Final Technical Report

Distribution Statement: Approved for public release; distribution is unlimited.

ONR YIP Award: N00014-16-1-3055
Multi-Dimensional Control in Laterally-Confined Atomically-Thin Nanostructures

June 1, 2016 – July 31, 2021

Recipient:
Northwestern University,
633 Clark Street, Evanston, IL 60208-1108

Principal Investigator: Nathaniel P. Stern
Report Date: 10/27/2021

Major Goals

The objective of this research was to advance the state-of-the-art methods in opto-electronic control in layered nanomaterials, including the use of lateral confinement and ultrafast manipulation. This research theme seeks to explore opportunities for creating nanoscale low-dimensional material systems that simultaneously enable multiple means of control including size, optical manipulation, and magnetic field. The knowledge pursued in this research will help enable new highly-integrable, size-tunable monolayer material platform with multiple intrinsic quantum degrees of freedom that can be deliberately engineered at the atomic size scale.

This research built on new understanding of lateral confinement of monolayer semiconductors to advance the state-of-the-art in opto-electronic control in nanomaterials. The overall objective of multi-dimensional control was addressed through several technical themes throughout the research program, outlined here.

1. Improve understanding of confinement, composition, and edges on dynamics in transition metal dichalcogenides (TMDs) and their heterostructures, thereby demonstrating deliberate control of optoelectronic effects in atomically-thin semiconductors.

2. Understand methods of lateral confinement of two-dimensional excitons in TMDs to realize monolayer quantum dots with discrete energy levels.

3. Leverage the variety of external tools available with 2D TMDs to demonstrate multi-dimensional control of excitations, thereby advancing a new platform for configurable materials with multiple engineered intrinsic quantum properties.

In pursuing these technical aims, progress was made in each theme, while the advances taken together form a foundation for future work in manipulation of layered heterostructures. The early phase of this project built on techniques previously developed in the Stern laboratory for Theme2 1 and 2, whereas the later efforts in Theme 3 expanded on ideas of coherent control of valley pseudospin in two-dimensional materials. Theme 1 was addressed both by developing nanopatterning methods and studying electrical properties and imaging methods on layered heterostructures of TMDs with hexagonal boron nitride. The second theme was approached by pushing the technical limits of top-down nanopatterning to form laterally confined nanodots. Although this did not reach the strong confinement regime, alternative approaches toward strong quantum confinement were explored by investigating electrical pumping of localized defect quantum dots in gate-defined p-n diodes. This work set the stage for new research interfacing these quantum dots with molecular systems that is currently underway. The third theme focused on ultrafast coherent control of valley pseudospin in 2D materials. Initial results showed the improvement of sensitivity in observing valley-selective energy shifts caused by ultrafast pulses. Later work extended this tool for coherent control to light-matter hybrid states, or polaritons, in TMDs embedded in optical microcavities. These last two themes have led to new collaborative research opportunities that will be explored in a newly-funded DOE-funded Energy Frontier Research Center, the Center for Molecular Quantum Transduction. The capabilities developed in this ONR YIP research was the foundation that the Stern Lab will bring to this new federally-funded center, demonstrating the impact of early career funding such as the YIP in shaping the important science being explored as careers evolve.

As an Office of Naval Research Young Investigator Program award, this research also helped develop the infrastructure, knowledge, and skills in the young Stern Laboratory, with significant investment through this YIP and associated DURIP awards in expanding the experimental capabilities for studying magneto-optical phenomena in low-dimensional materials. The impact of these investments will shape the Stern Laboratory research program long after the conclusion of this YIP award.

Project Activities and Accomplishments

This ONR Young Investigator Program award has generated accomplishments addressing each of the project objectives, with associated investigations also contributing to new scientific advances growing out of these themes. The primary accomplishments of this ONR-supported research are summarized here, including how they address the primary aims of the program.

1. Size-dependent confinement effects in nanostructures

Nanopatterning resolution-limited laterally-confined monolayer nanostructures: 2D materials are strongly confined in the vertical direction, but the remaining lateral dimensions are typically unconstrained on the nanoscale. Confined nanostructures are a standard method in nanoengineering, but observing size-induced effects is challenging in 2D materials such as TMDs because of their reduced screening and tightly bound excitons. The ability to control lateral confinement in TMD nanostructures would bring a new dimension to optimizing of layered heterostructure optoelectronics. With typical semiconductor materials, traditional semiconductor

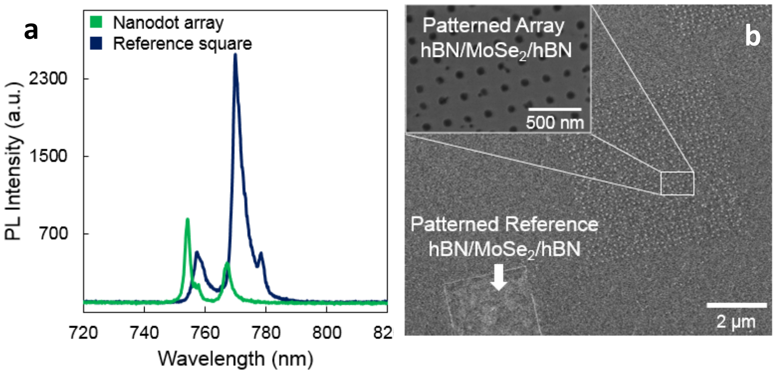

Figure 1: (a) Patterned array of hBN-encapsulated $MoSe_2$ nanodots. (b) Luminescence of patterned nanodots of $MoSe_2$ with and patterned a reference square of $MoSe_2$. Although there is a small shift, the emission of the small hBN-encapsulated nanodots is preserved. Images from Ref. 13.

processing with top-down nanolithography methods is an attractive approach for controlling lateral size, but electron beam lithography can damage few-layer optical properties in 2D materials. Positive-resist lithography avoids direct beam exposure, but the high doses required to make small structures leads to proximity effects that reduce pattern resolution. To overcome this limitation for writing lateral nanostructures, a negative resist process was developed for controllable and repeatable nanoscale patterning using direct electron beam writing that avoids damage to monolayer optical quality. By encapsulating a monolayer in hexagonal boron nitride (hBN), the monolayer is protected from damage from the electron beam up to high beam dose. This protection allows direct writing with a negative resist process for patterning small features and devices in monolayer semiconductors with lateral sizes down to 10 nm (). Although only weak confinement regimes are reached by this process, this process has implications for designing monolayer nanostructures for optoelectronics with controlled size and geometry. These results address Themes 1, 2, and 3 and are being revised for publication (Ref. 13).

Quantized conduction in top-down patterned TMD nanostructures: The ability for nanopatterning of monolayer TMDs allows the top-down design of functional optical and electronic devices with sizes scales of few tens of nanometers with near-pristine properties. To demonstrate the versatility of this method, a 50 nm narrow channel was patterned on an hBN-encapsulated MoS_2 heterostructure. This device is written with direct electron beam writing, yet the resulting patterned channel exhibits quantized conduction (**Fig. 2**). Although conductance steps have been observed

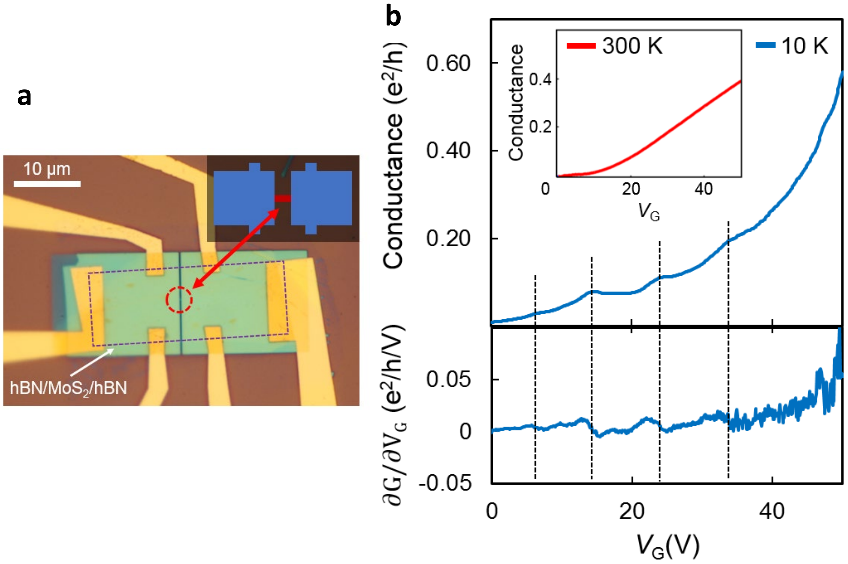

Figure 2: (a) hBN/MoS_2/hBN heterostructure with a 50-nm wide patterned nanochannel. (b) Conductance of the channel device showing step-like features of quantized conduction. Image from Ref. 13.

in MoS_2 before, they are typically induced by gate-defined channels rather than direct patterning of monolayer nanostructures. Results from this achievement addressing Themes 1 and 3 are under review (Ref. 13).

Size-dependent emission from halide perovskite nanocrystals: Distinct from the main layered heterostructure themes of this project, a collaboration with Profs. Chad Mirkin and Vinayak Dravid explored size-dependent emission from halide nanocrystal perovskites. These materials have promising optoelectronic properties, but poorly understood relationships between composition and properties limit their utility. A scanning probe method for synthesizing diverse size-controlled perovskite nanocrystal s is developed. Size-dependent photoluminescence emission from single halide perovskite nanocrystals were explored, utilizing similar approaches used in other materials in this project. This collaborative research is published in Ref. 9.

2. Advances in electrical contacts and visualization of layered heterostructures

In collaboration with Prof. Vinayak Dravid (Material Science and Engineering), advances were made in understanding heterostructure electrical contacts and electron microscope imaging of 2D materials. This new atomic-scale understanding can help provide new tools for studying opto-electronic phenomena in 2D materials and thereby help design and assemble functional layered devices (Theme 3).

Transport in tunneling contacts for monolayer lateral heterostructures: Harnessing monolayer materials and heterostructures for optoelectronic applications such as photosensitive devices requires extracting currents through electrical contacts. In lateral monolayer heterostructures, traditional metallic contacts can perturb the electronic character of the material, including novel nanoscale interfacial phenomena, so that only indirect measurements are possible. In this work, hBN tunneling contacts were employed to study optoelectronic properties of lateral heterostructures. A two order of magnitude reduction in the rectification ratio of MoS_2/WS_2 lateral heterostructures is observed when using boron nitride encapsulation and tunneling contacts. This implies that the metal/semiconductor Schottky barrier in traditional lateral heterostructure devices influences the extraction of electronic characteristics of the nontrivial lateral interface. This improved perspective on extracting monolayer properties with tunneling contacts will facilitate study of electronic properties of nanoscale layered heterostructures. This work is the first time that the hBN encapsulation and tunneling contact scheme is applied to layered heterostructures, which has been published in Ref. 3.

TEM visualization of grain boundaries in monolayers: Heterostructure stacking techniques developed in this project have been leveraged to help solve outstanding challenges in materials characterization of layered heterostructures. *In situ* observation of atomistic dynamics in TMDs has been difficult, leaving the underlying mechanisms determining macroscopic device properties unclear. In collaboration with Prof. Vinayak Dravid, a new set of *in situ* TEM and scanning TEM techniques that enable study of TMD grain boundaries under electrical bias were developed. The key contribution of the Stern Laboratory was to apply heterostructure 'stacking' techniques where layers of various materials (TMDs, hBN, InSe, etc) could be picked up and transferred to new substrates or to build heterostructures. For imaging purposes, it is necessary to transfer materials onto TEM grids. Applying past methods directly, this would not be possible as the layer would break over several hundred-micron wide holes due to pressure from the stamp. Smaller TEM holes and grids are not suitable since their copper composition would make electrical biasing impossible to apply in a controllable manner. The ONR-funded team solved the pressure problem by creating a new suspended-stamp that sits rigidly suspended by roughly 1 mm over a glass slide. Interdigitated gold contacts with spacing of <2 microns were developed to create a bridge over the TEM carve out. These contacts served as both controllable sources for biasing and structural support for layers. This approach was used to look at the impact of biasing on the formation of voids at grain boundaries in polycrystalline MoS_2 (**Fig. 3**). Mass transit under applied bias is observed in TEM images. Images reveal nanoparticulate clumps of gold or molybdenum forming near the contact/TMD boundary due to the applied bias and the voids present at grain boundaries combine to induce structural deformation. The electric field mediates a net vacancy flux from the grain boundary interior to the exposed surface edge sites that leaves molybdenum clusters. This new grain boundary visualization approach is published in Ref. 8.

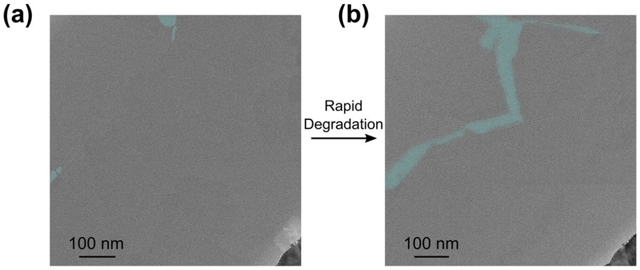

Figure 3: Image of void coalescence. (a) Low-magnification TEM of MoS_2 grain boundary region prior to biasing. (b) Same region after an electrical bias is applied. Neighboring voids (blue) coalesce to form porous chains. Image from Ref. 8.

Spatial mapping of electrostatic fields in 2D heterostructures: Building on the approaches used for TEM imaging of layered heterostructures, differential phase contrast (DPC) imaging is used to probe local electrostatic fields during electrical operation with nanoscale spatial resolution in hBN-MoS_2 heterostructures. Using a filtering algorithm, *a priori* electric field expectations can be directly compared with experimentally derived values to identify inhomogeneities and problematic regions. This platform is used to analyze the electric field and charge density distribution across layers of hBN and MoS_2. This collaborative work has been published in Ref. 12.

3. Valley-polarized photocurrents in monolayer devices

To harness valley pseudospin as a tool for control in monolayer materials, transport of valley must be better understood. Spin and valley-polarized electronic currents can be induced entirely optically through the circular photogalvanic effect (CPGE). In a monolayer TMD, this phenomenon is due to the symmetry of the bandstructure. Control of CPGE spin photocurrents has been explored only in bulk TMDs by gate electric fields, where this field is used to induce inversion asymmetry. So far no work has reported the electric control of CPGE spin photocurrent in a monolayer, in which there is intrinsic inversion asymmetry. In this unexplored regime, the gate voltage can be a tool for electrical control and optimization of optically-injected spin polarized photocurrents. Tuning of CPGE spin-valley photocurrent in monolayer MoS_2 at room temperature by both source-drain voltage and gating control was demonstrated. The polarization degree of the photocurrent can be tuned significantly (from 0.5% to 16.6%) by gate control (**Fig. 4**), showing transistor-like modulation behavior of spin polarization with the maximum “on/off ratio” of 34. Backed by gate-dependent photoluminescence experiments revealing the quenching of defect-related emission tuned by gate voltage, these results suggest a gate-controlled free carrier-screening effect on defect-associated valley scattering. Improved understanding of circular photogalvanic effects in layered materials can have applications in polarized current generation. This research addresses control of valley in heterostructures (Theme 3), and is published in Ref. 6.

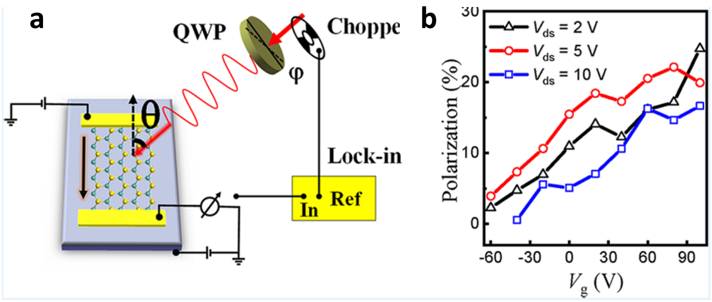

Figure 4: (a) Schematic of the experimental set up for circular photogalvanic effect in TMDs. (b) Spin photocurrent polarization degree P versus gate voltage. Nearly linear response shows capability of gate tuning the spin-polarized photocurrent to both on and off regimes. Image from Ref. 6.

4. Coherent control of valley pseudospin in 2D materials

One of the unique features of TMDs is the valley pseudospin, which can be selectively accessed using circular polarization of light. The so-called valley pseudospin labels regions of momentum space, but it can take two values, much like a traditional spin. Because valleys can be excited in coherent superpositions, there is potential to sue valley as a store of quantum information. This research program sought to develop methods of controlling the valley pseudospin (Theme 3), eventually combining these methods with the laterally confined nanostructures of Theme 1. Although this ONR-supported project did not demonstrate coherent control of the confined nanostructures, the approaches for coherent control were successfully applied to a different TMD excitation: a hybrid state of light and matter, or polariton.

Improved sensitivity for measuring Stark shifts for coherent control of valley pseudospin: Intense polarized optical pulses can break the degeneracy of valley energy levels in monolayer semiconductors. This effect can act as an effective all-optical magnetic field, enabling coherent control of valley pseudospin. This research has shown that time-resolved Kerr rotation can be used as a sensitive probe of valley-dependent level splitting induced by the optical Stark effect in monolayer transition metal dichalcogenides. Kerr rotation rejects polarization-independent backgrounds and probes a complementary dielectric response from established absorption-based techniques used to study the valley Stark effect. This allows for detection of shifts as small as 4 μeV – the lowest reported value in literature. Improving the detection sensitivity of experiments opens the door to deeper understanding, and eventually broader application, of new phenomena by enabling precise observations of smaller features. The pulse techniques are useful for coherent control of valley pseudospin, and our detection scheme can be harnessed to observe coherent valley control in smaller monolayer nanostructures where higher sensitivity is required. These results have been published in Ref. 4.

Valley-selective Stark shifts of light-matter hybrid polaritons: Valleytronics aims to use the valley degree of freedom to encode and process information. The field of polaritonics aims to harness the unique properties of half-light half-matter polaritons for new technologies. The Stern Lab bridges

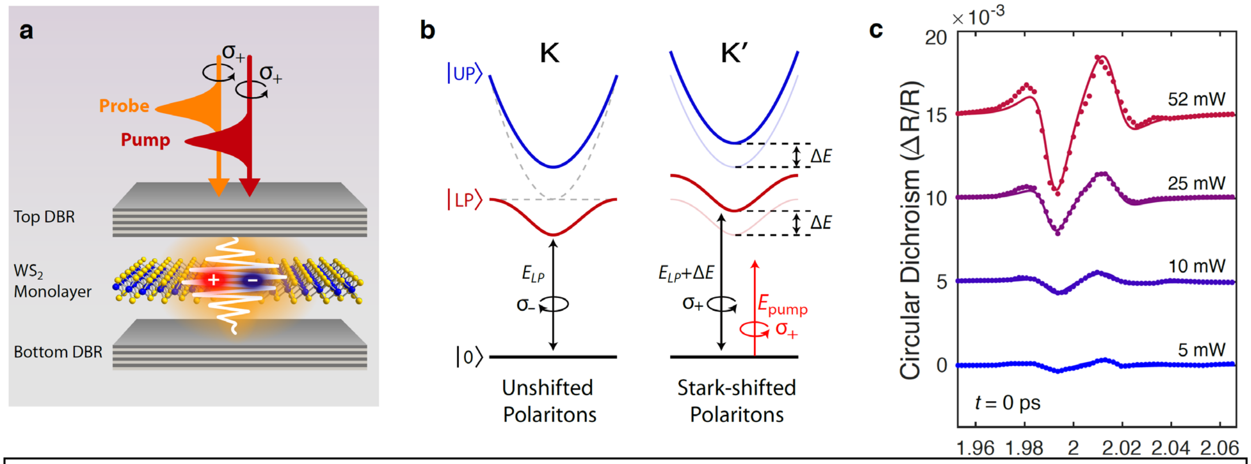

Figure 5: (a) Polarization-dependent transient reflectance of exciton-polaritons. (b) Valley-selective optical Stark shift. The σ+ polarized pump only couples to polaritons in the K' valley, causing an optical Stark shift ΔE of both the upper and lower polariton branches. (c) Pump-induced circular dichroism for increasing pump intensities. The dotted data is well fit to a Lorentz oscillator model incorporating the Stark shifted excitons and microcavity polariton spectra. Image from Ref. 11.

these two fields by investigating how the optical Stark effect can be used to manipulate the energy levels and valley degree of freedom of exciton-polaritons in monolayer TMDs. Monolayer TMDs exhibit unique valley-dependent optical properties that are also imparted to exciton-polaritons. Using polarization-dependent transient reflectance, the valley-selective optical Stark effect in WS_2 exciton-polaritons is demonstrated (**Fig. 5**). There is a simultaneous shift of both polariton branches when pump and probe are co-polarized and no appreciable shift when they are cross-polarized, demonstrating a polarization-selective stark shift in exciton-polaritons. Ultrafast pump-probe spectroscopy reveals polarization-selective spectra of the polaritons that originate from valley-selective energy shifts. These results establish a new approach for coherent control of spin phenomena at picosecond timescales in valley polaritonics. A report on these results is published in Ref. 11.

5. Electrical pumping of deterministic confined quantum emitters in dual-gated layered heterostructures

Single photon sources comprise an essential element of a number of emerging quantum optical technologies. The recent discovery of strain defects in layered TMD materials that can act as single photon emitters is a promising development since these emitters luminesce brighter than the normal excitonic species and exhibit narrow linewidths on the order of 100 micro eV. Electrical pumping of emitters in WSe_2 has been achieved in vertical LEDs, but this implementation is not ideal for scalability as it depends on the presence of crystalline defects which are largely randomly distributed over monolayers. To address this situation, lateral p-n junctions in monolayer WSe_2 defined by local electrostatic gates were fabricated (**Fig. 6**). Single photon emitters states are deterministically achieved in the interfacial region between the two gates by fabricating dielectric nanopillars to induce localized strain profiles in transferred monolayers. Since all electroluminescence is generated within this narrow region, the spectral background from the continuum of intrinsic exciton states can be minimized with tailored gate design. The presence of

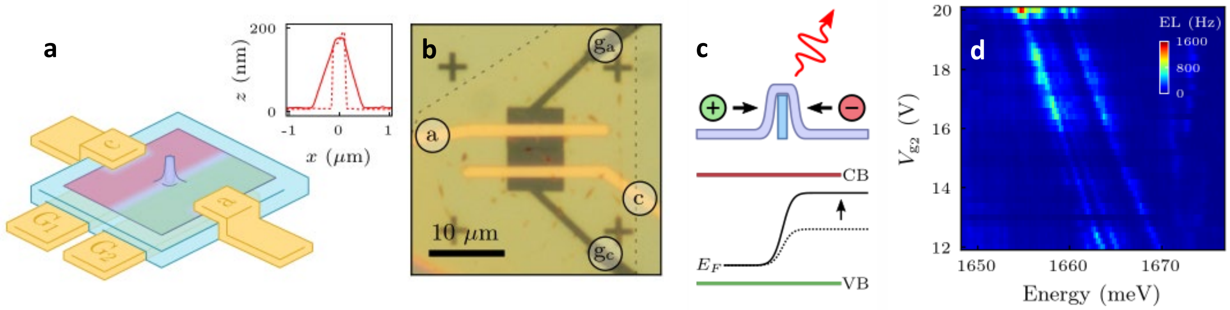

Figure 6: Tuning excitonic levels of defect single photon emitters in monolayer p-n diode. (a) Lateral gate-defined p-n diode assembled from WSe_2, with a nanopillar strain defect deterministically creating a quantum emitter. (b) Optical image of the device. (c) Recombination mechanism at quantum emitter under bias. (d) Tunable luminescence energy from defect emitters depending on the applied bias voltage, demonstrating gate tuning of quantum emitters in WS_2 p-n diodes. From Ref. 14.

the localized single emitters in the p-n junction are confirmed with $g^{(2)}$ second order correlation measurements. The lateral gate-defined p-n junction allows tuning of the emission energy via the Stark effect (**Fig. 6d**). The large in-plane fields of the p-n junctions can cause a red shift of the excitonic energies. With the dual bottom gate structure employed in our device architecture, high in-plane electric fields on the order of several tens of V per micron can be achieved. This geometry was exploited to observe energy shifts of quantum emitters caused by lateral electric fields. These shifts are observed in both photoluminescence and electroluminescence of single photon emitters in WSe2. This tuning from the local electric fields in lateral p-n diodes provides electrical control analogous to gate-defined quantum dots. These effects are observed in both monolayer and bilayer devices, suggesting compatibility with more complex heterostructures. This work, involving both ONR (quantum dots) and NSF (electrical TMD devices)-supported activities, directly addressing Themes 2 and 3 of this project with electrical control of strongly-confined emitters. The results are in preparation for publication (Ref. 14).

<u>Impact of this YIP Research Program – Training Opportunities</u>

The work of this project has contributed to the granting of three Ph.D. degrees at Northwestern University, each recipient receiving significant student support from ONR during their career. The YIP award also supported research that provided technical training opportunities to two other graduate students who were primarily funded from other sources. In addition to these graduate researchers, the project provided research opportunities for four undergraduate researchers, one of whom was a coauthor on published papers. The research also provided research opportunities for five high school interns, two of which participated in this and related research for over a year before leaving for college. Two more would have participated in the last (no-cost extension) year of the project but were prevented from doing so by the Covid-19 pandemic. These interns have primarily come from Evanston Township High School and a partnership with the advanced science instructor there. These numbers highlight the significant opportunities for participation created by this research program and the impact that funding a young and growing laboratory can have on workforce development.

Dissemination of Information

Results supported by this research have been disseminated through numerous conference presentations and journal publications. In addition to the refereed journal publications, results from this project are documented in four Ph.D. theses and one undergraduate research thesis. These theses have a particularly high impact on dissemination of information to future generations of researchers in the group since they contain significant technical details and are closely read by young students.

Meetings that included presentations based on work related to this grant include:

- March Meeting of the American Physical Society (annually, several presentations per year by ONR-supported students and postdocs)
- Invited presentations at SPIE Photonics West and the Materials Research Society meetings
- Three international presentations at the GRAPHENE conference in Europe (Spain, Germany, Italy)
- Over 10 invited academic seminars and colloquia

This research has also been used in outreach to younger students. The Stern research lab has been used for numerous lab tours for recruiting both undergraduates and graduate students, with ONR-funded research using ONR-funded infrastructure (sample preparation equipment, cryostats, lasers) a key component of these tours. The optical and cryogenic facilities funded in this research program often impress the general public, and the Physics and Astronomy department at Northwestern recognizes the value that the Stern lab provides for outreach to these populations. The Stern Lab is often a highlighted destination for these campus tours.

With outreach to younger students, this ONR project is helping dissemination of scientific skills and knowledge outside of the traditional technical and graduate student communities. Numerous lab tours to undergraduate and high school students have facilitated the exposure of research ideas and processes to help interest younger students and recruit them to science. The Stern Lab has established a joint interaction with the advanced science classes at the local Evanston Township High School in which talented high school students are brought into the laboratory to see real science in action. The research is described, and the students are encouraged to seek summer internship opportunities. Specifically, for this ONR program, two high school students were recruited for 15 month internship experiences, demonstrating the value of exposure of science for helping young people get involved at an early age. Although the pandemic hindered these experiences from continuing at the end of this project (2020 and 2021), the early years were a highlight of targeted dissemination of federally-funded science to improve interest and opportunities in non-technical students.

Impact of this YIP Research Program

Impact on Scientific Career

As a Young Investigator Program award, this project has helped build a foundation for future scientific impact. New research directions have arisen from the research in this program, especially the Theme 3 work. Capabilities from Theme 3 have been the basis for the Stern Laboratory's involvement in the new Department of Energy supported Energy Frontier Research Center, the Center for Molecular Quantum Transduction. This highly collaborative center has research thrusts that build on the valley coherence work done during this project. Additional research directions studying the manipulation of valley coherence are also being proposed which build on this work. These new scientific directions grow directly from the ONR-supported work of this award, demonstrating how the early career support has helped shape longer term directions of inquiry and collaboration.

Impact on Infrastructure

At the beginning of the period of performance for this project, the Stern Laboratory moved to a newly renovated laboratory facility Northwestern University. This new laboratory has increased space and better infrastructure for optics experiments. The new facility allowed more space for instrumentation. This ONR project helped equip this new lab space with nitrogen glove box and optical microscope for heterostructure assembly and a new source/measure unit for electrical experiments.

The research of the ONR YIP also was supported by two associated Defense University Research Instrumentation Program (DURIP) awards, N00014-18-1-2131 and N00014-18-1-2135. These awards provided additional funds for this new laboratory for a tunable continuous wave laser and two magneto-optical cryostats. This new instrumentation helped equip the new laboratory space and made several of the experiments in this project technically possible. The associated DURIP awards will have long term impact on the Stern Laboratory research beyond this YIP award by enabling sophisticated experiments. These DURIP awards also facilitated a partnership with Quantum Design to test a new OptiCool cryostat, which will have a lasting impact on the available scientific instrumentation to the community.

Honors

Nothing to report.

Technology Transfer

As part of two ONR DURIP awards associated with this ONR YIP, we acquired the new Quantum Design OptiCool magneto-optical cryostat. The system was acquired under a beta test agreement, in which research procedures, protocols, and experiences with the new system are shared with the vendor engineering team so that they can improve the system and its support for users. Although the system was acquired under separate DURIP awards, it was installed during the period of performance of this research and used by participants in this research. The insights gained from using this system as part of this project will guide information transfer from academic users in the Stern lab to the instrument manufacturers in industry.

Participants

The following personnel participated on this project. Interns, undergraduates, and summer participants are generally not directly funded by ONR but participate on a volunteer or university-supported summer opportunity basis.

Participant	Role	Person-Months worked
Stern, Nathaniel	PI	4
Dong, Yiyun	Undergraduate Student	3
Doppelt, Jonah	High school intern	2
Gottardi-Littel, Christopher	High school intern	12
Hansen, Gavin	High school intern	2
Hyman, Ross	K-12 teacher (intern, unfunded)	2
Kaup, Lydia	High school intern	3
LaMountain, Trevor	Graduate Student	18
Lenferink, Erik	Graduate Student	25
Liu, Lei	Postdoctoral fellow	12
Liu, Pufan	Graduate Student	18
Meyer, Joseph	High school intern	3
Nelson, Jovan	Graduate Fellow	3
Speiser, Nathaniel	Undergraduate Student	6
Stanev, Teodor	Graduate Student	24
Sun, Yuhan	Undergraduate Student	3
Wei, Guohua	Graduate Student	6
Yang, Madison	High school intern	12

Products

Publications

1. "Valley-selective photon-dressed states in transition metal dichalcogenides." T. LaMountain, Y. J. Chen, T. K. Stanev, and N. P. Stern. *Proc. SPIE 10530, Ultrafast Phenomena and Nanophotonics XXII,* 1053016 (2018). doi:10.1117/12.2285666

2. "Environmental Engineering of Transition Metal Dichalcogenide Optoelectronics." T. LaMountain, E. J. Lenferink, Y. J. Chen, T. K. Stanev, and N. P. Stern. *Frontiers of Physics* **13**, 138114 (2018). doi:10.1007/s11467-018-0795-x

3. "Intrinsic Transport in 2D Heterostructures Mediated through h-BN Tunneling Contacts." A. A. Murthy, T. K. Stanev, J. D. Cain, S. Hao, T. LaMountain, S. Kim, N. Speiser, K. Watanabe, T. Taniguchi, C. Wolverton, N. P. Stern, and V. P. Dravid. *Nano Letters* **18**, 2990-2998 (2018). doi:10.1021/acs.nanolett.8b00444

4. "Valley-selective optical Stark effect probed by Kerr rotation." T. LaMountain, H. Bergeron, I. Balla, T. K. Stanev, M. C. Hersam, and N. P. Stern. Physical Review B 97, 045307 (2018). doi:10.1103/PhysRevB.97.045307

5. "Manipulating valley-sensitive light-matter states in monolayer transition metal dichalcogenides." Y. J. Chen, T. LaMountain, T. K. Stanev, and N. P. Stern. *Proc. SPIE 10920, 2D Photonic Materials and Devices II,* 1092007 (2019). doi:10.1117/12.2510919

6. "Electrical Control of Circular Photogalvanic Spin-Valley Photocurrent in a Monolayer Semiconductor." L. Liu, E. J. Lenferink, G. Wei, T. K. Stanev, N. Speiser, and N. P. Stern. *ACS Applied Materials and Interfaces* **11**, 3334-3341 (2019). doi:10.1021/acsami.8b17476

7. "Valley-selective optical Stark effect of exciton-polaritons in a monolayer semiconductor." T. LaMountain, E. Lenferink, S. Amsterdam, M. Hersam, and N. P. Stern. *Proc. SPIE 11282, 2D Photonic Materials and Devices II, 112820D* (2020). doi:10.1117/12.2546644

8. "Direct Visualization of Electric-Field-Induced Structural Dynamics in Monolayer Transition Metal Dichalcogenides." A. A. Murthy, T. K. Stanev, R. dos Reis, S. Hao, C. Wolverton, N. P. Stern, and V. P. Dravid. *ACS Nano* **14**, 1569-1576 (2020). doi:10.1021/acsnano.9b06581

9. "Halide perovskite nanocrystal arrays: Multiplexed synthesis and size-dependent emission." J. S. Du, D. Shin, T. K. Stanev, C. Musumeci, Z. Xie, Z. Huang, M. Lai, L. Sun, W. Zhou, N. P. Stern, V. P. Dravid, and C. A. Mirkin. *Science Advances* **6**, eabc4959 (2020). doi:10.1126/sciadv.abc4959

10. "Tailoring the Optical Response of Pentacene Thin Films via Templated Growth on Hexagonal Boron Nitride." S. H. Amsterdam, T. LaMountain, T. K. Stanev, V. K. Sangwan, R. López-Arteaga, S. Padgaonkar, K. Watanabe, T. Taniguchi, E. A. Weiss, T.

J. Marks, M. C. Hersam, and N. P. Stern. *J. Phys. Chem. Lett.* **12**, 26-31 (2021). doi:10.1021/acs.jpclett.0c03132

11. "Valley-selective optical Stark effect of exciton-polaritons in a monolayer semiconductor." T. LaMountain, J. Nelson, E. J. Lenferink, S. H. Amsterdam, A. A. Murthy, H. Zeng, T. J. Marks, V. P. Dravid, M. C. Hersam and N. P. Stern. Nature Communications 12, 4530 (2021). doi:10.1038/s41467-021-24764-8
12. "Spatial Mapping of Electrostatic Fields in 2D Heterostructures." A. A. Murthy, S. M. Ribet, T. K. Stanev, P. Liu, K. Watanabe, T. Taniguchi, N. P. Stern, R. dos Reis, and V. P. Dravid. *Nano Letters* **21**, 7131-7137 (2021). doi:10.1021/acs.nanolett.1c01636
13. "Direct Patterning of Optoelectronic Nanostructures using Encapsulated Layered Transition Metal Dichalcogenides." T. K. Stanev, P. Liu, H. Zeng, E. J. Lenferink, A. Murthy, N. Speiser, K. Watanabe, T. Taniguchi, V. P. Dravid, and N. P. Stern. *In review* (2021).
14. "Deterministic and tunable localized exciton emission from monolayer p-n junctions." E. J. Lenferink, T. LaMountain, T. K. Stanev, E. Garvey, K. Watanabe, T. Taniguchi, and N. P. Stern. *In preparation* (2021).

Ph.D. Theses containing material supported by this ONR YIP project

1. "Optical Properties of Layered Semiconductor Nanostructures." G. Wei. Dissertation, Northwestern University. *ProQuest Dissertations Publishing*, 10288969 (2017).
2. "Broken Symmetries in Low-dimensional Materials." E. J. Lenferink. Dissertation, Northwestern University. *ProQuest Dissertations Publishing*, 22617874 (2019).
3. "Optical Stark Shifts of Hybrid Light-Matter States in Two-Dimensional Semiconductors." T. LaMountain. Dissertation, Northwestern University. *ProQuest Dissertations Publishing*, 28649661 (2021).
4. "Engineering Low-Dimensional Layered Structures." T. K. Stanev. Dissertation, Northwestern University. *ProQuest Dissertations Publishing*, 28713953 (2021).

Acknowledgement

This material is based upon work sponsored by the Department of the Navy, Office of Naval Research under Award Number N00014-16-1-3055.

REPORT DOCUMENTATION PAGE

PLEASE DO NOT RETURN YOUR FORM TO THE ABOVE ORGANIZATION.

1. REPORT DATE 20211027	**2. REPORT TYPE** Final Technical Report	**3. DATES COVERED** **START DATE** 20160601	**END DATE** 20210731

4. TITLE AND SUBTITLE
Multi-Dimensional Control in Laterally-Confined Atomically-Thin Nanostructures

5a. CONTRACT NUMBER	**5b. GRANT NUMBER** N00014-16-1-3055	**5c. PROGRAM ELEMENT NUMBER** YIP
5d. PROJECT NUMBER	**5e. TASK NUMBER**	**5f. WORK UNIT NUMBER**

6. AUTHOR(S)
Stern, Nathaniel, P.

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
Northwestern University
633 Clark St.
Evanston, IL 60208-0001

8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
Office of Naval Research
875 N. Randolph St. Suite 1425
Arlington VA 22203-1995

10. SPONSOR/MONITOR'S ACRONYM(S)
ONR

11. SPONSOR/MONITOR'S REPORT NUMBER(S)

12. DISTRIBUTION/AVAILABILITY STATEMENT
Approved for Public Release; distribution is Unlimited

13. SUPPLEMENTARY NOTES

14. ABSTRACT
This is the final technical report for ONR Young Investigator Program Award N0014-16-1-3055. The objective of this research was to advance the state-of-the-art methods in opto-electronic control in layered nanomaterials, including the use of lateral confinement and ultrafast manipulation. Activities, impact, and products of the research program are summarized.

15. SUBJECT TERMS
Nanostructures, monolayers, control, YIP

16. SECURITY CLASSIFICATION OF:			**17. LIMITATION OF ABSTRACT**	**18. NUMBER OF PAGES**
a. REPORT	**b. ABSTRACT**	**C. THIS PAGE**		13

19a. NAME OF RESPONSIBLE PERSON Nathaniel P. Stern	**19b. PHONE NUMBER** *(Include area code)* 847-467-7625

PREVIOUS EDITION IS OBSOLETE.