DoE Final Technical Report

1. **DoE Career Award #:** DE-SC0008145

   Recipient: *University of Washington*

2. **Project Title:** Photon-Electron Interactions in Dirac Quantum Materials

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3. **Date of the report:** Nov. 10, 2017
   Period covered by the report: 07/01/2012 – 06/30/2017

4. **Research Accomplishments**

Breakthroughs in both science and technology often accompany the discovery of new material systems with unique physical properties. The goal of our project is to develop, investigate, formulate and understand a new class of Dirac quantum material, which is layered group VI transition metal dichalcogenides (MX_2), ranging from MoS_2 to WSe_2. Chemically stable and weakly bound to each other, monolayers of MX_2 have strongly bound hexagonal layers of X-M-X with trigonal prismatic coordination resulting in a honeycomb structure like graphene (Fig. 1a). This fact gives them analogous Dirac-like electronic valleys at the corners (K-points) of the hexagonal Brillouin zone. A key difference from graphene is that MX_2 has inversion asymmetry, which gives rise to direct band gaps in the visible regime and non-trivial Berry-phase related physics. But more profoundly, $\pm K$ valleys have circularly polarized optical selection rules providing the first solid state system for dynamic control of valley degrees freedom (Fig. 1b). A second important difference is the large spin-orbit coupling (150 ~ 450 meV) which arises due to the transition metal. Combined with the inversion-symmetry breaking, this truly two-dimensional semiconductor affords new possibilities for manipulating charge, layer (dipole), spin, and valley degrees of freedom to explore exotic physical phenomena (Fig. 1c).

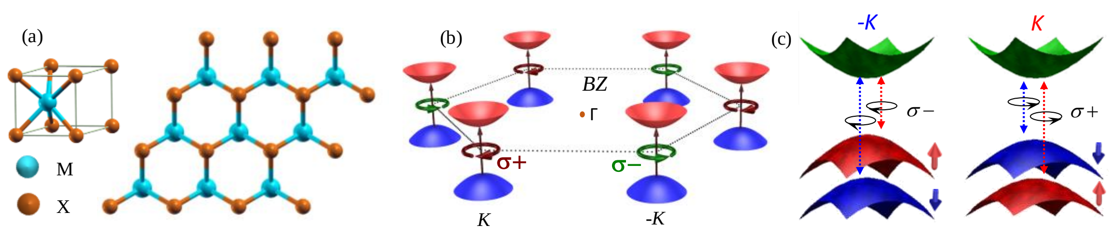

Figure 1| Physical properties of monolayer dichalcogenides. (**a**), Left: unit cell. Right: top view of the hexagonal lattice structure of monolayer MoX_2. (**b**), Schematic of inequivalent valleys at the band edges located at the K points with circularly polarized optical selection rules. (**c**), Energy level diagram of coupled spin-valley degrees of freedom with optical selection rules.

Supported by DoE career award, the PI established his group as the leader in the investigation of this new class of 2D materials and heterostructures by developing a suite of advanced nano-optical spectroscopy tools. We have made a number of breakthroughs which resulted in a total of *41* publications, including *Nature* (1), *Science* (1), *Science Advances* (2), *Nature Physics* (7), *Nature Nanotechnology* (3), *Nature Materials* (1), *Nature Communications* (7), *Nature Review Materials* (1), *Physical Review Letters* (4), *Nano Letters* (2), and *Physical Review B* (2). Below are the brief summaries of the major results.

a. *Electrical Control of Neutral and Charged Excitons in a Monolayer Semiconductor*, J.S. Ross, et al. ***Nature Communication***, 4, 1474 (2013) | doi: 10.1038/ncomms2498.

In this work, we demonstrated the full tunability of positively charged (X^+), neutral (X^o), and negatively charged (X^-) excitons in a 2D semiconductor, monolayer $MoSe_2$, for the first time (Fig. 2a). The charging energy for X^+ and X^- are nearly identical, implying the same effective mass for electrons and holes, consistent with their description as massive Dirac Fermions at the band edges. Due to reduced screening effects in the monolayer limit, the charging energy of about 30 meV is an order of magnitude larger than that of 3D bulk semiconductors. The linewidth of these excitons can be as narrow as 2 meV at a temperature of 30K. These narrow, well-separated resonances provide remarkable opportunities to selectively probe and control individual excitons in 2D crystals.

b. *Electrical Tuning of Valley Magnetic Moment Through Symmetry Control in Bilayer MoS_2*, S. Wu, et al. ***Nature Physics*** 9, 149–153 (2013) | doi:10.1038/nphys2524.

Monolayer MoS_2 has broken inversion symmetry, which leads to valley-dependent physical properties, such as magnetic moment (***m***), Berry curvature (**Ω**), and circularly polarized optical selection rules. Unlike monolayer, bilayer MoS_2 is inversion symmetric and thus valley-dependent physical properties will vanish. However, the intrinsic inversion symmetry can be broken simply by applying a perpendicular electric field. In principle, this offers the possibility of switching on/off and continuously tuning valley-dependent properties near the Dirac valleys by reversible

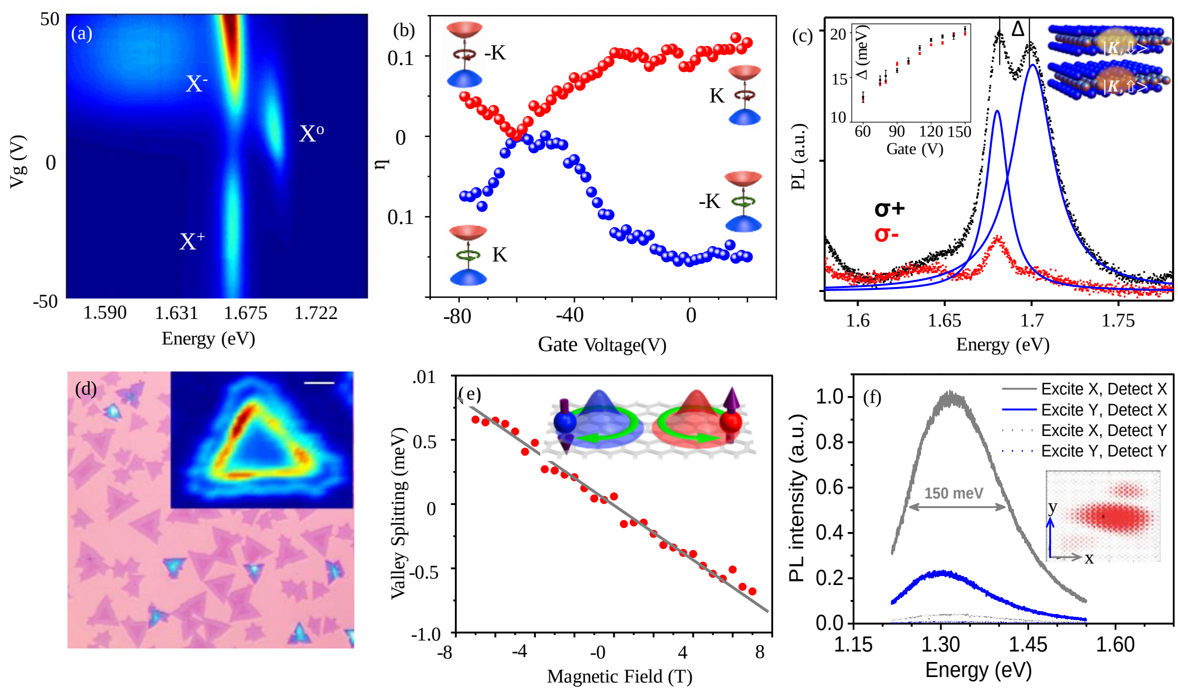

Figure 2. Selected results | (**a**) Photoluminescence (PL) intensity map of monolayer WSe_2 as a function of gate voltage and emission energy, showing charge tunable excitons. (**b**) Degree of circular polarization (η) of PL from bilayer MoS_2 as a function of perpendicular gate voltage. (**c**) Polarization resolved PL under σ^+ excitation with σ^+ (black) and σ^- detection (red). The doublet is due to the electric field induced spin Zeeman splitting. Inset: Peak splitting as a function of gate voltage (left) and schematic of spin and layer pseudospin coupling (right). (**d**) Optical image of lateral $MoSe_2/WSe_2$ heterostructure. Inset: PL intensity map of a triangular sample. (**e**) Valley Zeeman splitting as a function of magnetic field. Inset: cartoon depicting the valley magnetic moments. (**f**) Polarization-resolved PL spectra, revealing the excitonic nature of the emission from monolayer black phosphorus. Inset: real space exciton wave function by DFT calculation.

electrical control. We investigated this possibility using polarization-resolved photoluminescence (PL) of bilayer MoS_2. We find that in bilayer MoS_2 the circularly polarized PL can be continuously tuned from -15% to 15% as a function of gate voltage (Fig. 2b), whereas in structurally non-centrosymmetric monolayer MoS_2, the PL polarization is gate-independent. The observations are well explained as resulting from the continuous variation of orbital magnetic moments between positive and negative values via symmetry control.

c. *Spin-Layer Locking Effects in Optical Orientation of Exciton Spin in Bilayer WSe_2,* A. M. Jones, *et al.* ***Nature Physics,*** 10, 130 -134 (2014) | doi:10.1038/nphys2848.
A central theme in condensed matter physics is to study and understand the consequences of the interplay between distinct quantum degrees of freedom of electrons. One seminal example is the coupling between the electronic spin and motional degrees of freedom (spin-orbit interaction). In bilayer WSe_2, we discovered a new coupling effect between spin, valley, and layer pseudospins. Here, both spin and valley degrees of freedom are associated with magnetic moments. The layer degree of freedom is associated with electrical polarization, which corresponds to electrons either in the top or bottom layers (Fig. 2c, inset). We demonstrated the strong coupling effects between spin and layer pseudospin, resulting in the electrical control of spin Zeeman splitting without applied magnetic fields (Fig. 2c). Further, we provided spectroscopic evidence of interlayer and intralayer trion states, where intralayer (interlayer) means the exciton binds an additional electron or hole from the same (different) layer. We also demonstrated the optical generation of valley coherence in the interlayer trions.

d. *Lateral heterojunctions within monolayer $MoSe_2$-WSe_2 semiconductors*, C. Huang *et al*. ***Nature Materials*** 13, 1096 (2014) | doi:10.1038/nmat4064.
We demonstrated for the first time epitaxial growth of seamless lateral heterostructures in an atomic plane by physical vapor transport method. Figure 2d is an optical micrograph of an as-grown sample. Remarkably, each large crystal here exhibits two concentric regions with different optical contrast. We identified that the inside and outside triangles are $MoSe_2$ and WSe_2, respectively. Using high-resolution transmission electron microscopy, we confirmed that all the atoms lie in a single MX_2 honeycomb lattice. There are no dislocations or grain boundaries with nearly perfect lateral epitaxy. The importance of heterojunctions stems from the substantial difference between the electronic and optical properties of the two joined semiconductors. For instance, the inset in Fig. 2d is a spectrally integrated PL intensity map for a lateral heterostructure. Interestingly, the emission from the heterojunction is brighter than the bulk, highlighting the 1D heterojunction, possibly due to local potential traps at the interface.

e. *Magnetic Control of Valley Pseudospin in Monolayer WSe_2*, G. Aivazian, et al., ***Nature Physics*** 11, 148 (2015) | doi:10.1038/nphys3201.
In monolayer TMDs, electrons in the two valleys can have finite orbital contributions to their magnetic moments which are equal in magnitude but opposite in sign by time-reversal symmetry. The orbital magnetic moment has two parts: a contribution from the parent atomic orbitals, and a "valley magnetic moment" contribution from the lattice structure (Inset, Fig. 2e). In this project, we investigated the magnetic response of valley degrees of freedom. We observed the valley Zeeman splitting (Fig. 2e) and magnetic tuning of polarization and coherence of the excitonic valley pseudospin, by performing polarization-resolved magneto-PL on monolayer WSe_2. We also realized magnetic control of valley polarization and valley coherence via this magnetic moment, analogous to what is possible with real spin.

f. *Highly Anisotropic and Robust Excitons in Monolayer Black Phosphorus*, X. Wang *et al.* ***Nature Nanotechnology*** 10, 517–521 (2015) | doi:10.1038/nnano.2015.71.

Recently, black phosphorus emerged as a promising 2D semiconductor due to its widely tunable and direct bandgap, high carrier mobility, and remarkable in-plane anisotropic electrical, optical and phonon properties. In collaboration with Prof. Fengnian Xia's group at Yale University, we revealed highly anisotropic and strongly bound excitons in monolayer black phosphorus (Fig. 2f), using polarization-resolved PL measurements at room temperature. We show that regardless of the excitation laser polarization, the emitted light from the monolayer is linearly polarized along the light effective mass direction and centers around 1.3 eV, a clear signature of emission from highly anisotropic bright excitons. In addition, PL excitation spectroscopy suggests a quasiparticle bandgap of 2.2 eV, from which we estimate an exciton binding energy of around 0.9 eV, consistent with theoretical results based on first-principles.

g. Electrical Tunable Excitonic Light emission from monolayer p-n junctions, J. Ross et al., ***Nature Nanotechnology*** 9, 268 (2014).

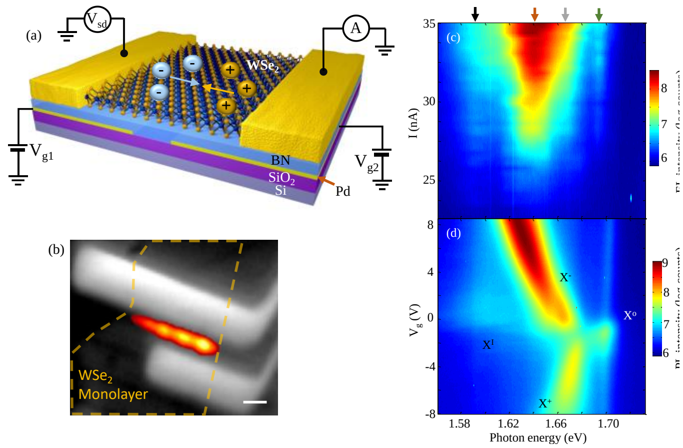

Figure 3 | Monolayer light emitting diode. (**a**) Schematic of monolayer WSe_2 p-n junctions. (**b**) Electroluminescence image (red) superimposed on a pn junction image (grayscale). The orange dashed lines outline the WSe_2 monolayer. Scale bar: 2 µm. (**c**) Electroluminescence intensity plot as function of bias current and photon energy. (**d**) Plot of photoluminescence intensity as a function of photon energy and gate voltage.

How to efficiently covert electronic energy into light emission is one of the DoE grand challenges. We address this topic by investigating the electroluminescence at single atomic layer limit. We design a monolayer WSe_2 p-n junction and observe bright electroluminescence from the p-n junction area (Figs. 3a-b). We identify that the light emission is from the radiative combination of excitons, which are created by strong Coulomb interaction between electrically injected electrons and holes. By tuning the amplitude of the electrically current, we observe electroluminescence from impurity bound exciton, negatively charged exciton, positively charged exciton, and neutral exciton (Fig. 3c). The tuning of light emission by current demonstrates a sequential population of excitonic levels from low to high energy states. We further compare the electroluminescence intensity plot as a function of current with the photoluminescence intensity plot as a function of gate voltage and photon energy (Figs. 3c-d). We find that these excitons involved in the electroluminescence are valley excitons, i.e. excitons localize at the corner of hexagonal Brillouin zone, which will be important for further investigation of polarized light emission using unique spin-valley coupled physics in MX_2.

h. Observation of long-lived interlayer excitons in monolayer $MoSe_2$-WSe_2 heterostructures, P. Rivera et al., ***Nature Communications*** 6, 7472 (2015).

The discovery of two-dimensional quantum materials provide unprecedented opportunities to explore new physical phenomena by precisely assemble 2D van der Waals heterostructures. We investigate such possibilities using monolayer $MoSe_2$-WSe_2 heterostructures, which is expected to have a type II band alignment of the heterostructure (Fig. 4a). Using photoluminescence spectroscopy, we found that the heterostructure not only maintains the intralayer excitons from individual monolayers, but also host interlayer excitons, i.e. bound electron and hole localized in different layers (Fig. 4b). We find that the energy and luminescence intensity of interlayer excitons are highly tunable by an applied vertical gate voltage. Moreover, the power dependent measurements shows the blue shift of the interlayer excitons, implying the repulsive dipole-dipole interactions, which implies 2D heterostructure is a promising system for study exciton many-body interaction effect.

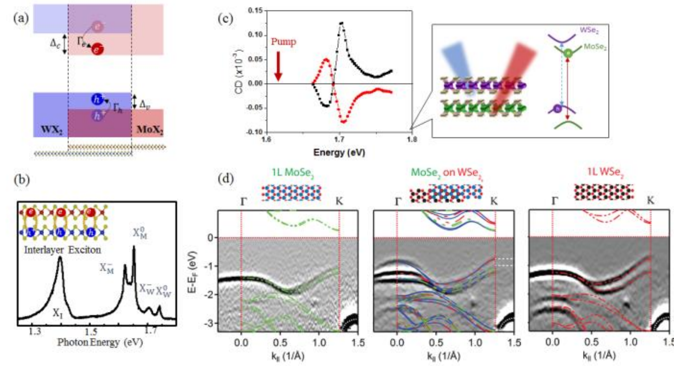

Figure 4 | Interlayer Exciton in $MoSe_2/WSe_2$ Heterostructures. (**a**) Schematic of type-II band. (**b**) PL spectra of a heterostructure. (**c**) Non-degenerated circular dichroism spectroscopy reveals directional charge transfer with spin/valley conservation. (**d**) μ-ARPES spectra of monolayer $MoSe_2$ (left), $MoSe_2/WSe_2$ heterostructure (middle), and monolayer WSe_2 (right).

i. Directional interlayer spin-valley transfer in 2D heterostructures, J. R. Schaibley et al, ***Nature Communications,*** Article 13747 (2016) | doi:10.1038/ncomms13747.

The advancement of precisely assembling isolated atomic monolayers has opened up an exciting new field based on quantum engineering of van der Waals heterostructures, which promises as a new material platform for potential breakthrough in both science and technology. However, most currently studies focus on interface effects on charge degrees of freedom, i.e. the effect of the interaction between layers on the electronic structure and thus charge transport phenomena (both within and between layers). Based on our progress in creation of 2D semiconductor heterostructures, we performed the first study of spin and valley pseudspin transport *between* monolayers in 2D $MoSe_2/WSe_2$ heterostructures (Fig. 4c). Monolayer materials such as WSe_2 are famous for unique spin-valley coupling properties, which enables the first solid state system for optical generation of spin-valley polarization in individual monolayers. Using nondegenerate optical circular dichroism spectroscopy, we report the direct observation that optically generated spin-valley polarization in a monolayer can be transferred between layers. We show that charge transfer between two monolayers conserves spins, which is robust against twist angle between layers from near zero to near 60 degree. We demonstrate directional pumping of spin polarized carriers into individual layers, i.e. polarized hole spins into WSe_2 and electron spins into $MoSe_2$. Spin initialization is a crucial operation for spintronic devices which require a net spin polarization for reading, writing, and transferring information. Our work not only provides a fundamental understanding of spin transfer across the 2D interface, but also points to a new spin pumping scheme in nanoscale devices by using 2D semiconductors as a spin-valley generator for storing and processing information.

j. Band parameters and hybridization in 2D semiconductor heterostructures from photoemission spectroscopy, Neil R. Wilson, et al., ***Science Advances*** Vol. 3, no. 2, e1601832 (2017);

Although optical spectroscopy has revealed emerging physical phenomena in 2D $WSe_2/MoSe_2$ heterostructures, the restrictions of optical characterization leave many key questions open. For example, is a semiconductor heterobilayer still a direct-bandgap system with band edges at the K points? To what extent do the orbitals hybridize at the K and Γ points, and can one regard the bands at K simply as being those from isolated monolayers? What is the band offset? Is the interlayer exciton as strongly bound as excitons in the isolated monolayers are known to be? These questions illustrate the pressing need for direct and accurate measurements of the band parameters in order to establish authoritative basis of understanding of 2D heterostructures. We employed angle-resolved photoemission spectroscopy with submicron spatial resolution (μ-ARPES) to study MoSe2/WSe2 van der Waals heterostructures (Fig. 4d). We find that in a MoSe2/WSe2 heterobilayer the bands in the K valleys are weakly hybridized, with the conduction and valence band edges originating in the MoSe2 and WSe2 respectively. There is stronger hybridization at the Γ point, but the valence band edge remains at the K points. This is consistent with the recent observation of interlayer excitons where the electron and hole are valley polarized but in opposite layers. We determine the valence band offset to be 300 meV, which combined with photoluminescence measurements implies that the binding energy of interlayer excitons is at least 200 meV, comparable with that of intralayer excitons.

k. Valley-Polarized Exciton Dynamics in a 2D Semiconductor heterostructure, P. Rivera et al, ***Science*** 351, 688 (2016)).

The observation of interlayer exciton enables us to investigate its associated valley degrees of freedom. We performed polarization-resolved PL of interlayer exciton. We apply circularly polarized continuous wave laser excitation and separately detect the right circular (σ^+) and left circular (σ^-) PL. Figure 5a shows the σ^+ (black) and σ^- (red) components of the X_I PL under circularly polarized excitation. We find that the degree of X_I valley polarization can be electrically controlled by the gate, as shown in Fig. 5b. Time-resolved measurements reveal the dynamics of the X_I PL polarization. In Fig. 5c, we show the decay of co-polarized (black) and cross-polarized

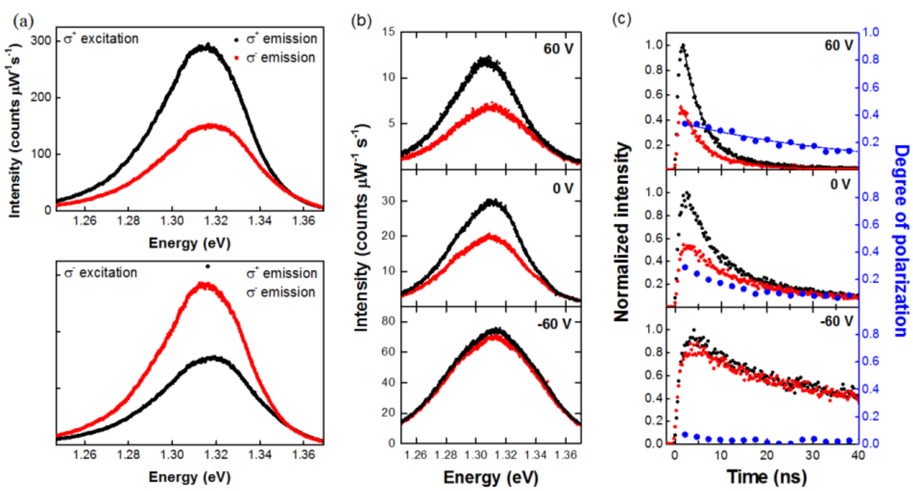

Figure 5. Interlayer exciton valley dynamics | (**a**) Circular polarization-resolved PL spectra of the interlayer exciton showing the generation of strong valley polarization. (**b**) Polarization-resolved interlayer exciton photoluminescence at selected gate voltages. (**c**), Time-resolved interlayer exciton photoluminescence at selected gate voltages. The blue curve (right axis) shows the decay of valley polarization.

(red) interlayer exciton PL, as well as the degree of polarization (blue). The valley polarization lifetime increases with V_g, reaching 39$\pm$2 ns at +60 V, as determined by fitting a single exponential decay. These measurements imply a strong suppression of intervalley scattering for interlayer excitons and a valley lifetime several orders of magnitude longer than that of intralayer excitons in monolayers, where valley depolarization occurs on picosecond timescales.

The long valley lifetime of the X_I allows visualization of their lateral drift and diffusion. The bottom panels of Fig. 6 display a sequence of spatial maps of the X_I PL polarization under pulsed excitation (40 MHz repetition rate) at $V_g = 60$ V for selected average excitation powers. The spatial pattern of ρ shows the evolution of a ring with diameter that increases with excitation intensity. The pattern of polarization stands in stark contrast to the spatial distribution of the emission. The top panels of Fig. 6 show the polarization-resolved PL intensity spatial maps, where both σ^+ and σ^- PL components display an approximately Gaussian profile centered at the excitation spot. The data shows striking difference between the spatial distribution of polarization and the total density of X_I. The observed spatial patterns in the valley polarization can be understood well as manifestations of valley-dependent many-body interactions in the dense interlayer exciton gas.

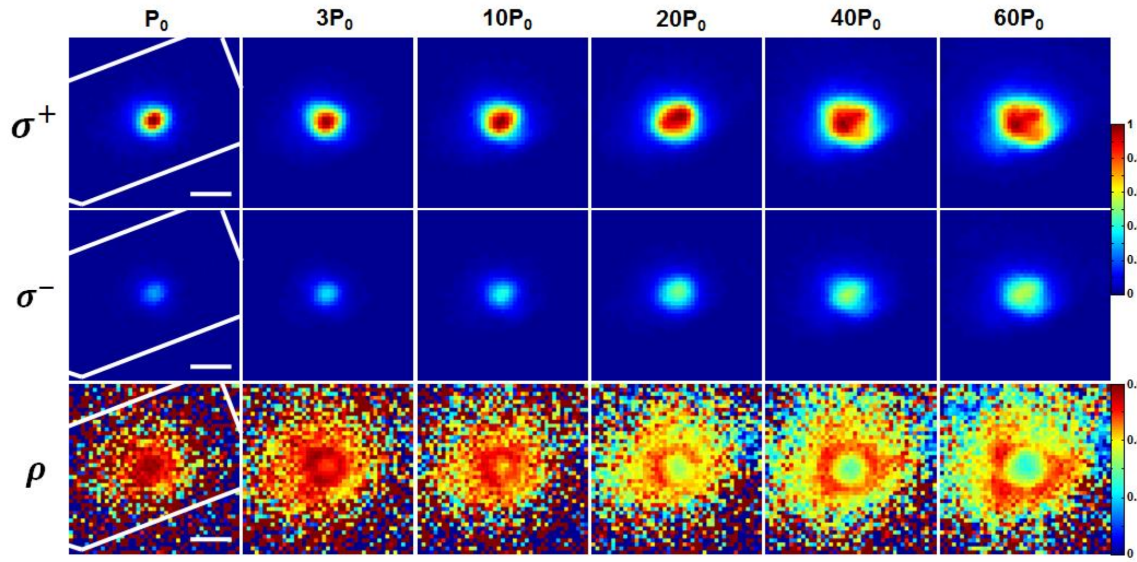

Figure 6 | Spatially resolved valley transport. Spatial maps of valley polarization under σ^+ pulsed laser excitation with $P_0 = 1$ μW. Scale bar is 2 μm. At each power, the spatial profile of co- and cross-polarized PL is shown (normalized to the peak co-polarized intensity) in the top and middle panels, respectively, and the degree of valley polarization is shown in the bottom. The spatial pattern of valley polarization displays the evolution of a ring with increasing diameter under higher excitation power.

5. Publication (All work at UW is exclusively supported by DMSE)

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6. Students involved in this project

Graduate Student: Kyle Seyler (50%), Gen Clark (0%), Aaron M Jones (50%); Jason Ross (50%); Sanfeng Wu (50%); Grant Aivazain (50%);
Postdoc: John Schaibley (100%);
Undergraduate Student: Jacob Watkins (0%); Essance Ray (0%); Mari Scott (10%)
Visiting Scholar: Chunming Huang (0%); Liefeng Feng (10%)

Aaron Jones and Grant Aivazian defended their thesis and obtained PhD degree in summer, 2015. Both of their thesis works are mainly based on this project. Sanfeng Wu and Jason Ross got their PhD degrees summer 2016. 50% of their thesis work are based on this project.

Postdoc John Schaibley is now a tenure-tracked assistant professor in Physics Department at U. Arizona.

7. Unexpended funds

The grant is fully expended.