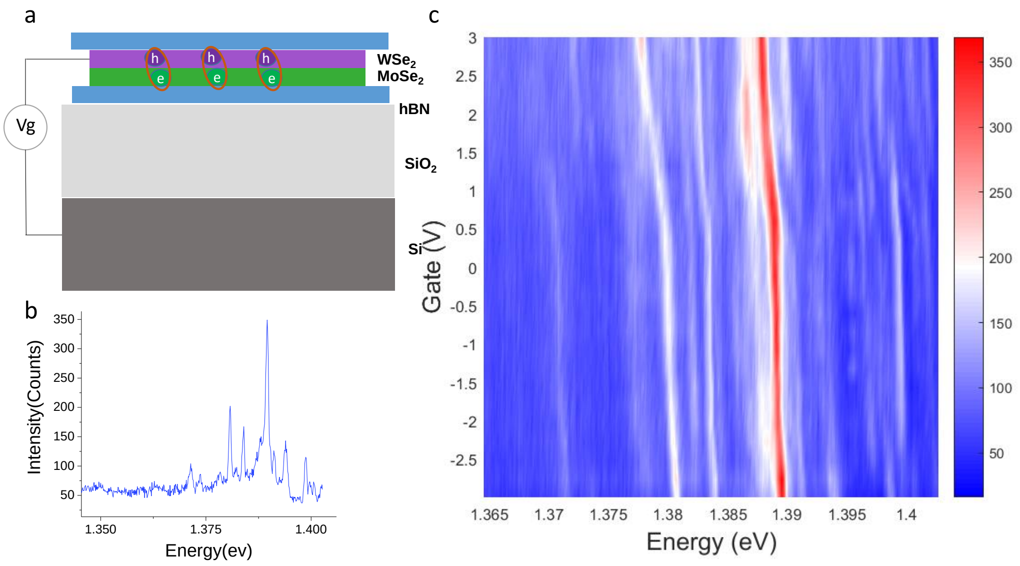

Figure 6| **a** Depiction of a $MoSe_2$-WSe_2 field effect heterostructure hosting interlayer excitons. **b** Narrow photoluminescence lines around the interlayer exciton resonance, attributed to localized single quantum emitters. **c** Gate dependent quantum light emission showing evidence of charged single quantum emitters.

4. Conclusion:

The acquisition of the closed-cycle optical cryostat has greatly enhanced both of our current AFOSR funded research projects. In particular, this acquisition has allowed for the construction of a dedicated optical setup that is optimized for nonlinear plasmonics measurements. We have also used the setup to perform nonlinear spectroscopy on hBN-encapsulated $MoSe_2$ to understand the fundamental exciton-trion interactions, have observed evidence of plasmonic enhancement of dark excitons in monolayer WSe_2, and investigated single quantum emitters in 2D heterostructures that will be interfaced with SWCNT plasmons. Beyond these current projects, we expect that the cryostat will be in constant use for decades, enabling various DoD, NSF and DOE funded projects focusing on novel light-matter interactions that have applications to information processing, and nonlinear optics.

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